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Various Domination Parameters in Mycielski's graphs

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Abstract: Given a graph G and any integer $m \geq 0$, Mycielski constructed a graph $\mu(G)$ and one can transform G into a generalized mycielskian of G, $\mu_m(G)$. This paper investigate the mycielskian number of $\mu_m(G)$ under the domination parameters strong domination, weak domination, dom chromatic, chromatic strong domination and chromatic weak domination. Also we show that for a graph G without isolated vertices, $\mu(G)$ and $\mu_m(G)$ are not strong(weak) efficient open dominatable whenever G is strong(weak) efficient open dominatable.

Key words: Mycielski's Graph, Strong (Weak) Domination, Dom Chromatic set, Chromatic Strong(Weak) Domination, Strong(Weak) Efficient Open Domination.

1 Introduction

Let $G = (V, E)$ be an undirected graph with vertex set V and edge set E. For graph theoretic terminology, we refer to [4] and [5]. The *open neighborhood* of $v \in V$ is $N(v) = \{u \in V \mid uv \in E\}$ and *closed neighborhood* of $v \in V$ is $N[v] = N(v) \cup \{v\}$. The *strong open neighbourhood* of a point u is the set $N_s(u)$ consisting of all points v such that $degu \leq degv$ which are adjacent with u. The *strong neighbourhood* is $N_s[u] = N_s(u) \cup \{u\}$. The *weak open neighbourhood* of a point u is the set $N_w(u)$ consisting of all points v such that $degu \geq degv$ which are adjacent with u. The *weak neighbourhood* is $N_w[u] = N_w(u) \cup \{u\}$.

E. Sampathkumar and L. Pushpalatha introduced the concepts of strong(weak) domination in [12]. A subset S of $V(G)$ is called a *strong dominating set* of G if for every $v \in V - S$, there exists $u \in S$ such that u and v are adjacent and $degu \geq degv$. The *strong domination number* $\gamma_s(G)$ of G is the minimum cardinality of a strong dominating set. A subset S of $V(G)$ is called a *weak dominating set* of G if for every $v \in V - S$, there exists $u \in S$ such that u and v are adjacent and $degu \leq degv$. The *weak domination number* $\gamma_w(G)$ of G is the minimum cardinality of a weak dominating set.

T. N. Janakiraman and M. Poobala ranjani [8] introduced a new conditional dom chromatic set and S. Balamurugan et al [2] extended this dom chromatic set to chromatic strong (weak) dominating set. A subset D of V is said to be a *dom chromatic set* if D is a dominating set and $\chi(< D >) = \chi(G)$. The minimum cardinality of a dom chromatic set in a graph G is called the *dom chromatic number* and is denoted by $\gamma_{ch}(G)$. A dom chromatic set with cardinality γ_{ch} is called $\gamma_{ch} -$ set of G. A subset D of V is said to be a

chromatic strong dominating set if D is a strong dominating set and $\chi(< D >) = \chi(G)$. The minimum cardinality of a chromatic strong dominating set in a graph G is called the *chromatic strong domination number* and is denoted by $\gamma_s^c(G)$. A chromatic strong dominating set with cardinality γ_s^c is called $\gamma_s^c -$ set of G. A subset D of V is said to be a *chromatic weak dominating set* if D is a weak dominating set and $\chi(< D >) = \chi(G)$. The minimum cardinality of a weak strong dominating set in a graph G is called the *chromatic weak domination number* and is denoted by $\gamma_w^c(G)$. A chromatic weak dominating set with cardinality γ_w^c is called $\gamma_w^c -$ set of G.

We introduced the concept of strong(weak) efficient open domination in [1]. A subset D of $V(G)$ is called a *strong efficient open dominating set* (or SEOD set, for short) of G if $|N_s(v) \cap D| = 1$, for every $v \in V(G)$. A subset D of $V(G)$ is called a *weak efficient open dominating set* (or WEOD set, for short) of G if $|N_w(v) \cap D| = 1$, for every $v \in V(G)$. The strong (weak) efficient open domination number, denoted by $\gamma_{ste}(G)$ ($\gamma_{wte}(G)$), is the minimum cardinality of a strong (weak) efficient open dominating set of G. We also call the corresponding set that γ_{ste} (γ_{wte}) - set of G. A graph G is called a strong (weak) efficient open dominating graph or SEOD (WEOD) graph if it contains a strong(weak) efficient open dominating set. Also says that G is strong (weak) efficient open dominatable. In this paper we investigate the mycielskian number of $\mu_m(G)$ under the domination parameters strong domination, weak domination, dom chromatic, chromatic strong domination and chromatic weak domination. Also we show that $\mu(G)$ and $\mu_m(G)$ are not strong(weak) efficient open dominatable whenever G is strong(weak) efficient open dominatable, for a graph G without isolated vertices.

2 The Mycielski Construction

In 1955, Mycielski, [7] introduced a admirable construction to increase the chromatic number of triangle free graphs without increasing a clique number. W. Lin et al [13] call this mycielski's graph as mycielskian of G.

The Mycielskian of a graph G is defined as follows:

Let G be a graph with vertex set $V = \{v_1, v_2, \cdots, v_n\}$ and edge set E. Let V^1 be a copy of the vertex set and u be a single vertex. Then the Mycielskian $\mu(G)$ has the vertex set $V^0 \cup V^1 \cup \{u\}$. The edge set of $\mu(G)$ is the set $\{v_i^0 v_j^0 : v_i v_j \in E\} \cup \{v_i^0 v_j^1 : v_i v_j \in E\} \cup \{v_j^1 u : \forall v_j^1 \in V^1\}$.

In general,

Let G be a graph with vertex set $V = \{v_1, v_2, \cdots, v_n\}$ and edge set E and let m be any positive integer. For each integer $k(0 \leq k \leq m)$, let V^k be a copy of vertices in V, that is $V^k = \{v_1^k, v_2^k, \cdots, v_n^k\}$. The m - mycielskian $\mu_m(G)$ has the vertex set $V^0 \cup V^1 \cup \cdots \cup V^m \cup \{u\}$ where u is a single vertex. The edge set of $\mu_m(G)$ is the set $\{v_i^0 v_j^0 : v_i v_j \in E\} \cup \left(\cup_{k=0}^{m-1} \{v_i^k v_j^{k+1} : v_i v_j \in E\}\right) \cup \{v_j^m u : \forall v_j^m \in V^m\}$.

W. Lin et al [13] define $\mu_0(G)$ to be the graph obtained from G by adding a universal vertex u.

We observe that every vertex v_i^k in V^k is adjacent to the vertices v_j^{k+1} in V^{k+1}

and v_j^{k-1} in V^{k-1}, $k = 1,2,\cdots,m-1$ if v_i is adjacent to v_j in G. No two vertices in V^k are adjacent to each other except $k = 0$ and v_i^k and v_i^l are not adjacent, for all i,k,l. Also, $degv_i^j = 2degv_i$, for all $j = 0,1,\dots,m-1$; $degv_i^m = degv_i + 1$ and $degu = |V(G)|$.

We define the subset A of $V(G)$ as $A = \{x|x^k \in A^k\}$ where A^k is the subset of $V^k, (k = 0,1,2,\dots,m-1)$.

3 Various Dominations on Mycielski's Graph

Theorem 3.1

For any graph G, $\gamma_s(\mu(G)) = \gamma_s(G) + 1$.

Proof :

Let A be a γ_s-set of G. Then $A \cup \{u\}$ is a strong dominating set of $\mu(G)$. Hence $\gamma_s(\mu(G)) \leq \gamma_s(G) + 1$. Suppose $\gamma_s(\mu(G)) \leq \gamma_s(G)$. Let D be γ_s-set of $\mu(G)$. Then, D must contain the vertex u. Otherwise no vertex in D can dominate u. Now, let $D = A^0 \cup B^1 \cup \{u\}$. If B^1 is empty, then $D = A^0 \cup \{u\}$ implies that A is a γ_s- set of G with $|A| < \gamma_s(G)$. If B^1 is non empty, Then $D_1 = A^0 \cup B^0 \cup \{u\}$ where $B^0 = \{x^0|x^1 \in B^1\}$. Since B^0 does not affect the condition of strong domination, D_1 is also γ_s - set of $\mu(G)$. Then $A \cup B$ is a γ_s - set of G with the cardinality less than $\gamma_s(G)$. Since both cases leads to the contradiction, $\gamma_s(\mu(G)) = \gamma_s(G) + 1$, for any graph G.■

Theorem 3.2

For any graph G, $\gamma_w(G) + 1 \leq \gamma_w(\mu(G)) \leq 2\gamma_w(G)$

Proof :

Let D be a optimal weak dominating set of $\mu(G)$. Then $D = A^0 \cup B^1 \cup \{u\}$. If B^1 is empty, then all the vertices in V^1 may be weakly dominated by the vertices of A^0. Hence $\gamma_w(\mu(G) = |D| = |A^0| + 1$. It clear that the set $A = \{x \in V(G)|x^0 \in V^0(G)\}$ is weakly dominating set of G. $|A| \geq \gamma_w(G)$. $\gamma_w(\mu(G)) \geq \gamma_w(G) + 1$. If B^1 is non empty, there exists a vertex $v^1 \in B^1$ such that $degv^1 \leq degu$. Hence $u \notin D$. $D = A^0 \cup B^1$. $\gamma_w(\mu(G)) = |D| = |A^0 \cup B^1| \leq 2\gamma_w(G)$.■

Theorem 3.3

The Mycielskian number of a graph G under the dom chromatic, strong chromatic and weak chromatic domination is $V(\mu(G))$.

Proof :

Let D be the dominating set under the domination parameters such as dom chromatic, strong chromatic and weak chromatic dominating set of $\mu(G)$. Then, $\chi(\langle D\rangle) = \chi(\mu(G))$. Since $\mu(G)$ is χ - critical, the only dominating set under these domination parameter is the vertex set of $\mu(G)$.

$$\therefore \gamma_{ch}(\mu(G)) = V(\mu(G))$$
$$\gamma_c^s(\mu(G)) = V(\mu(G))$$
$$\gamma_c^w(\mu(G)) = V(\mu(G)).$$

■

4 Various Dominations on Generalized Mycielskian Graph

Theorem 4.1

For a graph G, $\left\lceil \frac{m}{2} \right\rceil \gamma_s(G) + 1 \leq \gamma_s(\mu_m(G)) \leq m\gamma_s(G) + 1$

Proof :

Let D be a γ_s - set of G. Then D^0 may or may not be strongly dominate all the vertices of V^1.

Case : 1

Suppose if D^0 may not be strongly dominate all the vertices of V^1. Then $D^0 \cup D^1 \cup D^2 \cup \ldots \cup D^{m-1} \cup \{u\}$ is the strong dominating set of $\mu_m(G)$. $\gamma_s(\mu_m(G)) \leq |D^0 \cup D^1 \cup D^2 \cup \ldots \cup D^{m-1}| + 1 = m\gamma_s(G) + 1$

Case : 2

Suppose if D^0 may be dominate all the vertices in V^1. Then, $D^i \cup D^{i+1}$ is strongly dominate all the vertices in $V^{i-1} \cup V^i \cup V^{i+1} \cup V^{i+2}$. If $m = 4k, 4k-1$, choose $2k$ appropriate strong dominating subsets D^r and if $m = 4k+1, 4k-2$, choose $2k+1$ appropriate strong dominating subsets D^r. Also, the vertex u strongly dominates all the vertices in V^m. Hence $\gamma_s(\mu_m(G)) \geq \left\lceil \frac{m}{2} \right\rceil \gamma_s(G) + 1$.■

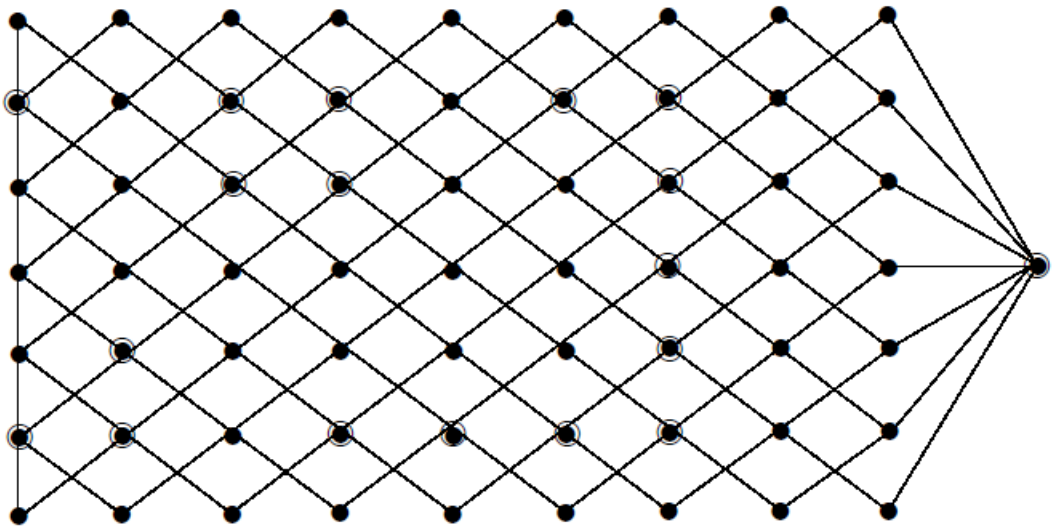
Illustrative Example 4.2 *For upper Bound,*

Consider the graph $G = P_3$ then the strong dominating set $D = \{x\}$, where x is a unique full degree vertex in P_3 and $\gamma_s(P_3) = 1$. It is clear that there exists no vertex v to strongly dominate the vertex x^i in each V^i, for $i = 0,1,\ldots,m-1$. Hence $S = \{x^0, x^1, \ldots, x^{m-1}, u\}$ is the γ_s - set of $\mu_m(G)$. Also, $\gamma_s(\mu_m(G)) = m + 1 = m\gamma_s(G) + 1$

Illustrative Example 4.3 *For lower Bound,*

Consider the graph $G = P_4$ then the strong dominating set $D = \{x, y\}$, where x and y are supporting vertices in P_4 and $\gamma_s(P_4) = 2$. Let S be a strong dominating set of $\mu_m(G)$. It is clear that the vertices x^0, y^0 strongly dominates all the vertices in $V^0 \cup V^1$ then for $m = 4k-2$ choose $S = D^0 \cup D^3 \cup D^4 \cup \ldots \cup D^{m-3} \cup D^{m-2} \cup \{u\}$; for $m = 4k-1$ choose $S = D^0 \cup D^1 \cup D^4 \cup D^5 \cup \ldots \cup D^{m-3} \cup D^{m-2} \cup \{u\}$; for $m = 4k$ choose $S = D^1 \cup D^2 \cup D^5 \cup D^6 \cup \ldots \cup D^{m-3} \cup D^{m-2} \cup \{u\}$ and for $m = 4k+1$ choose $S = D^0 \cup D^2 \cup D^3 \cup D^6 \cup D^7 \cup \ldots \cup D^{m-3} \cup D^{m-2} \cup \{u\}$. Hence $\gamma_s(\mu_m(P_4)) = \left\lceil \frac{m}{2} \right\rceil \gamma_s(P_4) + 1$.

Illustrative Example 4.4 *Consider the following graph* $\mu_8(P_7)$



For this graph, $m = 8$; $\gamma_s(P_7) = 3$ and $\gamma_s(\mu_8(P_7)) = 18$.The encircled vertices form a γ_s - set. Also, $\left\lceil \frac{m}{2} \right\rceil \gamma_s(G) + 1 = 13 \leq \gamma_s(\mu_8(P_7)) \leq 25 = m\gamma_s(G) + 1$

Theorem 4.5

For a graph G, $\gamma_w(\mu_m(G)) \leq (m+1)\gamma_w(G)$ *and*

$$\gamma_w(\mu_m(G)) \geq \begin{cases} \left(\left\lfloor \frac{m}{2} \right\rfloor + 1\right)\gamma_w(G) & if m is even \\ \left(\left\lfloor \frac{m}{2} \right\rfloor + 1\right)\gamma_w(G) + 1 & if m is odd \end{cases}$$

Proof :

Let D be a γ_w - set of G. Then D^0 may or may not be weakly dominate all the vertices of V^1.

Case : 1

Suppose if D^0 may not be weakly dominate all the vertices of V^1. Then $D^0 \cup D^1 \cup D^2 \cup \ldots \cup D^{m-1} \cup D^m$ is the weak dominating set of $\mu_m(G)$. Therefore, $\gamma_w(\mu_m(G)) \leq |D^0 \cup D^1 \cup D^2 \cup \ldots \cup D^{m-1} \cup D^m| = (m+1)\gamma_w(G)$

Case : 2

Suppose if D^0 may be dominate all the vertices in V^1. Then, $D^i \cup D^{i+1}$ is weakly dominate all the vertices in $V^{i-1} \cup V^i \cup V^{i+1} \cup V^{i+2}$. If $m = 4k, 4k+1$, choose $2k+1$ appropriate weakly dominating subsets D^r and if $m = 4k-1, 4k-2$, choose $2k$ appropriate weak dominating subsets D^r. Also, the vertex v^m in V^m weakly dominates the vertex u, for an odd $m.\gamma_w(\mu_m(G)) \geq \begin{cases} \left(\left\lfloor \frac{m}{2} \right\rfloor + 1\right)\gamma_w(G) & if m is even \\ \left(\left\lfloor \frac{m}{2} \right\rfloor + 1\right)\gamma_w(G) + 1 & if m is odd \end{cases}$. ■

Illustrative Example 4.6 *For upper Bound,*

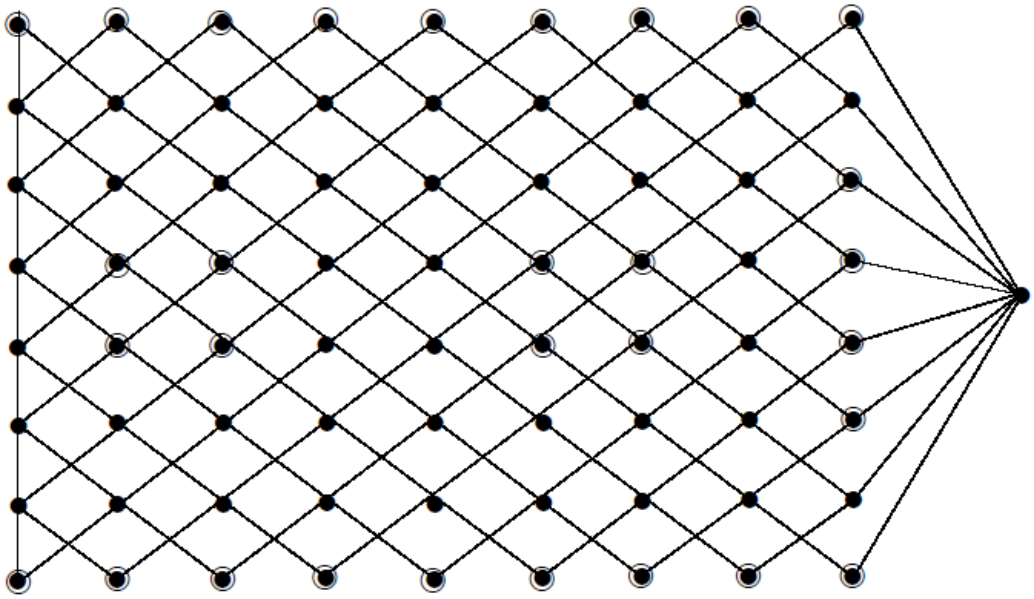
Consider the graph $G = P_4$ then the weak dominating set $D = \{x, y\}$, where x and y are pendant vertices in P_4 and $\gamma_w(P_4) = 2$. It is clear that there exists no vertex v to weakly dominate the vertex x^i, y^i of degree two in each V^i, for $i = 0,1, \dots, m-1$. Hence $W = \{x^0, y^0, x^1, y^1, \dots, x^m, y^m\}$ is the γ_w - set of $\mu_m(G)$. Also, $\gamma_w(\mu_m(G)) = 2(m+1) = (m+1)\gamma_w(G)$

Illustrative Example 4.7 *For lower Bound,*

Consider the graph $G = C_4$ then the weak dominating set $D = \{x, y\}$, where x and y are any two adjacent vertices in C_4 and $\gamma_w(C_4) = 2$. Let W be a weak dominating set of $\mu_m(G)$. It is clear that the vertices x^0, y^0 weakly dominates all the vertices in $V^0 \cup V^1$ then for $m = 4k-2$ choose $S = D^1 \cup D^2 \cup D^5 \cup D^6 \cup \dots \cup D^{m-1} \cup D^m$; for $m = 4k-1$ choose $S = D^1 \cup D^2 \cup D^5 \cup D^6 \cup \dots \cup D^{m-2} \cup D^{m-1} \cup \{v^m\}$; for $m = 4k$ choose $S = D^0 \cup D^3 \cup D^4 \cup D^7 \cup D^8 \cup \dots \cup D^{m-1} \cup D^m$ and for $m = 4k+1$ choose $S = D^0 \cup D^2 \cup D^3 \cup D^6 \cup D^7 \cup \dots \cup D^{m-2} \cup D^{m-1} \cup \{v^m\}$.

Hence $\gamma_w(\mu_m(G)) \geq \begin{cases} \left(\left\lfloor \frac{m}{2} \right\rfloor + 1\right)\gamma_w(G) & if\, m\, is\, even \\ \left(\left\lfloor \frac{m}{2} \right\rfloor + 1\right)\gamma_w(G) + 1 & if\, m\, is\, odd \end{cases}$.

Illustrative Example 4.8 *Consider the following graph* $\mu_8(P_8)$



For this graph, $m = 8$; $\gamma_w(P_8) = 3$ and $\gamma_w(\mu_8(P_8)) = 30$.The encircled vertices form a γ_w - set. Also, $\left(\left\lfloor\frac{m}{2}\right\rfloor + 1\right)\gamma_w(P_8) = 20 \leq \gamma_w(\mu_8(P_8)) \leq 36 = (m+1)\gamma_w(P_8)$

Theorem 4.9
The generalized mycielskian graph, $\mu_m(G)$ of a strong efficient open dominating graph $G \neq K_2$ not a strong efficient open dominatable.

Proof:

Let G be a strong efficient open dominating graph. Then $degv \leq \Delta \leq \frac{n}{2}$. Let Δ_i denotes the maximum degree of the vertices $V^i \subseteq V(\mu_m(G))$, for $i = 0,1,\ldots,m$. Then $\Delta_i \leq n$, for $i = 0,1,\ldots,m-1$; $\Delta_m \leq \frac{n}{2} + 1$ and $degu = n$. Since $G \neq K_2$, $degu \neq degv_i^m$, for all i. Hence the strong neighborhood set of a vertex u is empty. Hence $\mu_m(G)$ is not a strong efficient open dominatable. ■

Corrolary 4.10
The mycielski's graph $\mu(G)$ is not a strong efficient open dominatable, for a strong efficient open dominating graph G.

Theorem 4.11
The Mycielski's graph $\mu_0(G)$ is not a strong efficient open dominatable.

Theorem 4.12
For a graph $G \neq K_2$, the generalized mycielskian graph, $\mu_m(G)$ is not a weak efficient open dominatable.

Proof:

Let x be a vertex of G such that $degx = \delta(G)$. Then $degx^m = \delta + 1$. Since $G \neq K_2$, $degx^m \neq degu$ and $degx^m \neq degy^{m-1}$, for all $y \in N(x)$, because $degy^{m-1} \geq 2\delta$. Hence $N_w(x^m) = \phi$. Hence clearly, $\mu_m(G)$ is not a weak efficient open dominatable, for any graph G.■

Corrolary 4.13
For a graph $G \neq K_2$, the mycielski's graph $\mu(G)$ is not a weak efficient open dominatable.

Theorem 4.14
The Mycielski's graph, $\mu_0(G)$ is not a weak efficient open dominatable.

Remark 4.15
Let $G = K_2$, then $\mu_m(K_2) = C_{2m+3} \neq C_{4k}$. Hence $\mu_m(G)$ is neither efficiently open dominatable nor strong(weak) efficient open dominatable.

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Some Results On Strong Efficient Open Domination

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Abstract: Let $G = (V, E)$ be a simple, undirected, finite graph without isolated vertices. A subset D of $V(G)$ is a *Strong(Weak) efficient open dominating set* of G if $|N_s(v) \cap D| = 1(|N_w(v) \cap D| = 1)$, for every $v \in V(G)$ where $N_s(v)$ and $N_w(v)$ are strong and weak neighborhood respectively. A graph G is *strong(weak) efficient open dominating graph* if it contains a strong(weak) efficient open dominating set. In this paper we determine the Nordhaus - Gaddum type results, strong efficient open domatic partition, and some operations on strong efficient open domination.

Key words: Strong(weak) Efficient Open Domination, Nordhaus-Gaddum type results, Domatic Partition.

1 Introduction

Let $G = (V, E)$ be a finite, simple, undirected graph without isolated vertices. The *open neighborhood* of a vertex $v \in V(G)$ is $N(v) = \{u \in V(G)|uv \in E(G)\}$ and *closed neighborhood* of $v \in V$ is $N[v] = N(v) \cup \{v\}$. If v is a vertex of $V(G)$, then the degree of v is defined by the cardinality of $N(v)$ and is denoted by $degv$. The minimum and maximum degree of the vertices of G is denoted by δ and Δ respectively. For graph theoretic terminology, we refer to [3] and [5]. The strong neighborhood and weak neighborhood of a vertex $v \in V(G)$ are defined by $N_s(v) = \{u \in V(G)|uv \in E(G)\ anddegu \geq degv\}$ and $N_w(v) = \{u \in V(G)|uv \in E(G)\ anddegu \leq degv\}$ respectively. A subset D of V is a *dominating set* of G if every vertex in $V - D$ is adjacent to atleast one vertex in D. The *domination number*, $\gamma(G)$ of G is the minimum cardinality of a dominating set of G. In [10], Prof. E. Sampathkumar and L. Pushpalatha have defined strong(weak) domination in graphs. A subset D of V is called a *strong(weak) dominating set* of G if for every vertex, v in $V - D$ there exists $u \in D$ such that $uv \in E(G)$ and $degu \geq degv$ $(degv \geq degu)$. The *strong(weak) domination number*, $\gamma_s(G)$ $(\gamma_w(G))$ of G is the minimum cardinality of a strong(weak) dominating set of G. D.W.Bange et al [4] defined an efficient dominating set D as a set of vertices of a graph G such that $|N[v] \cap D| = 1$, for every $v \in V(G)$ and N. Meena et al [9] extend this into strong(weak) efficient dominating set D as a set of vertices of a graph G such that $|N_s[v] \cap D| = 1$ ($|N_w[v] \cap D| = 1$), for every $v \in V(G)$. The *strong(weak) efficient domination number*, $\gamma_{se}(G)$ $(\gamma_{we}(G))$ of G is the minimum cardinality of a strong(weak) efficient dominating set of G.

A subset D of V is a *total dominating set* of G if every vertex in V is adjacent to atleast one vertex in D. The *total domination number*, $\gamma_t(G)$ of G is the minimum

cardinality of a total dominating set of G. Gavlas and Schultz are defined an efficient open dominating set D as a set of vertices of a graph G such that $|N(v) \cap D| = 1$, for every $v \in V(G)$ in [7]. The *efficient open domination number*, $\gamma_{te}(G)$ of G is the cardinality of a efficient open dominating set of G. A study of domination, total domination and efficient open domination in graphs and its advanced topics are given in [2, 6, 8, 11]. We introduced a strong(weak) efficient open domination in graphs [1]. In this paper we determine the Nordhaus - Gaddum type results, strong efficient open domatic partition, and some operations on strong efficient open domination.

2 Strong(Weak) Efficient Open Domination

Definition 2.1 *[1]*
A subset D of $V(G)$ is called a strong efficient open dominating set (or SEOD set, for short) of G if $|N_s(v) \cap D| = 1$, for every $v \in V(G)$. A subset D of $V(G)$ is called a weak efficient open dominating set (or WEOD set, for short) of G if $|N_w(v) \cap D| = 1$, for every $v \in V(G)$.

The strong (weak) efficient open domination number, denoted by $\gamma_{ste}(G)$ ($\gamma_{wte}(G)$), is the minimum cardinality of a strong (weak) efficient open dominating set of G. We also call the corresponding set that γ_{ste} (γ_{wte}) - set of G.

A graph G is called a strong (weak) efficient open dominating graph or SEOD (WEOD) graph if it contains a strong(weak) efficient open dominating set. Also says that G is strong (weak) efficient open dominatable.

3 Nordhaus - Gaddum Type Results

Lemma 3.1
Let G be a strong efficient open dominating graph with non increasing degree sequence $(d_1, d_2, \dots, d_n)$ and the complement $\bar{G}$ of G is strong efficient open dominatable then G is either regular or

$$d_i = \begin{cases} \frac{n}{2} & if\ i \le t \\ \frac{n}{2} - 1 & if\ i > t \end{cases}$$ where both n and $t > 0$ are even.

Proof:

Let G and its complement $\bar{G}$ be strong efficient open dominationg graph. Then $\Delta(G) \le \frac{n}{2}$ and $\Delta(\bar{G}) \le \frac{n}{2}$. Hence $\Delta(G) \le \frac{n}{2}$ and $\delta(\bar{G}) \ge n - 1 - \frac{n}{2} = \frac{n}{2} - 1$ Therefore, $\frac{n}{2} - 1 \le degv \le \frac{n}{2}$, for all $v \in V$. If n is odd, then G is $\frac{n-1}{2}$ - regular graph. If n is even, $degv$ is either $\frac{n}{2}$ or $\frac{n}{2} - 1$, for all $v \in V$. Hence G may be regular.

Otherwise, let t be an positive integer such that t number of vertices have a degree $\frac{n}{2}$ and $n - t$ vertices have a degree $\frac{n}{2} - 1$ in G. Since n is even, either $\frac{n}{2}$ or $\frac{n}{2} - 1$ is odd. Suppose if $\frac{n}{2}$ is odd, then t is even and also $n - t$ is even. Suppose if $\frac{n}{2}$ is even, then $\frac{n}{2} - 1$ is odd. Therefore $n - t$ is even implies t is even.

$$\therefore d_i = \begin{cases} \frac{n}{2} & if i \le t \\ \frac{n}{2} - 1 & if i > t \end{cases}$$

Theorem 3.2

A graph G and its complement $\bar{G}$ are connected strong efficient open dominating graphs then both G and $\bar{G}$ are efficient open dominatable. Moreover, $\gamma_{ste}(G) = \gamma_{ste}(\bar{G}) = 2$.

Proof:

Let a graph G and its complement, $\bar{G}$ be strong efficient open dominating graph. Then if G is regular, then clearly, both G and $\bar{G}$ are efficient open dominatable. otherwise if $d_i = \begin{cases} \frac{n}{2} & if i \le t \\ \frac{n}{2} - 1 & if i > t \end{cases}$, where both positive integer n and t are even. Since $\Delta = \frac{n}{2}$, then there exists adjacent maximum degree vertices u, v in $V(G)$ such that $\{u, v\}$ is a strong efficient open dominating set of G. Hence $\gamma_{ste}(G) = 2$.Similarly, $\gamma_{ste}(\bar{G}) = 2$.

Hence both G and $\bar{G}$ are efficient open dominatable.

Theorem 3.3

For a connected graph G, $4 \le \gamma_{ste}(G) + \gamma_{ste}(\bar{G}) \le \frac{3n}{2}$;

$4 \le \gamma_{ste}(G).\gamma_{ste}(\bar{G}) \le \frac{n^2}{2}$. The upper limit is true for C_4 and the lower limit is sharp for a connected graph, $\bar{G}$.

Proof:

Since both G and $\bar{G}$ are strong efficient open dominating graph,

$\therefore \gamma_{ste}(G) \ge 2$ and $\gamma_{ste}(G) \ge 2$

$\Rightarrow \gamma_{ste}(G) + \gamma_{ste}(\bar{G}) \ge 4$ and $\gamma_{ste}(G) \cdot \gamma_{ste}(\bar{G}) \ge 4$.

Also, for upper limit, $\gamma_{ste}(G) \le \frac{n}{2}$ and $\gamma_{ste}(\bar{G}) \le \frac{n}{2}$, for a connected $\bar{G}$.

Suppose if $\bar{G}$ is not connected and contains K_2 as a component then, $\gamma_{ste}(\bar{G}) \le n \therefore \gamma_{ste}(G) + \gamma_{ste}(G) \le \frac{n}{2} + n = \frac{3n}{2}$ and $\gamma_{ste}(G) \cdot \gamma_{ste}(G) \le \frac{n}{2} \cdot n = \frac{n^2}{2}$.

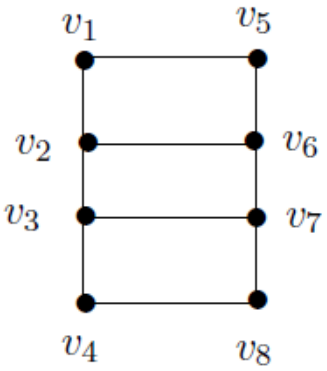
Further, $\gamma_{ste}(C_4) = 2$ and $\gamma_{ste}(\bar{C_4}) = 4$. Hence the upper limit is sharp for C_4. By theorem (3.2), the lower limit is sharp for a connected graph, $\bar{G}$.

4 Domatic Number in Strong Efficient Open Dominating Graphs

Definition 4.1

A **strong efficient open domatic partition** *is a partition of the vertices of a graph into disjoint strong efficient open dominating sets. The maximum number of disjoint strong efficient open dominating sets in a domatic partition of a graph G is called its* **strong efficient open domatic number** *and is denoted by $d_{ste}(G)$.*

Example 4.2 *Consider the following graph G*



There exists two strong efficient open dominating sets D_1 and D_2, where $D_1 = \{v_1, v_2, v_3, v_4\}$ and $D_2 = \{v_5, v_6, v_7, v_8\}$. Also, $D_1 \cap D_2 = \phi$ and $D_1 \cup D_2 = V(G)$. Hence $d_{ste}(G) = 2$.

Proposition 4.3

For a cycle $C_{4n}, (n \geq 1)$, $d_{ste}(C_{4n}) = 2$.

Proof:

Let $V(C_{4n}) = \{x_1, x_2, \dots, x_{4n}\}$. It is clear that the γ_{ste} - set of C_{4n} are $\{x_1, x_2, x_5, x_6, \dots, x_{4n-3}, x_{4n-2}\}$ and $\{x_3, x_4, x_7, x_8, \dots, x_{4n-1}, x_{4n}\}$.

Hence, clearly $d_{ste}(C_{4n}) = 2, n \geq 1$.

Proposition 4.4

For a cycle $K_{m,m}, (m \geq 1)$, $d_{ste}(K_{m,m}) = m$.

Proof:

Let (X, Y) be the partition of the vertex set of $K_{m,m}$, where $X = \{x_1, x_2, \dots, x_m\}$ and $Y = \{y_1, y_2, \dots, y_m\}$ the distinct γ_{ste} - sets of $K_{m,m}$ are $\{x_1, y_1\}$, $\{x_2, y_2\}$,..., $\{x_m, y_m\}$.

Hence, $d_{ste}(K_{m,m}) = m$.

Theorem 4.5

For any strong efficient open dominating graph G *with* $\delta > 1$, $d_{ste}(G) = 1$.

Proof:

Let G be a strong efficient open dominating graph with $\delta = 1$. Then all the strong efficient open dominating set of G must contains the supporting vertices of G. Hence, $d_{ste}(G) = 1$.

Theorem 4.6

For any strong efficient open dominating graph G *with exactly two maximum degree vertices,*

$d_{ste}(G) = 1$.

Proof:

Let G be a strong efficient open dominating graph with exactly two maximum degree vertices v_1 and v_2. Then v_1 and v_2 must be adjacent. Also, $N_s(v_1) = \{v_2\}$ and $N_s(v_2) = \{v_1\}$. Then every strong efficient open dominating set of G must contain the vertices $\{v_1, v_2\}$. Hence $d_{ste}(G) = 1$

5 Operations on Strong Efficient Open Domination

In this section, we disscuss about the strong efficient open domination in the union, $G_1 \cup G_2$, cartesian product $G_1 \blacksquare G_2$, and direct product $G_1 \times G_2$ of the graphs G_1 *and* G_2.

Proposition 5.1

A graph $G_1 \cup G_2$ is need not to be a strong efficient open dominating graph if G_1 and G_2 is strong efficient open dominatable.

Example 5.2

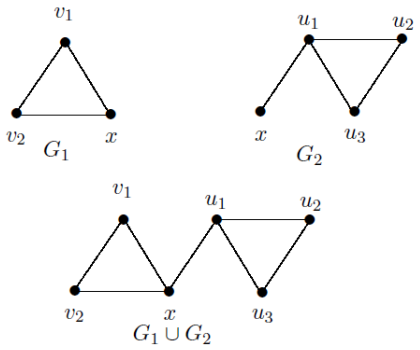
- P_2 and P_4 are strong efficient open dominating graph with a common unique end vertex. Then clearly, $P_2 \cup P_4 = P_5$, is not a strong efficient open dominating graph.
- $D_{r,r}$ and P_2 are strong efficient open dominating graph with a common unique end vertex. Then clearly, $D_{r,r} \cup P_2$, is not a strong efficient open dominating graph.

Remark 5.3

A graph $G_1 \cup G_2$ is a strong efficient open dominating graph for a non strong efficient open dominating graph G_1 and G_2.

Example 5.4

Consider the following graphs G_1 and G_2.



Clearly, both G_1 and G_2 are not a strong efficient open dominatable whereas $G_1 \cup G_2$ is a strong efficient open dominating graph.

Theorem 5.5
Let G_1 and G_2 be any two strong efficient open dominating graph with a γ_{ste} - set D_1 and D_2 respectively. Then $G_1 \cup G_2$ is a strong efficient open dominatable if $V(G_1) \cap V(G_2)$ contains the adjacent pair of maximum degree vertices in both $\langle D_1 \rangle$ and $\langle D_2 \rangle$.

Proof:
Let G_i be a strong efficient open dominating graph with γ_{ste} - set D_i, $(i = 1,2)$ and let x, y be the adjacent maximum degree vertices of both G_1 and G_2 with an edge. Then $x, y, xy \in \langle D_1 \rangle \cap \langle D_1 \rangle$ Then $deg_{G_1 \cup G_2} x = deg_{G_1 \cup G_2} y = \Delta(G_1 \cup G_2) = \Delta(G_1) + \Delta(G_2)$ Hence, $G_1 \cup G_2$ is a strong efficient open dominating graph with $\gamma_{ste}(G_1 \cup G_2) = \gamma_{ste}(G_1) + \gamma_{ste}(G_2) - 2$.

Note 5.6
A graph $G_1 \cup G_2$ is strong efficient open dominating graph if either $G_1 \subseteq G_2$ or $G_2 \subseteq G_1$

Remark 5.7
A join sum of two complete graph, G has exactly two maximum degree vertices u, v (say) with a bipartition $N(u)$ and $N(v)$ of $V(G)$. Hence G is strong efficient open dominatable with $\gamma_{ste}(G) = 2$.

Proposition 5.8
A graph $G_1 \blacksquare G_2$ is need not to be a strong efficient open dominating graph if G_1 and G_2 is strong efficient open dominatable.

Example 5.9
- $P_4 \blacksquare P_8$ and $P_4 \blacksquare P_4$ are strong efficient open dominatable.
- $P_4 \blacksquare P_{12}$ and $P_8 \blacksquare P_8$ are not a strong efficient open dominatable

Remark 5.10
A graph $G_1 \times G_2$ is need not to be a strong efficient open dominating graph if G_1 and G_2 is strong efficient open dominatable.

Theorem 5.11
For a strong efficient open dominating paths P_{n_1} and P_{n_2}, $P_{n_1} \times P_{n_2}$ is a strong efficient open dominating graph.

Proof:
Let P_{n_1} and P_{n_2} be the distinct strong efficient open dominating paths with the vertex set $V(P_{n_1}) = \{u_1, u_2, \dots, u_{n_1}\}$ and $V(P_{n_1}) = \{v_1, v_2, \dots, v_{n_1}\}$ respectively. Then $V(P_{n_1} \times P_{n_1}) = \{(u_i, v_j) | 1 \le i \le n_1; 1 \le j \le n_2\}$
Let $Y_i = \{2,3,6,7, \dots, 4k-2, 4k-3, \dots, n_i - 2, n_i - 1\}, (i = 1,2)$.
Then $D = \{(u_{y_1}, v_{y_2}) | y_1 \in Y_1 and y_2 \in Y_2\}$ form a strong efficient open dominating set

of $P_{n_1} \times P_{n_2}$. Hence proved.

Remark 5.12

Clearly all the vertices of D, defined in above theorem, are equal in degree ($deg\ v = 4$, for all $v \in D$) and $\langle D \rangle = mK_2, (m > 1)$. Also the strong efficient open dominating set with less than $|D|$ can not be found.

$\therefore \gamma_{ste}(P_{n_1} \times P_{n_2}) = \gamma_{ste}(P_{n_1}) \cdot \gamma_{ste}(P_{n_2})$.

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Chromatic total domination in graphs

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Chromatic total domination in graphs

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Abstract

Let $G = (V, E)$ be a simple, finite and undirected graph and without isolated vertex. A set $D \subseteq V$ is said to be chromatic total dominating set of G if D is a total dominating set and $\chi(<D>) = \chi(G)$. The minimum cardinality of a chromatic total dominating set of G is called the chromatic total domination number of G and is denoted by $\gamma_{ch}^{t}(G)$. In this paper, we discuss the chromatic total domination number for standard graphs.

Subject Classification: *05C69*

Keywords: *Total domination, Chromatic total domination, Chromatic total domination number.*

1. Introduction

Let $G = (V, E)$ be a simple, finite and undirected graph and without isolated vertex. A subset D of V is said to be dominating set if for every u in $V - D$ there exist a vertex u in D such that u and v are adjacent. The minimum cardinality of a dominating set of G is called the domination number of G and is denoted by $\gamma(G)$. D is minimal dominating set of a graph G if no proper subset of D is a dominating set of G. D is a total dominating set of G if $< D >$ has no isolates. The minimum cardinality of a total dominating set of G is called the total domination number of G and is denoted by $\gamma_t(G)$. The total domination in graphs was introduced by the authors Cockayne, Dawes and Hedetniemi[2]. For a recent survey of total domination in graphs can be found in [4]and further notations used in the paper we refer[3] .A clique C is a subset of vertices of G such that every two distinct vertices in the clique are adjacent that is , its induced subgraph is complete .The clique number of a graph G, denoted $\omega(G)$, is the number of vertices in a maximum clique of G. In this paper we obtain the chromatic total domination number for standard graphs.

Theorem 1.1 : [3] *If G contains full degree vertex,* $\gamma_t(G) = 2$

Corollary 1.2 : [3]

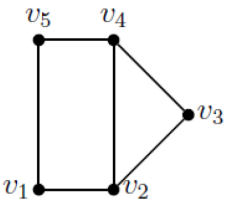
(*i*) $\gamma_t(K_n) = 2$

(*ii*) $\gamma_t(C_n) = \gamma_t(P_n) = \begin{cases} n/2 & \text{if } n \equiv 0 (\text{mod } 4) \\ (n+1)/2 & \text{if } n \equiv 1,3 (\text{mod } 4) \\ \dfrac{n}{2}+1 & \text{if } n \equiv 2 (\text{mod } 4) \end{cases}$

2. Chromatic Total Dominating set

Definition 2.1 : A set $D \subseteq V$ is said to be chromatic total dominating set of G if D is a total dominating set and $\chi(<D>) = \chi(G)$. The minimum cardinality of a chromatic total dominating set of G is called the chromatic total domination number of G and is denoted by $\gamma_{ch}^{t}(G)$.

Examples 2.2 : Consider the following graph:



G

Let $D = \{v_2, v_3, v_4\}$. Then D is a total dominating set and $\chi(<D>) = \chi(G)$. Hence D is a chromatic total dominating set of G.

Theorem 2.3 : *Let $G = K_n$, then $\gamma_{ch}^{t}(K_n) = n$.*

Proof : Let G be a complete graph with n vertices. Let D be a γ_t set of G. Then $\chi(G) = \chi(K_n)$. Since $\chi(G) \leq \gamma_{ch}^{t}(G)$, $n \leq \gamma_{ch}^{t}(G)$. Therefore $\gamma_{ch}^{t}(K_n) = n$.

Theorem 2.4 : *Let $G = F_n$. Then $\gamma_{ch}^{t}(F_n) = 3$ for all $n \geq 3$.*

Proof : Let G be a fan graph with n vertices. $\gamma_t(F_n) = 2$ and $\chi(F_n) = 3$. Let u be vertex of F_n with $deg(u) = n - 1$. Let $x, y \in (F_n) - \{u\}$ such that $xy \in E(F_n)$. Let $D = \{u, x, y\}$. Then $\chi(<D>) = \chi(F_n)$. Therefore D is a chromatic total dominating set of G. Therefore $3 = \chi(F_n) \leq \gamma_{ch}^{t}(F_n) \leq |D| = 3$.Therefore $\gamma_{ch}^{t}(F_n) = 3$.

Theorem 2.5 : *Let $G = K_{1,n}$. Then $\gamma_{ch}^{t}(K_{1,n}) = 2$.*

Proof : Let $G = K_{1,n}$ be a star graph with $n + 1$ vertices. $\gamma_t(K_{1,n}) = 2$ and $\chi(K_{1,n}) = 2$. Let u be a vertex of $K_{1,n}$ with $deg(u) = n - 1$. Let $x \in V(K_{1,n}) - \{u\}$ such that $ux \in E(K_{1,n})$. Let $D = \{u, x\}$. Then $\chi(<D>) = 2 = \chi(K_{1,n})$. D is a chromatic total dominating set. Therefore $2 = \chi(K_{1,n}) \leq \gamma_{ch}^{t}(K_{1,n}) \leq |D| = 2$.Therefore $\gamma_{ch}^{t}(K_{1,n}) = 2$.

Theorem 2.6 : *Let $G = K_{m,n}$. Then $\gamma_{ch}^{t}(K_{m,n}) = 2$.*

Proof : Let G be a complete bipartite graph with m+n vertices. $\gamma_t(K_{m,n}) = 2$ and $\chi(K_{m,n}) = 2$. Let u be a vertex of $K_{m,n}$. Let $x \in V(K_{m,n}) - \{u\}$ such that $ux \in E(K_{m,n})$. Let $D = \{u, x\}$. Then $\chi(<D>) = \chi(K_{m,n})$. Therefore D is a chromatic total dominating set. $2 = \chi(K_{m,n}) \leq \gamma_{ch}^t(K_{m,n}) \leq |D| = 2$. Therefore $\gamma_{ch}^t(K_{m,n}) = 2$.

Theorem 2.7 : *Let $G = W_n$. Then $\gamma_{ch}^t(W_n) = n$ if n is even $\gamma_{ch}^t(W_n) = 3$ if n is odd*

Proof : Let W_n be a wheel with n vertices and $V(W_n) = \{x, v_1, v_2, ..., v_{n-1}\}$. Let x be a full degree vertex of W_n.

Case (i) : If n is even and let $D = (x, v_j)$ for some $i = 1, 2, \ldots, n-1$.

Then D is a total dominating set of W_n, but $\chi(<D>) \neq \chi(W_n) = 4$. Let $D_1 = \{x, v_1, v_2, ..., v_{n-1}\}$. Then $\chi(<D>) = 4 = \chi(G)$. Therefore D is a unique chromatic total dominating set of W_n. Therefore $\gamma_{ch}^t(W_n) = |D_1| = n$.

Case (ii) : If n is odd and let $D = \{x\}$.

Then D is the total dominating set of W_n. But $\chi(<\{x\}>) \neq \chi(W_n) = 3$. Let $D_1 = \{x_2, v_i, v_j\}$ where $v_i v_j \in E(W_n)$. Then clearly $\chi(<D_1>) = 3 = \chi(W_n)$. D is a chromatic total dominating set of W_n.

Therefore, $\gamma_{ch}^t(W_n) = 3$.

Observation 2.8 :

(i) If $\chi(G) = 2$, then $\gamma_t(G) = \gamma_{ch}^t(G)$.

(ii) If G is χ critical, then $\gamma_{ch}^t(G) = \gamma_{cc}(G) = \gamma_{ch}(G) = \gamma_s^c(G) = \gamma_w^c(G)$.

Theorem 2.9 : *Let $G = P_n$. Then* $\gamma_{ch}^t(P_n) = \begin{cases} n/2 & \text{if } n \equiv 0 (\text{mod } 4) \\ (n+1)/2 & \text{if } n \equiv 1, 3 (\text{mod } 4) \\ \frac{n}{2} + 1 & \text{if } n \equiv 2 (\text{mod } 4) \end{cases}$

Proof : Let G be a path with n vertices and $V(G) = \{v_1, v_2, ..., v_n\}$. Let D be a minimal total dominating set of G. Then $\chi(<D>) = 2$. Therefore D is a minimal chromatic total dominating set of G. Therefore $\gamma_{ch}^t(G) = |D| = \gamma_t(G)$.

Theorem 2.10 : *Let $G = C_n$. Then* $\gamma_{ch}^t(C_n) = \begin{cases} n & \text{if } n \text{ is odd} \\ n/2 & \text{if } n \equiv 0 (\text{mod } 4) \text{ and } n \text{ is even} \\ \frac{n}{2} + 1 & \text{if } n \equiv 2 (\text{mod } 4) \text{ and } n \text{ is odd} \end{cases}$

Proof : Let $G = C_n$ be a cycle on $n \geq 3$ vertices and $V(C_n) = \{v_1, v_2, ..., v_n\}$.

Case (i): n is odd

Then G is χ-critical graph. Since every chromatic total dominating set is a dom chromatic set, $n = \gamma_{ch}(G) \leq \gamma^t_{ch}(G)$. Also, $\gamma^t_{ch}(G) \leq |V(G_n)| = n$. Therefore $\gamma^t_{ch}(G) = n$.

Case (ii): n is even

Let D be a minimal total dominating set of G. Also $\chi(<D>) = 2 = \chi(G)$. Therefore D is chromatic total dominating set of G with minimum cardinality. $\gamma^t_{ch}(G) = |D| = \gamma_t(G)$.

Theorem 2.11 : *Let $G = D_{r,s}$. Then $\gamma^t_{ch}(D_{r,s}) = 2$.*

Proof : Let $G = D_{r,s}$, $(r \leq s)$ be a double star with $r + s$ vertices. Let $V(G) = \{x, y, v_1, v_2, ... v_r - 1, u_1, u_2, ..., u_{s-1}\}$ such that $xv_i \in E(G)$ and $yu_j \in E(G)$ for all $i = 1, 2, ... r - 1$ and $j = 1, 2, ... s - 1$. Let $D = \{x, y\}$ be a total dominating set of G. Since $\chi(<D>) = 2 = \chi(G)$, D is chromatic total dominating set of G. $\gamma^t_{ch}(G) \leq |D| = 2$. Since $\gamma^t_{ch}(G) \geq 2$, for all G. Therefore $\gamma^t_{ch}(G) = 2$.

Theorem 2.12 : *Let $G = K_m(a_1, a_2, ..., a_m)$, $a_i \geq 1$. Then $\gamma^t_{ch}(G) = m$.*

Proof : Let $G = K_m(a_1, a_2, ..., a_m)$ be a multistar graph and let $V(G) = \{1, 2, ..., m, a_1, a_2, ..., a_m\}$, $a_i \geq 1$. Let $D = \{1, 2, \ldots, n\}$ be the total dominating set. Then $\chi(<D>) = \chi(G)$. Therefore D is a chromatic total dominating set of G. Since $\chi(G) \leq \gamma^t_{ch}(G) \leq n$. Therefore $\gamma^t_{ch}(G) = n$.

Corollary 2.13 : Let G be any graph with corona. Then $\gamma^t_{ch}(G) = n$.

Theorem 2.14 : Given a positive integer $k \geq 1$ there exists a graph G such that $\gamma^t_{ch}(G) - \gamma_t(G) = k$

Proof : Let $G = C_{2k+1}$ then $\gamma^t_{ch}(G) = 2k + 1$ and $\gamma_t(G) = k + 1$. Therefore $\gamma^t_{ch}(G) - \gamma_t(G) = 2k + 1 - (k + 1) = k$.

Observation 2.15 :

(i) If G is χ critical then $\gamma^t_{ch}(G) \geq \omega(G)$

(ii) If G is **critical then $\gamma^t_{ch}(G) \geq \chi(G)$

(iii) Every chromatic total dominating set is a total dominating set.

Proposition 2.16 : *If G is a connected split graph and $\Delta(G) < n - 1$ then $\overline{G}$ is connected split graph with $\Delta(\overline{G}) < n - 1$*

Theorem 2.17 : *If G is connected split graph of order n and* $\Delta(G) < n - 1$ *then*

(*i*) $\gamma_{ch}^{t}(G) = \omega(G) = cpn(G)$

(*ii*) $\gamma_{ch}^{t}(\overline{G}) = \omega(\overline{G}) = cpn(\overline{G})$

Proof :

(i) Since G is a split graph, its vertex set can be partitioned into two sets X and Y such that $<X>$ is complete and $<Y>$ is totally disconnected graph. Without loss of generality $<X>$ can be assumed to be a maximum clique. When $<X>$ is maximum clique, each vertex of Y is not adjacent to atleast one vertex of X. Since $<Y>$ is totally disconnected graph, $\chi(G) = \chi(<X>)$. Therefore $cpn(G) = |X| = \omega(G)$. Let D be a γ_{ch}^{t} set of G. Then $X \subseteq D$. Since G is connected each vertex of Y is adjacent to atleast one vertex of X. This implies that each vertex of $D - X$ is adjacent to atleast one vertex of X. $D = X \Rightarrow \gamma_{ch}^{t}(G) = \omega(G) = cpn(G)$.

(ii) If G is connected split graph then from proposition (2.16) $\overline{G}$ is also connected split graph and hence the result follows.

Theorem 2.18 : *If G is any graph, then* $\gamma_t(G) \leq \gamma_{ch}^{t}(G) \leq \gamma_t(G) + \omega(G) - 2$

Proof : Let D be chromatic total dominating set of G. By the definition, D is the total dominating set of G. Then $\gamma_t(G) \leq |D| = \gamma_{ch}^{t}(G)$. Let S be a γ_t set of G. Clearly, $|S| \geq 2$. If $\chi(<S>) = \chi(G)$, then S is a chromatic total dominating set of G. Therefore $\gamma_{ch}(G) \leq |S| = \gamma_t(G)$. Otherwise, we need atmost $\omega(G) - 2$ vertices in $V(G) - S$ such that $\chi(<S \cup T>) = \chi(G)$ where T has newly collected $\omega(G) - 2$ vertices. Therefore $S \cup T$ is a chromatic total dominating set of G. $\gamma_{ch}(G) \leq |S \cup T| = \gamma_t(G) = \omega(G) - 2$.

Theorem 2.19 : *If G is a planar graph with* $diam(G) = 2$, $\chi(G) = 3$ *and* $\gamma_t(G) = 2$ *then* $3 \leq \gamma_{ch}^{t}(G) \leq 5$.

Proof : Lower bound is trivial. Let $D = \{a, b\}$ be a γ_t-set of G. Since $diam(G) = 2$, $g_0(G) = 3$ or 5.

Case (i) :

$g_0(G) = 3$, Let C be a 3-cycle $xyzx$. If $a, b \in C$ then two vertices of C are adjacent to a and one vertex is adjacent to b or vice versa, for otherwise K_4 is induced, a contradiction to $\chi(G) = 3$. Let x and y be adjacent to a and z be adjacent to b. Then $axya$ is a 3-cycle. Hence $\{a, x, y, b\}$ is a chromatic total dominating set of G. If a or b is in the 3-cycle, C then the 3-Cycle, C together with the remining vertex of D is a chromatic total dominating set of G. If $a, b \in C$ then $\{x, y, z\}$ is a chromatic total dominating set of G.

Case (ii)

$g_0 = 5$. Let C be a 5-cycle $uvwxyu$. If $a, b \in C$ then the vertices of G are adjacent to a or b and not to both otherwise 3-cycle is induced. Then D can totally dominate atmost 4 vertices of C, a contradiction. Hence a or $b \in C$. Let $a \in C$ and $b \in C$. Let $u = a$ then xw are adjacent to b and hence a 3-Cycle is induced a contradiction. Therefore both $a, b \in C$. Hence $\{u, v, w, x, y\}$ is a γ_{ch}^{t}- set of G.

Case(i) and (ii) shows that $3 \leq \gamma_{ch}^{t}(G) \leq 5$.

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Chromatic connected domination in graphs

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Abstract

Let $G = (V, E)$ be a undirected, finite, simple graph. A subset D of V is said to be dominating set if for every v in $V - D$ there exists u in D such that u and v are adjacent. A dominating set D is said to be connected dominating set if $< D >$ has connected. We introduce the concept of chromatic connected dominating set. A subset D of V is said to be chromatic connected dominating set if D is a connected dominating set and $\chi(< D >) = \chi(G)$, where $\chi(G)$ is a chromatic number of G. The minimum cardinality of chromatic connected dominating set of a graph G is called a chromatic connected domination number and is denoted by $\gamma_{cc}(G)$. We find the chromatic connected domination number for some standard graphs and characterize the graph for $\gamma_{cc}(G) = 2$.

Subject Classification: *05C69*

Keywords: *Connected domination, Chromatic connected domination, Chromatic connected domination number.*

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1. Introduction

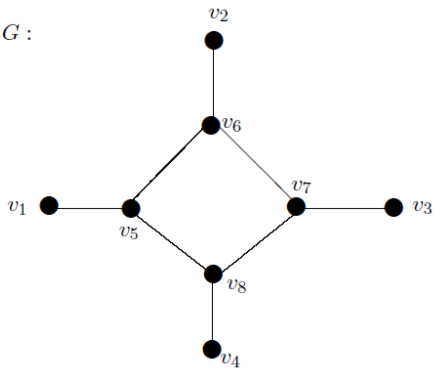
Let $G = (V, E)$ be a graph with vertex set V and edge set E of order n and size m. A *walk* W in G is a finite non-null sequence, whose terms are alternatively vertices and edges. In a walk W, if the vertices are distinct then W is called a *path*. A graph G is said to be *connected* if any two vertices of G are joined by a path; Otherwise disconnected. A graph G is said to be *k-connected* if the removal of minimum number, k of vertices from a graph G so that the resulting graph is disconnected. A *colouring* of G is an assignment of colours to the vertices of G such that no two adjacent vertices have the same colour. A *chromatic number* of G, is the minimum number of colours needed for colouring of G and is denoted by $\chi(G)$. A graph G is called χ*-critical* if $\chi(H) < \chi(G)$, for every proper subgraph, H of G[1].

Berge and Ore are introduced the concept of domination in graphs. A subset D of V is a *dominating set* of G if every vertex v in $V - D$ is dominated by some u in D. The minimum cardinality of the dominating set is called the *domination number* and is denoted by $\gamma(G)$[2]. T. N. Janakiraman and M. Poobalaranjani introduced dom-chromatic sets of graphs. A subset D of V is said to be a *dom-chromatic set* if D is a domiating set and $(< D >) = \chi(G)$. The minimum cardinality of a dom-chromatic set is called the *dom-chromatic number* and is denoted by $\gamma_{ch}(G)$[3]. E.Sampathkumar and H.B.Walikar introduced connected domination number of graphs. A dominating set D of $V(G)$ is said to be a *connected dominating set* if the subgraph $< D >$ induced by D is connected in G. The minimum cardinality of the connected dominating set of G is called the *connected domination number* of G and is denoted by $\gamma_c(G)$[4]. In this paper we define chromatic connected dominating set and find the chromatic connected domination number for some graphs.

2. Chromatic Connected Domination

Definition 2.1 : Let $G = (V, E)$ be a graph. A subset D of V is said to be chromatic connected dominating set (or *CCD*-set) if D is a connected dominating set and $\chi(< D >) = \chi(G)$. The minimum cardinality of chromatic connected dominating set in a graph G is called a chromatic connected domination number (or *CCD*- number) and is denoted by $\gamma_{cc}(G)$. The chromatic connected dominating set with cardinality $\gamma_{cc}(G)$ is called γ_{cc}-set of G.

Example 2.2 :



The chromatic connected dominating sets of G are $\{v_3, v_5, v_6, v_7\}$ and $\{v_5, v_6, v_7, v_8\}$ therefore $\gamma_{cc}(G) = 4$.

Remark 2.3 :

1. For any graph G, $\gamma_{cc}(G) \leq |V(G)|$.
2. If G is χ - critical, then $\gamma_{cc}(G) \leq |V(G)|$.
3. We observe that $1 \leq \gamma_{cc}(G) \leq n$.

Theorem 2.4 : *For any non-trivial connected graph G, $\chi(G) \leq \gamma_{cc}(G)$.*

Proof : Let D be a γ_{cc} - set of G. By the definition $< D >$ is connected dominating set and $\chi(< D >) = \chi(G)$. Therefore D is contains atleast $\chi(G)$ vertices. Hence $\gamma_{cc}(G) = |D| \geq \chi(G)$. Therefore $\gamma_{cc}(G) \geq \chi(G)$.

Theorem 2.5 : *Let P_n be a path with n vertices. Then $\gamma_{cc}(P_n) = n - 2$, for all $n \geq 4$.*

Proof : Let P_n be a path with n vertices and $V(P_n) = \{v_1, v_2, ..., v_n\}$ such that $deg(v_1) = deg(v_n) = 1$. Clearly, $\chi(P_n) = 2$. Let $D = \{v_2, v_3, ..., v_{n-1}\}$, then $< D > = P_{n-2}$. Clearly D is a connected dominating set and $\chi(< D >) = 2 = \chi(P_n)$. This implies that D is a chromatic connected dominating set.

$$\therefore \qquad \gamma_{cc}(P_n) \leq n - 2 \qquad (1)$$

Suppose $\gamma_{cc}(P_n) < n - 2$, Let D be a γ_{cc} - set of P_n. Then $V - D$ contains atleast 3 vertices. Let $X = \{x, y, z\}$ such that $V - D = X$.

Case (i) :

Suppose if $< X > = P_3$, then there exist a vertex, v in $V - D$ such that $deg(v) = 2$ in $< X >$. Then v can be dominated by no vertex of D.

Case (ii) :

Suppose $< X > = K_2 \cup K_1$

If $v_1 \in V(K_2)$ or $v_n \in V(K_2)$, then either v_1 or v_n dominated by no vertex of D. If neither $v_1 \in V(K_2)$ nor $v_n \in V(K_2)$, then clearly $< V - X >$ is disconnected. This implies that $< D >$ is disconnected.

Case (iii) :

If $< X > = 3K_1$, then clearly $< D >$ is disconnected.

Since the above cases leads to contradiction,

$$\therefore \quad \gamma_{cc}(P_n) \geq n - 2 \tag{2}$$

From (1) and (2) we get, $\gamma_{cc}(P_n) = n - 2$.

Remark 2.6 : $\gamma_{cc}(P_n) = 2$ if $n = 2, 3$

Theorem 2.7 : *Let C_n be a cycle with n vertices.*

Then

$$\gamma_{cc}(C_n) = \begin{cases} n, \text{ if } n \text{ is odd} \\ n-2, \text{ if } n \text{ is even} \end{cases}.$$

Proof : Let C_n be a cycle with n vertices and $V(C_n) = \{v_1, v_2, v_3, ..., v_n\}$.

Case (i) :

If n is even, then $\chi(C_n) = 2$. Let $D = \{v_1, v_2, v_3, ..., v_{n-1}\}$, then $< D > = P_{n-2}$. Clearly, D is a connected dominating set and $\chi(< D >) = 2 = \chi(C_n)$, since $n \geq 4$. Therefore D is a chromatic connected dominating set. Hence

$$\gamma_{cc}(C_n) \leq n - 2. \tag{3}$$

Suppose $\gamma_{cc}(C_n) < n - 2$. Let D be a γ_{cc} – set of C_n. Then $V - D$ contains atleast 3 vertices. Let $X = \{x, y, z\}$ such that $V - D = X$. Suppose if $< X > = P_3$, then there exists a vertex v in $V - D$, such that $deg(v) = 2$ in $< X >$. Then v can be dominated by no vertex of D. Otherwise, $< V - X >$ is disconnected, since C_n is 2-connected. Hence $< D >$ is disconnected. Hence $\gamma_{cc}(C_n) = n - 2$

Case (ii) :

If n is odd. Since C_n is χ- critical, $V(C_n)$ is a unique chromatic connected dominating set of C_n. Hence $\gamma_{cc}(C_n) = n$.

Remark 2.8 : If G is k-connected then $\gamma_{cc}(G) \geq n - k$.

Theorem 2.9 : *Let K_n be a complete graph with n vertices. Then $\gamma_{cc}(K_n) = n$.*

Proof : Let K_n be a complete graph with n vertices. Since K_n is χ-critical, $\gamma_{cc}(K_n) = n$.

Theorem 2.10 : *Let $K_{m,n}$ be a complete bipartite graph with $m + n$ vertices. Then $\gamma_{cc}(K_{m,n}) = 2$ for all $m, n \geq 1$.*

Proof : Let $K_{m,n}$ be a complete bipartite graph with $m + n$ vertices and $V(K_{m,n}) = \{v_1, v_2, ..., v_m, u_1, u_2, ..., u_n\}$. Clearly $\chi(K_{m,n}) = 2$.

Let $D = \{\{v_i, u_j\}/1 \leq i \leq m, 1 \leq j \leq n\}$. Then $< X > = P_2$. Clearly D is a connected dominating set and $\chi(< D >) = 2 = \chi(K_{m,n})$. Hence D is a chromatic connected dominating set.

$$\therefore \qquad \gamma_{cc}(K_{m,n}) \leq 2 \tag{4}$$

Suppose $\gamma_{cc}(K_{m,n}) < 2$, let D be a chromatic connected dominating set of $K_{m,n}$, $\gamma_{cc}(K_{m,n}) = \gamma_{cc}(|D|) = 1$. Clearly $\chi(< D >) = \chi(K_1) = 1 \neq \chi(K_{m,n})$, which is contradiction. Therefore $\gamma_{cc}(K_{m,n}) = 2$.

Corollary 2.11 : *Let $K_{1,n-1}$ be a star graph with n vertices. Then $\gamma_{cc}(K_{1,n-1})$ for all $n \geq 2$.*

Theorem 2.12 : *Let F_n be a fan with n vertices. Then $\gamma_{ch}(F_n) = 3$, if $n \geq 3$.*

Proof : Let F_n be a fan with n vertices and $V(F_n) = \{v_1, v_2, v_3, ..., v_n\}$ such that $deg(v_1) = n - 1$. Clearly $\chi(F_n) = 3$. Let $D = \{v_1, v_2, v_3\}$. Then D is a connected dominating set with $\chi(< D >) = 3 = \chi(F_n)$. Therefore D is a chromatic connected dominating set. If $\gamma_{cc}(F_n) < 3$ then there is no chromatic connected dominating set D such that $\chi(< D >) = 3$. Hence $\gamma_{cc}(F_n) = 3$.

Theorem 2.13 : *Let W_n be a wheel with n vertices. Then*

$$\gamma_{cc}(W_n) = \begin{cases} n, \text{ if } n \text{ is even} \\ 2, \text{ if } n \text{ is odd} \end{cases}$$

Proof : Let W_n be a wheel with n vertices and $V(W_n) = \{x, v_1, v_2, ..., v_{n-1}\}$ such that $deg(x) = n - 1$.

Case (i) :

If n is even, let $D = \{x, v_i\}$ for some $i = 2, 3, \ldots, n$. Then D is a connected dominating set of W_n, but $\chi(< D >) \neq \chi(W_n) = 4$. Let $D_1 = \{x, v_1, v_2, \ldots, v_{n-1}\}$, then $\chi(< D_1 >) = \chi(W_n) = 4$. Therefore D_1 is a unique chromatic connected dominating set of W_n, then $\gamma_{cc}(W_n) = |D_1| = n$, since W_n is χ-critical, for an even n.

Case (ii) :

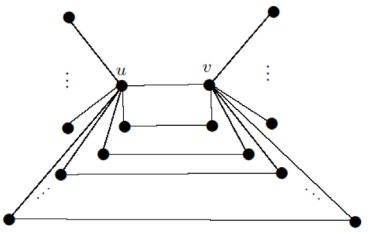
If n is odd. Let $D = \{x\}$ is the connected dominating set of W_n, but $\chi(< D >) \neq \chi(W_n) = 3$. Let $D_1 = \{v_1, v_i, v_j\}$ where $v_i v_j \in E(W_m)$, then clearly $\chi(< D >) = 3 = \chi(W_n)$. Therefore D_1 is a chromatic connected dominating set of W_n. Hence $\gamma_{cc}(W_n) = 3$.

Theorem 2.14 : *Let $D_{r,s}$ be a double star with $r + s$ vertices. Then $\gamma_{cc}(D_{r,s}) = 2$, for all $r, s \geq 1$*

Proof : Let $D_{r,s}$ be a double star with $r + s$ vertices and $V(D_{r,s}) = \{v_1, v_2, \ldots, v_r, u_1, u_2, \ldots, u_s\}$. Clearly $\chi(D_{r,s}) = 2$. Let $D = \{v_1, u_1\}$, since $\deg(v_1) = r$; $\deg(u_1) = s$. $< D > = K_2$. Clearly D is a connected dominating set and $\chi(< D >) = 2 = \chi(D_{r,s})$. Therefore D is a chromatic connected dominating set. Hence $\gamma_{cc}(D_{r,s}) \leq 2$. Suppose $\gamma_{cc}(D_{r,s}) < 2$. Let D is a chromatic connected dominating set of $D_{r,s}$. Therefore $\gamma_{cc}(D_{r,s}) = 1$. Clearly $\chi(< D >) = K_1 = 1 \neq \chi(D_{r,s})$. Which is contradiction. $\therefore \gamma_{cc}(D_{r,s}) \geq 2$. Hence $\gamma_{cc}(D_{r,s}) = 2$.

Theorem 2.15 : *For any graph G, $\gamma_{cc}(G) = 2$ if and only if G belongs to the family $\mathcal{A}$.*

Proof : Let G belongs to the family $\mathcal{A}$. Let $D = \{u, v\}$. Since $< D > = K_2$, D is a connected dominating set and $\chi(< D >) = 2 = \chi(G)$. Then D is a chromatic connected dominating set of G. Hence $\gamma_{cc}(G) = 2$. Conversely, Let G be a chromatic connected dominating graph with $\gamma_{cc}(G) = 2$. Let $D = \{u, v\}$ be a γ_{cc}- set of G. By the definition, $uv \in E(G)$.

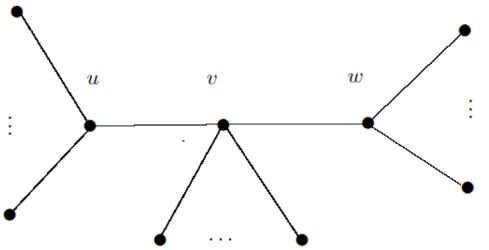


Graph in Family $\mathcal{A}$

Let $N(u) = \{v, u_1, u_2, ..., u_n\}$ and $N(v) = \{u, v_1, v_2, ..., v_m\}$ Then $V(G) = N(u) \cup N(v)$. Otherwise if there exists a vertex w in $V(G) - (N(u) \cup V(v))$ then w can be dominated by no vertex in D. Also, no two vertices v, w in $V(u)$ are adjacent to each other. Otherwise $V(<\{u, v, w\}>)$ form a triangle. This implies the contradiction that $\gamma_{cc}(G) \geq 3$. Similarly, no two vertices in $N(v)$ are adjacent to each other. But the vertices in $N(u) - \{v\}$ may be adjacent to the vertex in $V(v)$ $\{u\}$. The above discussions shows that G belongs to the family $\mathcal{A}$.

Theorem 2.16 : *For any tree T, $\gamma_{cc}(T) = 3$ if and only if T belongs to the family $\mathcal{T}$.*

Proof: Let T be a tree belongs to $\mathcal{T}$. Let $D = \{u, v, w\}$. Since $< D > = P_3$ and dominates all the vertices of T, D is a connected dominating set T and $\chi(< D >) = 2 = \chi(T)$. Then D is a chromatic connected dominating set of T. Hence $\gamma_{cc}(T) = 3$.



Conversely, let T be a tree such that $\gamma_{cc}(T) = 3$. Let $D = \{u, v, w\}$ be a γ_{cc} - set of T. Then by the definition, $< D >$ is connected and $\chi(< D >) = \chi(T) = 2$. Hence there exists exactly 2 edges in $< D >$. Without loss of generality, let us assume that $uv \in E(G)$; $vu \in E(T)$ and $uw \notin E(T)$. Hence $< D > = P_3$. Let $N(u) = \{v, v_1, v_2, ..., v_p\}$; $N(v) = \{u, w, v_1, v_2, ..., v_q\}$ and $N(w) = \{v, w_1, w_2, ..., w_r\}$ where p, q and r are the non-negative integers. Then it is clear that no two vertices in $N(u)$ are adjacent to each other. Otherwise, $G(<u,v, w>)$ form a triangle. This implies the contradiction to T is a tree. Similarly, no two vertices in $V(v)$ and $V(w)$ are adjacent to each other. It is clear that $V(T) = N(u) \cup N(v) \cup N(w)$. Otherwise if there exists a vertex x in $V(T) - (N(u) \cap N(v) \cap N(w))$ then x can be dominated by no vertex of D. Let x, y be two vertices of u, v and w. If $xy \in E(T)$ then T contains the cycle C_4. Which is contradiction to T is a tree. Therefore $xy \notin E(T)$. The above discussions shows that T belongs to the family $\mathcal{T}$.

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GALLAI-TYPE THEOREMS ON INDEPENDENCE AND IRREDUNDANCE IN GALLAI FUZZY GRAPHS

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Abstract: The Gallai fuzzy graph Γ(G) of a fuzzy graph G has the fuzzy edges of G as its fuzzy vertices and two distinct fuzzy edges of G are fuzzy incident in G, but do not span a fuzzy triangle in G. The Gallai fuzzy graphs are fuzzy spanning Gallai sub graphs of the well-known Class of fuzzy line graphs. Let Γ(Γ(G)) and β(Γ(G)) denote the maximum fuzzy cardinality of a fuzzy dominating set of a Gallai fuzzy graph Γ(G)= (σ, μ) with n fuzzy vertices and minimum fuzzy degree δ(Γ(G)), Γ(Γ(G))≤ n- δ(Γ(G)), β(Γ(G)) ≤ n- δ(Γ(G)).In this paper, we show for the upper fuzzy irredundance number, IR(Γ(G)): For a Gallai fuzzy graph Γ(G) with n fuzzy vertices and minimum fuzzy degree δ(Γ(G)), IR (Γ(G)) ≤ n- δ(Γ(G)).Characterizations are given for classes of Gallai fuzzy graphs which achieve this upper bound for the upper fuzzy irredundance, upper fuzzy domination and fuzzy independence numbers of a Gallai fuzzy graph.

Key words: Gallai fuzzy graph, Gallai-type theorems, On Domination,independent and irredundance

AMS Classification: 03E72, 05C99**.**

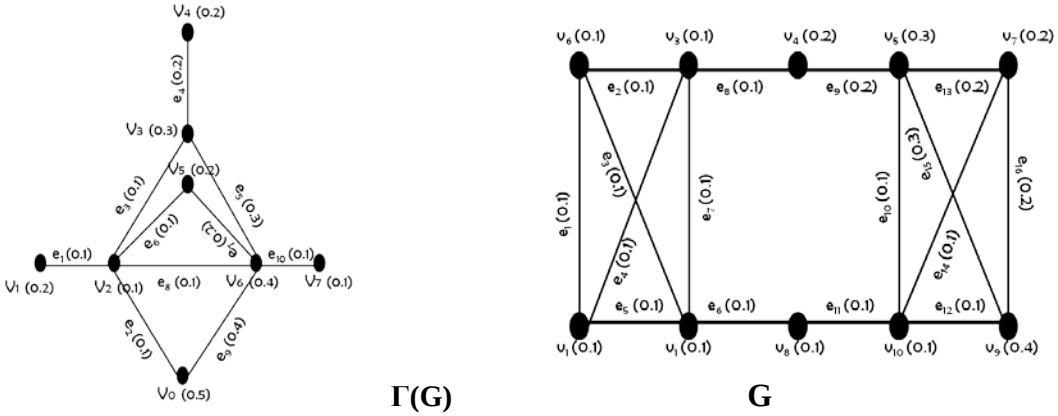
1. Introduction

The study of dominating sets in graphs was begun by Ore and Berge, the domination number, independent domination number are introduced by Cockayne and Hedetniemi. In 1965, L.A. Zadeh [1] introduced a mathematical frame work to explain the concepts of uncertainty in real life through are publication of a seminal paper. In 1975, A. Rosenfeld [2] introduced the notation of fuzzy graph theoretic concept such as paths, cycles and connectedness. In 1996, Van Bang Le [3] was discussed the paper of Gallai graphs and anti-Gallai graphs. Also S. Aparna Lakshmanan and S.B. Rao [4] were discussed in this paper. A. Somasundram and S. Somasundram [5] discussed domination in fuzzy graphs. A. Nagoorgani and

P.Vadivel [6] dealt with domination, independence and irredundance numbers. In this paper we discussed about Gallai- type theorems on domination, independence and irredundance in Gallai fuzzy graphs based on the concept of Gallai type theorems and domination parameters by GaylaS.Domkeet.al. [7]. In [8], our earlier work have discussed the concept of Gallai Fuzzy Graphs on Domination parameters. In this manuscript, we investigate the Gallai-type theorems in Gallai fuzzy graph involving upper fuzzy domination parameters combined with minimum fuzzy degree and establish the relationship with other parameters which is also investigated.

2. Preliminaries.

Definition 2.1 [4] A fuzzy graph G and its Gallai fuzzy graph Γ(G) as shown below:



Γ(G) **G**

A fuzzy graph with G as the underlying set is a finite non-empty unordered pair of G= (σ, μ), where σ : V →[0,1] is a fuzzy subset, μ: E→[0,1] is a fuzzy relation on the fuzzy subset σ such that μ(u, v) ≤ σ(u) ∧ σ(v) for all u, v Є V where ∧ and ∨ stands for minimum and maximum. The underlying crisp fuzzy graph of G=(σ, μ) is denoted by G^*=(V, E), where V ={ u ϵ V : σ(u) > 0}andE = {(u, v)ϵ VxV : μ(u, v) > 0 },the fuzzy order P and fuzzy size of the fuzzy graph G=(σ, μ) are defined by p = $\Sigma_{v \in V}$ σ(u) and q =$\Sigma_{u, v \in E}$ μ(u, v). Each pair μ = u, v of fuzzy vertices in σ is a fuzzy edge of G and μ is said to join u and v are fuzzy adjacent vertices, fuzzy vertex u and fuzzy edge μ are fuzzy incident with each other as are σ and μ if two distinct fuzzy edges are incident with a common fuzzy vertex, then they are called fuzzy adjacent edges. A fuzzy edge e = uv of a fuzzy graph is an fuzzy edge if μ(u, v) = σ(u) ∧ (v). N (u) ={v ∈ V / μ (u, v) = σ(u) ∧ σ(v) } is called the open fuzzy neighborhood of u and N[u]= N(u)∪{u}is the closed fuzzy neighborhood of u.

Definition 2.2 *Let* G be a fuzzy graph and u be a fuzzy vertex in G then there exists a fuzzy vertex v such that (u,v) is a fuzzy edge then we say that u dominates v.

Definition 2.3 Let G=(σ, μ) be a fuzzy graph. A subset D of V is said to be fuzzy dominating set of G if for every v ∈ V-D, there exists u∈ D such that u dominates v.

Definition 2.4 A fuzzy dominating set D of a fuzzy graph G is called minimal fuzzy dominating set of G, if for every fuzzy vertex v ∈ D,D-{v} is not a fuzzy dominating set.

Definition 2.5 Two uzzy vertices in a fuzzy graph G are said to be fuzzy independent if there is no fuzzy edge between them. A subset S of V is said to be fuzzy independent set of G if every two fuzzy vertices of S are fuzzy independent.

Definition 2.6 A fuzzy independent set S of G is said to be maximal fuzzy independent, if for every fuzzy vertex v∈ V-S, the set S∪{v} is not fuzzy independent.

3. Fuzzy independent and irredundant in Gallai sets.

Definition 3.1 Let Γ(G) be Gallai fuzzy graph and S be a set of fuzzy vertices. A fuzzy vertex v is said to be fuzzy private neighbour or private neighbour of u ∈S with respect to S if N[v]∩S={u}. Furthermore, we define fuzzy private neighborhood of u ∈ S with respect to S to be PN[u,S]={v:N[v]∩S={u}}.Stated in other words PN[u,S]=N[u]-N[S-{u}].If u ∈ PN[u,S], then u is an isolate fuzzy vertex in <S>.If is also stated that u is its own fuzzy private neighbor.

Definition 3.2 A Gallai fuzzy set of fuzzy vertices S is said to be Gallai fuzzy irredundant set if PN [u,S]≠ φ for everyfuzzy vertex in S.

Definition 3.3 A Gallai fuzzy irredundant set S is a maximal fuzzy irredundant if for every fuzzy vertex u ∈V-S, the set S∪{u} is not Gallai fuzzy irredundant set, which means that there exists at least one fuzzy vertex w ∈ S∪{u} which does not have fuzzy private neighbor.

Definition 3.4 Minimum fuzzy cardinality among all minimal fuzzy dominating sets in Γ(G) is called fuzzy domination number of Γ(G) and is denoted by γ(Γ(G)).

Definition 3.5 Maximum fuzzy cardinality among all minimal fuzzy dominating sets in Γ(G) is called upper fuzzy domination number of Γ(G) and is denoted by Γ(Γ(G)).
A fuzzy dominating set D of a Gallai fuzzy graph Γ(G) is a minimum fuzzy dominating set if $|D|$= γ(Γ(G)).

Definition 3.6 The maximum fuzzy cardinality among all maximal fuzzy independent set is called fuzzy independent number of Γ(G) and is denoted by β(Γ(G)).

Definition 3.7 The minimum fuzzy cardinality among all maximal fuzzy independent set is called independent fuzzy domination number of Γ(G) and is denoted by i (Γ(G)).

Definition 3.8 Minimum fuzzy cardinality among all maximal fuzzy irredundant set is called fuzzy irredundance number and is denoted by ir(Γ(G)).

Definition 3.9 Maximum fuzzy cardinality among all maximal Gallai fuzzy irredundant set is called upper fuzzy irredundance number and is denoted by IR(Γ(G)).

Definition 3.10 A property P of a Gallai fuzzy graph Γ(G) is hereditary if every Gallai fuzzy sub graph of Γ(G) also has this property. A fuzzyset S of fuzzy vertices of Γ (G) is called a fuzzy P-set if the induced Gallai fuzzy sub graph [S] has property P. A fuzzy set is called fuzzy $\bar{P}$- set, if the fuzzy set does not satisfy the property P. A property P is super hereditary if whenever a fuzzy set S has property P, so does every fuzzy super set $S_1^{\prime} \supset$ S.

A fuzzy P-set S is a maximal fuzzy P-set if every proper fuzzy super set $S_1^{\prime} \supset$ S is a fuzzy $\bar{P}$ – set. A fuzzy P-set S is 1- maximal fuzzy P-set if for every fuzzy vertex u ∈ V-S, S∪ $\{u\}$ is a fuzzy $\bar{P}$ – set.A fuzzy P-set S is a minimal fuzzy P-set if every proper fuzzy subset $S_1^{\prime} \subset$ S is a fuzzy $\bar{P}$ – set.A fuzzy P-set S is 1- minimal fuzzy P-set if for every fuzzy vertex
v∈ S, S−$\{v\}$ is a fuzzy$\bar{P}$ – set.

Proposition 3.11 Let Γ(G) =(σ,μ) be a Gallai fuzzy graph. Let P be hereditary property. Then a set D is a maximal fuzzy P-set if and only if D is 1-maximal fuzzy P-set.

Proposition 3.12 Let Γ(G) =(σ,μ) be a Gallai fuzzy graph. Let P be super hereditary property. Then a set D is a minimal fuzzy P-set if and only if D is 1-minimal fuzzy P-set.

Proposition 3.13 If D is independent fuzzy dominating set, then D is both a minimal fuzzy independent set and a maximal fuzzy independent set. Conversely, if D is a maximal fuzzy independent set then D is an independent fuzzy dominating set.

Definition3.14T he Gallai fuzzy graph Γ(G) of a fuzzy graph G has the fuzzy edges of G as its fuzzy vertices and two distinct fuzzy edges of G are fuzzy incident in G, but do not span a fuzzy triangle in G. The line fuzzy graph L(G) of a fuzzy graph G has the fuzzy edges of G as its fuzzy vertices and two distinct fuzzy edges of G are adjacent in L(G) if they are fuzzy incident in G.

Example 3.15 A fuzzy graph G and its Gallai fuzzy graph Γ(G) as shown below

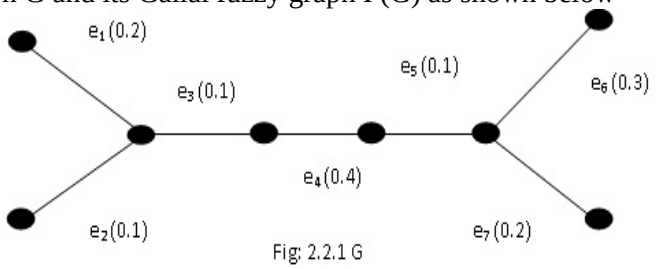


Fig: 2.2.1 G

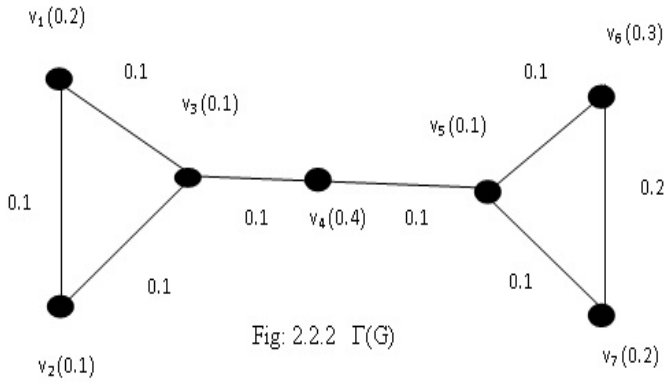


Fig: 2.2.2 Γ(G)

Theorem: 3.16 *(cockayne)* For any Gallai fuzzy graph Γ(G), we have ir(Γ(G)) ≤ γ(Γ(G)) ≤ i(Γ(G)) ≤ β(Γ(G)) ≤ Γ(Γ(G)) ≤ IR(Γ(G)).

Proof:First we have to prove that every minimal fuzzy dominating set in a maximal fuzzy irredundant set. Let D be a γ-set of Γ(Γ(G)), then for every fuzzy vertex v ∈ D, there exist a fuzzy vertex w ∈ V-(D-{v}), which is not fuzzy dominated by D-{v}. This implies that every fuzzy vertex in D has a fuzzy private neighbour. Then D is a fuzzy irredundant set.

Now, we have to prove that D is a maximal fuzzy irredundant set of Γ(G). The property of a set D being a fuzzy irredundant is a hereditary property. By Proposition 3.8, if P is hereditary property of a Gallai fuzzy graph Γ (G), then a set D is a maximal fuzzy P-set if and only if D is a 1-maximal fuzzy P-set. It is enough to prove that D is maximal fuzzy irredundant set. Suppose D is not a maximal fuzzy irredundant set. Then, there exist a fuzzy vertex u ∈ V-D for which D∪{u} is fuzzy irredundant. This means, in particular that PN[u, D∪{u}]≠ φ.That is there exist at least one fuzzy vertex W, which is a fuzzy private neighbor of u with request to D∪{u}.This implies no fuzzy vertex in D is fuzzy adjacent to W. Then D is not a fuzzy

dominating set, which is a contradiction to the assumption that D is a fuzzy dominating set. Hence D is a maximal fuzzy irredundant set. Therefore, $(\Gamma(G)) \leq \gamma(\Gamma(G))$.

Now, to prove the second inequality, we have to show that every maximal fuzzy independent set in a Gallai fuzzy graph$\Gamma(G)$ is a minimal fuzzy dominating set. Let D be a maximal fuzzy independent set in $\Gamma(G)$.

By proposition 3.10, a fuzzy independent set is maximal fuzzy independent if and only if it is fuzzy independent and fuzzy dominating. Let D be a maximal fuzzy independent set.

By the definition of maximal fuzzy independent set, for every fuzzy vertex u ∈ V-D, there is a fuzzy vertex v∈ D such that u is fuzzy adjacent to V. This implies that D is a fuzzy dominating set.

Now, we have to prove that D is a minimal fuzzy dominating set. By proposition 3.9, if P is super hereditary property of a Gallai fuzzy graph$\Gamma(G)$, then a set D is a 1-minimal fuzzy P-set if and only if D is minimal fuzzy P-set. A set being a fuzzy dominating set has a super hereditary property.It is enough to prove that, the set D is 1-minimal fuzzy P-set. A fuzzy dominating set D is a minimal fuzzy dominating set if for every fuzzy vertex v ∈ D, the set D-{v}is not a fuzzy dominating set.

Suppose that D is not a minimal fuzzy dominating set. Then there exist at least one fuzzy vertex v ∈ D for which D-{v} is a fuzzy dominating set. But if D-{v} dominates V-(D-{v}), then at least one fuzzy vertex in D-{v} fuzzy adjacent to v. This is a contradiction to the fact that D is fuzzy independent. Then D is a minimal fuzzy dominating set.

Therefore $\gamma(\Gamma(G)) \leq$ i($\Gamma(G)$).

By definition, i($\Gamma(G)$) is a minimum fuzzy cardinality of maximal fuzzy independent sets of $\Gamma(G)$and$\beta(\Gamma(G))$ is the maximum fuzzy cardinality of maximal fuzzy independent sets of$\Gamma(G)$. Then i($\Gamma(G)$) $\leq \beta(\Gamma(G))$.

Let S be a minimal fuzzy dominating set with maximum fuzzy cardinality. That is, $|S|$= $\Gamma(\Gamma(G))$. To prove that $\beta(\Gamma(G)) \leq \Gamma(\Gamma(G))$, we have to prove that S maximal fuzzy independent set with maximal fuzzy cardinality.

Suppose S is not a maximal fuzzy independent set. Then there is a fuzzy vertex W ∈ S-V such that S ∪{w} is a fuzzy independent set. This implies that W is not fuzzy adjacent to any fuzzy vertex in S, then S is not a fuzzy dominating set of $\Gamma(G)$, a contradiction. Therefore, S is a maximal fuzzy independent set. Then $\beta(\Gamma(G)) \leq \Gamma(\Gamma(G))$.

Let S be a maximal fuzzy irredundant set with maximum fuzzy cardinality. That is $|S|$= IR($\Gamma(G)$). To prove that $\Gamma(\Gamma(G)) \leq$ IR($\Gamma(G)$), we have to prove that S is a minimal fuzzy dominating set with maximum fuzzy cardinality. Suppose S is not a minimal fuzzy dominating

Set. Then S-{v} is a fuzzy dominating set. Then S-{v} is a fuzzy dominating set of $\Gamma(G)$,for some v in S. Then v has no fuzzy private neighbour. This implies that S is not an fuzzy irredundant set. Therefore, S is a minimal fuzzy dominating set. Then $\Gamma(\Gamma(G)) \leq$ IR($\Gamma(G)$).

Hence ir($\Gamma(G)$) $\leq \gamma(\Gamma(G)) \leq$ i($\Gamma(G)$) $\leq \beta(\Gamma(G)) \leq \Gamma(\Gamma(G)) \leq$ IR($\Gamma(G)$).

Example: 3.17 The Gallai fuzzy sets S_1= {v_3,v_5}, S_2 ={v_1,v_5}, S_3={v_3,v_6}, S_4={v_2,v_5,}, S_5={v_3,v_7}, S_6={v_1,v_4,v_6}, S_7={v_2,v_4,v_7} are maximal fuzzy irredundant sets in the Gallai fuzzy graph given in fig: 2.2.2$\Gamma(G)$. Here PN[v_3,S_1]={ v_1,v_2,v_3}, PN[v_5,S_1]={ v_5,v_6,v_7}, PN[v_1,S_6]={ v_1,v_2}, PN[v_4,S_6]={ v_4}, PN[v_6,S_6]={ v_6,v_7}.For this Gallai fuzzy graph with fuzzy domination numbers, independent fuzzy domination numbers and fuzzy irredundant numbers are as follows $\gamma(\Gamma(G))$ = 0.2,$i(\Gamma(G))$ = 0.2, $\Gamma(\Gamma(G))$ = 0.9 , $\beta(\Gamma(G))$ = 0.9 ir($\Gamma(G)$) = 0.2, and IR($\Gamma(G)$) = 0.9.

4. The upper fuzzy domination parameters and minimum fuzzy degree.

The upper fuzzy domination parameters, $\beta(\Gamma(G))$, $\Gamma(\Gamma(G))$ and IR($\Gamma(G)$) will be combined with minimum fuzzy degree for Gallai-type results of Gallai fuzzy graph.

Theorem 4.1 For any Gallai fuzzy graph$\Gamma(G)$, IR($\Gamma(G)$) + $\delta(\Gamma(G)) \leq$ n.

Proof: Let S be a maximal fuzzy irredundant set of size IR($\Gamma(G)$) and let u∈ S. Since S is fuzzy irredundant, there is a fuzzy vertex u such that u∈ N[u]-N[S-{u}].

We consider two cases,

Case (i) :v= u. Then u is not fuzzy adjacent to any fuzzy vertex in S, and must have at least δ(Γ(G)) fuzzy neighbours in V-S. Thus n- IR(Γ(G)) = $|V - S| \geq$ δ(Γ(G)) and IR((Γ(G)) + δ(Γ(G)) ≤ n.
Case (ii): v≠ u. By the choice of v, v does not belongs to S and N(v) ∩ S = {u}. Then N[v]-{u} is a subset of V-S, so that n-IR(Γ(G)) = $|V - S| \geq |N[v] - \{u\}| \geq$ δ(Γ(G)).
This implies, - IR(Γ(G)) ≥ δ(Γ(G)) – n.

IR(Γ(G)) ≤ - δ(Γ(G)) + n

IR(Γ(G)) +δ(Γ(G)) ≤ n

Using theorem 4.1 and 3.11, we get the following corollary:

Corollary 4.2 For any Gallai fuzzy graph Γ(G), Γ(Γ(G)) + δ(Γ(G)) ≤ n. and $\beta(\Gamma(G))$ + δ(Γ(G)) ≤ n.

Proof:

By Theorem 3.11, Γ(Γ(G)) ≤ IR(Γ(G))

Γ(Γ(G)) + δ(Γ(G)) ≤ IR(Γ(G)) + δ(Γ(G))

By Theorem 4.1, IR(Γ(G)) + δ(Γ(G)) ≤ n.

Therefore Γ(Γ(G)) + δ(Γ(G)) ≤ n.

By Theorem 3.11, $\beta(\Gamma(G))$ ≤ IR(Γ(G))

Then$\beta(\Gamma(G))$ + δ(Γ(G)) ≤ IR(Γ(G)) + δ(Γ(G))

By Theorem 4.1, IR(Γ(G)) + δ(Γ(G)) ≤ n.

Therefore$\beta(\Gamma(G))$ + δ(Γ(G)) ≤ n.

We will first consider Gallai fuzzy graphs for which$\beta(\Gamma(G))$ + δ(Γ(G)) = n.

Theorem 4.3 Let Γ(G) be a connected Gallai fuzzy graph and let I be a maximal fuzzy independent set of Γ(G) such that $|I| = \beta(\Gamma(G))$ .Then $\beta(\Gamma(G))$ + δ(Γ(G)) = n if and only if for each u∈ I, we have fuzzy degree (u) = δ(Γ(G)) and V- N(u) is an fuzzy independent.

Proof: Suppose that$\beta(\Gamma(G))$ + δ(Γ(G)) = n.Let u∈ I,

Then I is a subset or equal toV-N[u]. So $\beta(\Gamma(G) = |I| \leq$ n - $|N(u)|$

≤ n - δ(Γ(G)) = $\beta(\Gamma(G))$.

Since $|N(u)| \geq$ δ(Γ(G)), implies that - $|N(u)| \leq$ - δ(Γ(G)).

Thus $|N(u)|$ = δ(Γ(G)), and $|V - N(u)| = |I|$

So V- N(u) is a fuzzy independent set.

Now, suppose that for each u∈ I, fuzzy degree (u) = δ(Γ(G)) and V-N(u) is a fuzzy independent set. Then $\beta(\Gamma(G)) \geq |V - N(u)|$.

$\beta(\Gamma(G)) \geq$ n - δ(Γ(G)).

$\beta(\Gamma(G))$ + δ(Γ(G)) ≥ n.

By corollary 4.2, $\beta(\Gamma(G))$ + δ(Γ(G)) ≤ n.

Therefore $\beta(\Gamma(G)) = n - \delta(\Gamma(G))$.

Theorem 4.4 For any Gallai fuzzy graphΓ(G), IR(Γ(G)) +δ(Γ(G)) = n if and only if Γ(Γ(G)) + δ(Γ(G)) = n.

Proof: Let Γ(G) be a Gallai fuzzy graph with Γ(Γ(G)) + δ(Γ(G)) = n.

From the fact that Γ(Γ(G)) ≤ IR(Γ(G)), we have Γ(Γ(G)) + δ(Γ(G)) ≤ IR(Γ(G)) + δ(Γ(G)).

By Theorem 4.1, we have,IR(Γ(G)) + δ(Γ(G)) ≤ n.

Then n = Γ(Γ(G)) + δ(Γ(G)) ≤ IR(Γ(G)) + δ(Γ(G)) ≤ n. Hence IR(Γ(G)) + δ(Γ(G)) = n.

Conversely,

Suppose that Γ(G) is a Gallai fuzzy graph with IR(Γ(G)) + δ(Γ(G)) = n.

Let S be a maximal fuzzy irredundant set for Γ(G) with |S| = IR(Γ(G)) . We will show that S is fuzzy dominating and since S is fuzzy irredundant, it will be a minimal fuzzy dominating set. So suppose that S is not fuzzy dominating. Then there is a w ∈ V- S such that w is not fuzzy adjacent to any fuzzy vertex of S. Then N[w] is a subset or equal to V-S. But |N[w]|≥ δ(Γ(G)) +1,

Which implies δ(Γ(G)) + 1 ≤ $|N[w]| \leq |V - S|$.

$\delta(\Gamma(G)) + 1 = n - IR(\Gamma(G))$ and so $IR(\Gamma(G)) + \delta(\Gamma(G)) \leq n - 1$, a contradiction. Thus S is a fuzzy dominating set. Since $\Gamma(\Gamma(G)) \geq |S| = IR(\Gamma(G)) \geq \Gamma(\Gamma(G))$,
we must have $IR(\Gamma(G)) = \Gamma(\Gamma(G))$.Therefore $\Gamma(\Gamma(G)) + \delta(\Gamma(G)) = n$.

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Gel Combustion Synthesis and Characterization of ZnO/NiO Nanocomposite for Supercapacitor Application

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Abstract*: Energy demand is a major concern around the globe of the world. Electrochemical supercapacitors are one among various alternative and green energy devices. The performances of supercapacitors depend mainly on the enhanced properties of electrode materials. In the present work, ZnO/NiO nanocomposite (NCs) was synthesized by a simple and facile citrate-based gel combustion procedure. The crystal structure and phase identification, surface morphology and functional groups of the samples were analyzed by X-ray diffraction (XRD) pattern, scanning electron microscope (SEM) and Fourier-transform infrared spectroscopy (FTIR), respectively. X-ray Diffraction pattern is observed that the crystalline peaks are broader and confirmed nanoparticles. The mean size of the particle is found to be ~25 nm. The prepared sample is analyzed an electrochemical studies such as cyclic Voltammetry, charge discharge and electrochemical impedance spectrum, respectively. The maximum specific capacitance (Scp) is 450 Fg-1 at 0.5 mA/cm2.

***Keywords*: ZnO/NiO, gel combustion method, supercapacitors, XRD, FTIR, SEM.**

I. INTRODUCTION

In recent decades supercapacitors based devices have paid must attention in energy storage applications. The supercapacitors used in rapid charge-discharge rate, large operation temperature range and more cycle stability as compared with secondary battery [1-3]. Low Scp and low energy density degrades such potential applications of supercapacitors in energy storage area. In the search of new electrode materials, lot of researches has been implemented to overcome the drawbacks. Recently, transition metal oxides are widely used in electrode material for supercapacitors. In supercapacitors, the transition metal oxides attribute fast faradaic redox reactions [4-6]. Supercapacitors are describe as two kinds of effect such as pseudo capacitor and electric double layer capacitor. Based on the electrochemical reactions supercapacitors are divided into two types one is electrical double layer capacitor (EDLC) and another one is pseudocapacitor (PSc). Compared with EDLC, the transition metal oxides based PSc has better Scp and elevated energy density [7, 8]. There are many reports are available for binary metal oxides based supercapacitors due to their wealthier redox reactions and synergistic effects with different metal ions.

The pure NiO NPs possesses an excellent electrochemical properties which are including its remarkable theoretical supercapacitance value (2584 Fg^{-1}), inexpensiveness and good chemical stability [9-11]. However, the low surface area of a electrode material leads to less capacitance value than the theoretical one. Preparing of nanomaterials ought to be advanced in light of the fact that it will positively affect the materials electrochemical property [12]. By comprises the different kind of nanocrystalline transition metal oxides, improving of capacitance can be achieved through these structure of nanocomposite based electrode. Zinc oxide (ZnO), a significant semiconductor material is mostly preferred due its particular highlights. In addition, ZnO utilizing as a terminal substance, has evident focal points for supercapacitors. These properties incorporate high electron versatility, electrochemical stability, capacity and lower expensive [13- 15]. Encouragingly, a nanocomposite electrode which means by combined NiO and ZnO, has incredible value of voltage. Because of modification by ZnO, the capacitance value is also enhanced in the nanocomposite. Effective synthesis of nanocomposite is also one of the reason for the improvement. Hence, in this approach, the nanocomposite of transition metal oxides are developed as electrode materials for electrochemical storage device applications. [16, 17].

In the present work deals, the ZnO/NiO nanocomposite (NCs) was prepared by a Gel-combustion method with subsequent calcination. The structural, spectroscopic, surface morphological and electrochemical properties are examined through XRD, FTIR, SEM and cyclic voltametry and discharge with charge analysis, respectively.

II. EXPERIMENTAL SECTION

A. Experimental methods and characterization techniques

Citrate-based gel combustion method is adopted to obtain ZnO/NiO NCs. Analytical grade of zinc nitrate, nickel nitrate are

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acting as starting materials and citric acid is used as a chelating agent in gel combustion method. Appropriate amount of zinc nitrate, nickel nitrate and citric acid (2:6 molar ratio) are dissolved in double distilled (DD) water in separate beakers. Initially zinc nitrate and nickel nitrate precursors are dissolved in DD water. The citric acid solution was poured into above solution drop-wise at 80 ^{o}C. Later, the temperature of the above mixture was increased to 130 ^{o}C. Then, solution was kept overnight to convert into gel and dried gel was formed. The dried gel is calcined at 500 ^{o}C for 3 hrs. The entire reaction has shown in scheme.1.The samples are well characterized using XRD, FTIR and SEM analysis.

Scheme.1. Preparation of ZnO/NiO NCs

III. RESULTS AND DISCUSSION

A. Powder X-Ray Diffraction Analysis

X – Ray diffractogram for ZnO/NiO NCs is displayed in Fig. 1. In the crystal structure, ZnO and NiO are hexagonal and cubic geometry, respectively. Fig.1 diffractogram of the calcinated sample, characteristic diffraction peaks could shows X – Ray every part of the be matched to hexagonal ZnO (JCPDS No. 361451) and cubic NiO (JCPDS No. 897130) [18]. No diffraction peaks belonging to impurities could be found, indicating that the precursor was completely transformed into ZnO/NiO mixed metal oxides. The size of the crystallite is estimated by Scherrer's equation. The estimated average crystallite size is obtained about ~25 nm.

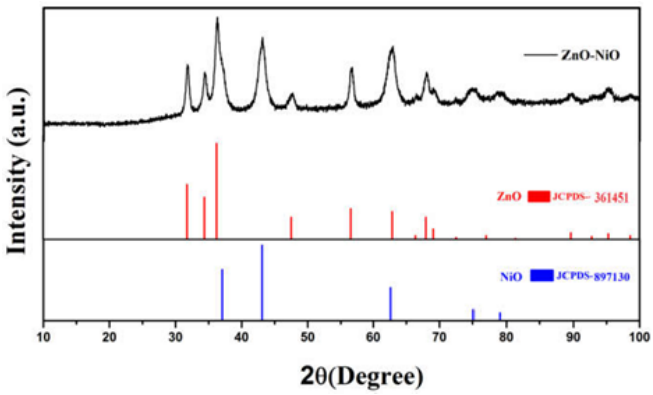

Fig. 1.XRD pattern of ZnO/NiO NCs

B. FTIR

The Fig.2, described FTIR spectra of ZnO/NiO nanocomposites, heated for 3h at 500 ^{o}C. Four new IR bands are found to be observed at 1602, 1478, 1100 and 417 cm-1[19]. A broad band around 3346 cm^{-1} attributes the stretching mode of OH group which is contributed by adsorbed water molecules. The peak around 2367 cm^{-1} was observed indicating CO2 group. Band formed at 1602 cm^{-1} can be contributed due to OH group in the sample. IR peak at 1478 cm^{-1} is obtained corresponds to C=O bond in asymmetric stretching mode and at 1100 cm^{-1} are obtained due to C-O bonding. IR band at 417 cm^{-1} is observed indexing the confirmation of the occurrence of crystalline Ni–O bond formation [20].

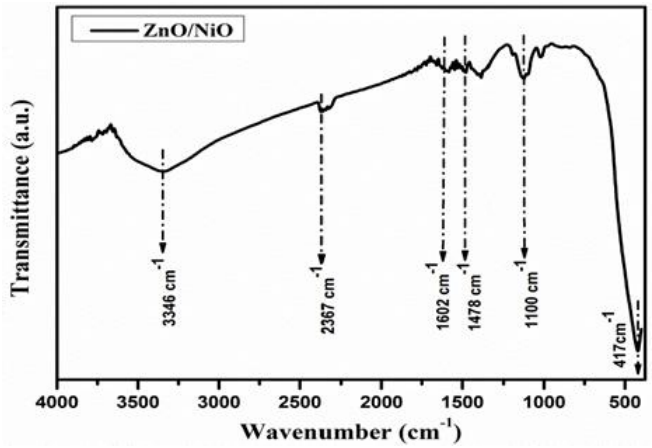

Fig. 2.FTIR spectrum of ZnO/NiO NCs

C. SEM

The morphological analysis of the ZnO/NiO composite is examined using SEM. The different magnification of SEM images of the ZnO/NiO composite is presented as shown in Fig. 3. (a and b). It is illustrated that the sample is composed nano spheres like agglomerated particles. The average size of the particles is measured as ~61 nm.

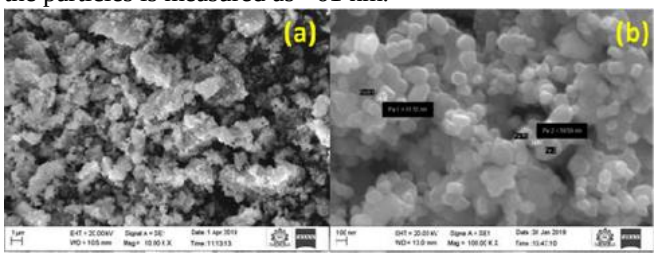

Fig. 3.(a, b) SEM images of ZnO/NiO NCs

D. Electrochemical characterizations

The microstructural analysis and ion transport behavior of the ZnO/NiO nanospheres is analyzed through cyclic voltammeteric studies. It is investigated by GCD, CV and EIS with KOH solution of electrolyte in three-electrode cell setup. The curves described in Cyclic voltammetry at the range of scan rates between 5mVs^{-1} and 100 mVs^{-1} which characteristically pseudocapacitive behaviour in the way of delineating redox peaks. It is ascribed in Fig. 4a. Because of the scan rate increments to the higher rates, cathodic curve drop down towards lower potential whereas the anodic peak shifts towards higher voltage proposing a superior electrochemical reversibility [21, 22]. The determined Csp values are 111, 124, 136, 166, 146 and 198 F g^{-1} for 100, 50, 25, 10 and 5 mV s^{-1} respectively. In Fig. 4. b., the GCD plot of the material demonstrates the curves applied in the range of 4 to 0.5 mA/cm^{2} current densities. The GCD graphs are profoundly regular in nature proposing the great capacitive profile. The deliberate Scp esteems from the

GCD curves are 450, 390, 312, 270 and 174 F g^{-1} for current densities 0.5, 1, 2, 3 and 4 mA/cm^2, individually. By the electrochemical investigation, the outcome is presumed that the expansion of current densities and scan rate diminish the execution of capacitance effect in nanocomposite materials. At the intension of lower current densities, the ionic charge carriers has efficient time to diffuse the layer in the interfacial region of electrode and electrolyte which provide the illustration of high capacitance value. Then again, at high current density the flow of ions is very quick and the accumulation of ions is very minimum on the surface of the working electrode henceforth the S_{cp} value reduces [23].

The EIS study of the ZnO/NiO nanospheres (Fig.4.4d) shows a semi-circle at the high frequencies with inclined spike with Y-axis obtained at lower frequencies, displaying great electrochemical behavior in modified working electrode. The appearance of small semi-circle indicates the miniature of charge transfer resistance which is remarkable for quick ion charge exchange and prompted predominant [22, 24]. The retentively of S_{cp} study for different cyclic performance is also much interesting property to analyze the cycle time of working electrode.

From fig.4.c displays cyclic durability for ZnO/NiO NCs were analyzed for 5000 cycles with applied current density of 3mA/cm2. After 5000 cycles, the retention of the ZnO/NiO NCs is about 74.7 % was observed. In the present study the retention graph reveals the long life of the ZnO/NiO NCs.

The electrochemical reaction mechanism of the prepared ZnO/NiO nanospheres as follows [24]

$ZnO + OH^- \leftrightarrow ZnOOH + e^-$ (1)

$NiO + OH^- \leftrightarrow NiOOH + e^-$ (2)

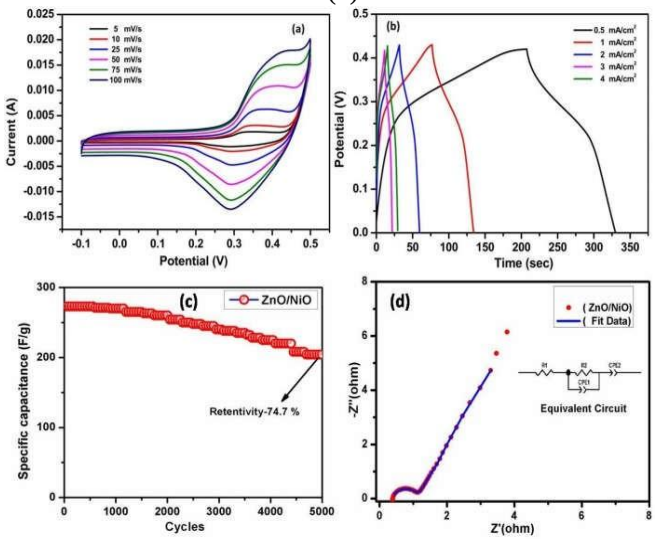

Fig. 4.(a) Cyclic Voltammetry curve (b) Charge discharge curve (c) Cyclic stability graph (d) Electrochemical impedance spectrum of ZnO/NiO NCs

IV. CONCLUSION

ZnO/NiO nanocomposite has been successfully synthesized by Gel-combustion process. The XRD revealed the mixed phases of ZnO/NiO nanocomposite. In the crystal structure, ZnO and NiO are hexagonal and cubic geometry, respectively. No diffraction peaks belonging to impurities could be found, indicating that the precursor was completely transformed into ZnO/NiO mixed metal oxides. The FTIR spectrum confirmed the formation pure metal oxides ZnO/NiO. The SEM study divulged sphere like particles. By the electrochemical study, the S_{cp} value of 198 F g^{-1} at a scan rate of 5mVs^{-1}. From charge discharge graph the calculated the maximum S_{cp} value is 450 F g^{-1} at 0.5 mA/cm^2.

The ZnO/NiO composite modified electrode has higher S_{cp} and also exhibited an excellent cyclic stability performance of 74.7 % retained up to 5000 cycles. The result obtained from these electrochemical performances suggests that the pristine ZnO/NiO nanosphere is considered as active- material for applications of electrochemical storage devices.

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Synthesis and Characterization of Sol-Gel Derived $Bivo_4$ Nanoparticles for Electrochemical Applications

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Abstract: ***$BiVO_4$ nanoparticles (NPs) were prepared sol-gel technique for the potential electrode of supercapacitor applications. The crystal structure, elemental composition, and surface morphology of the synthesized sample were characterized by powder X-ray diffraction (PXRD), scanning electron microscopy and elemental analysis (EDS) spectrum, respectively. The diffraction peaks were well indexed with monoclinic structure. The morphology of the synthesized sample exhibited that small flattened rice shaped structure with the average particle size of ~50 nm. The room temperature capacitive behaviour of $BiVO_4$ NPs electrode was recorded by cyclic voltammetry (CV) in 2 M of KOH electrolyte. The enhance specific capacitance (C_{SP}= 139 F g^{-1}) was observed at the scan rate of ~10 mVs^{-1}. The results show that the as synthesized $BiVO_4$ NPs is a potential candidate for electrochemical supercapacitor application.***

Keywords: PXRD, Monoclinic, Cyclic Voltammetry, Specific capacitance, Supercapacitor.

I. INTRODUCTION

Among the exhaustion of remnant energy, global warming and pollution, renewable energy capital have involved an enormous amount of concentration [1]. The insist for electrochemical energy storage devices have enlarged in the past few years. With the energy storage devices, supercapacitors [SCs] are considered one of the majority promising device due to their specific power being as high as conservative capacitors and a specific energy close to that of batteries, as well as their other advantages : eco-friendly and comparatively low cost [2]. Among the numerous supercapacitor based electrodes, pseudocapacitive based transition-metal oxides or inorganic compounds showed large energy density due to faradic redox charge storage mechanism, which has higher energy density compared to the electrochemical double-Layer capacitive carbon materials [3, 4]. In pseudocapacitors, the electrochemical charge storage is done by Faradic redox reactions. The pseudocapacitors based electrodes were fabricated by means of conducting polymers, metal oxides and hydroxides. Particularly some materials such as polyaniline, $NiMoO_4$, NiO, Co_3O_4 and MnO_2 were used as pseudocapacitors electrodes [5]. In this present study demonstrate the synthesized $BiVO_4$ modified working electrode used as a pseudocapacitor electrode material for supercapacitor application. Based on the previous reports, $BiVO_4$ used as efficacy photocatalyst, phosphor hosts, sensors and an electrode for electrochemical devices, etc. [6]. For the past few years, a small number of efforts have been made to use $BiVO_4$ as a pseudocapacitive material for high – performance supercapacitors because of its better electrical conductivity and high specific capacitance [7]. As we known, the synergistic effect of bismuth species and Vanadium species plays important roles in the high electrochemical performance of Bismuth vanadium binary oxides. Therefore, the bismuth vanadium binary metal oxides are expected to deliver higher supercapacitive performance than both bismuth oxide and vanadium oxides [8].

Herein, we report the synthesis of $BiVO_4$ NPs by a simplistic and eco – friendly sol-gel technique. The $BiVO_4$ sample loaded modified working electrode improved the electrochemical performance. The $BiVO_4$ modified electrode revealed the highest specific capacitance 139 Fg^{-1} at 10 mVs^{-1}.

II. EXPERIMENTAL

A. *Sample preparation*

All the starting precursors were purchased with AR grade and used for preparation without any addition further purification process. The BiVO4 NPs were synthesized by sol-gel method using Bi(NO3)3·5H2O (bismuth nitrate pentahydrate), NH4VO3 (ammonium metavanadate), HNO3 (Nitric acid), NH4OH (ammonium hydroxide) and CH3COOH (acetic acid) as the starting materials. In the typical synthesis, the stoichiometric amount of the starting precursors such as Bi $(NO_3)_3{\cdot}5H_2O$ and NH_4VO_3 chemicals were dissolved in 50 ml of double distilled (DD) water, separately. The above two solutions were mixed together. Additionally, 100 ml of ethanol was dropped wise added into the solution and hence the stirrer heat was increased at 70 ^oC. The yellow sol was formed.

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Further 1M of acetic acid (CH_3COOH) to make a 50 ml stock solution, the solution was added drop wise the solution maintained at 100 ^oC for a few hrs. The gel was collected and calcined at 600 ^oC in the furnace. The pure monoclinic $BiVO_4$ sample was formed.

III. RESULTS AND DISCUSSION

A. PXRD

The crystal structural and phase identification analysis of sol-gel derived $BiVO_4$ sample were characterized by powder X-ray Diffraction (PXRD) technique. The XRD pattern of $BiVO_4$ material is as shown in Fig. 1. The sharp powder X ray diffraction peaks showed that the sample has the high crystalline nature. The diffraction peaks are completely matched and good aggreement with the reference pattern of monoclinic-$BiVO_4$ (JCPDS card no- 75-1866) and with the space group I2/a [9]. No more other peaks are observed in these patterns. The crystalline size was measured by using Scherer's equation, the average size of the particle is ~26 nm.

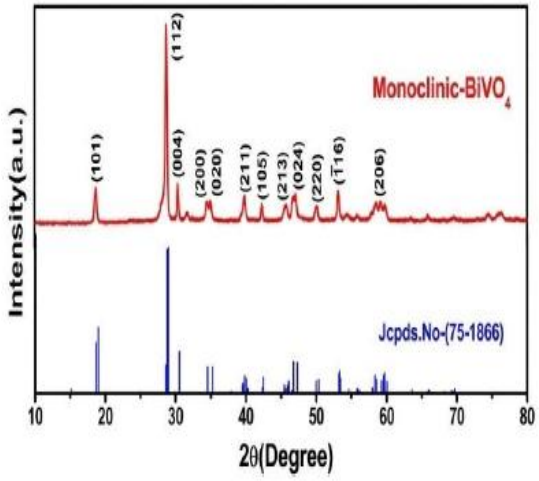

Fig. 1.XRD pattern of Monoclinic $BiVO_4$

B. SEM and EDX Analysis

The surface micrographs and present elements of the $BiVO_4$ sample was analyzed using SEM and EDX analysis. The SEM images of $BiVO_4$ were recorded with different magnifications and the SEM images are displayed in Fig. 2(a,b). The particles clearly show that the flattened rice shaped morphology for pure $BiVO_4$ (Fig. 2 (a & b)). From the SEM analysis the average size of the particle is measured. The mean size of the particle is ~50 nm. Fig.2. (c) EDX spectrum shows the purity of the sample. This is at last affirmed the presence of the elements such as Bi, V and O and no other extra impurity peaks detected which completely
favors the preparation $BiVO_4$ material through sol-gel technique.

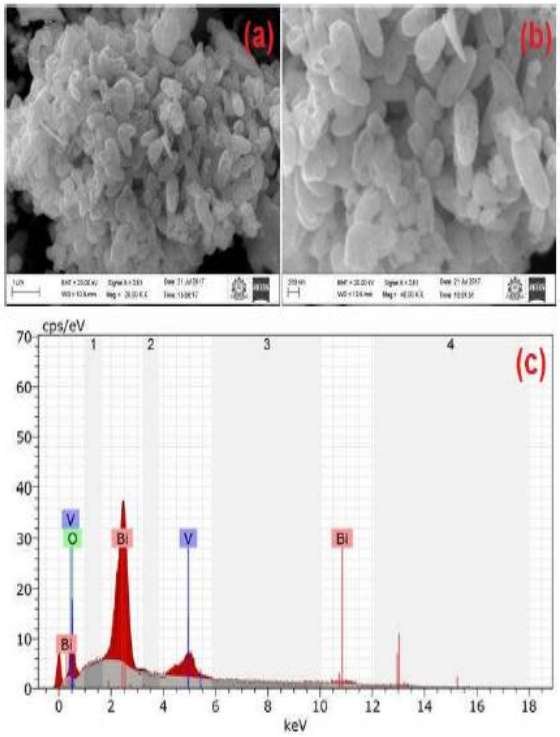

Fig. 2.SEM and EDX pattern of Monoclinic $BiVO_4$

C. *Cyclic Voltammetry Analysis*

The Cyclic Voltammetry (CV) is major tool to investigate the electrochemical behaviour of the sample modified electrode. This is confirming the either EDLC or Pseudocapacitance nature. The prepared sample was coated on nickel foam which act as a working electrode, reference electrode is Ag/Agcl and platinum wire is act as a counter electrode. The entire reaction was tested by 2M KOH solution. The Cyclic Voltammetry graph is shown in fig. 3a. From the CV graph divulged the pseduocapacitance nature of the $BiVO_4$ sample. The faradaic redox reactions were take place on the surface of the electrode material.

$$BiVO_4 + xK_+ + xe \leftrightarrow Kx\ BiVO_4 \quad (1)$$

Where, x represents the mole concentration of K^+ ions. There can be seen that there are one anodic and one cathodic peak in the CV graph as shown in Fig. 3a. The appearance of the anodic peak at -0.7 V occurrence of Bi^{3+} to Bi^0. The peak current versus the square root of the scan rate of the $BiVO_4$ NPs is shown in Fig. 3b. This indicates a linear relationship between peak current and square root scan rate, it denotes the electrode reaction is diffusion-controlled. The presence of cathodic peak at -0.2 V attributes the reduction reaction of Bi metal to Bi^{3+} [7, 8].The C_{SP} values of the working electrode found by the CV curve using above equation 1. The calculated C_{SP} values are 139, 109, 75, 70 and 64 F g^{-1} for different scan rates from 10 to 100 mV s^{-1} respectively. The calculated C_{SP} values with corresponding scan rate are presented in table 1. The C_{SP} values are reduced with increase of scan rate. This is due to the ion (K^+) transfer process between electrolyte and electrode surface [10]. The maximum C_{SP} (= 139 F g^{-1}) is observed for $BiVO_4$ modified
electrode with the scan rate of 10 mV s^{-1}.

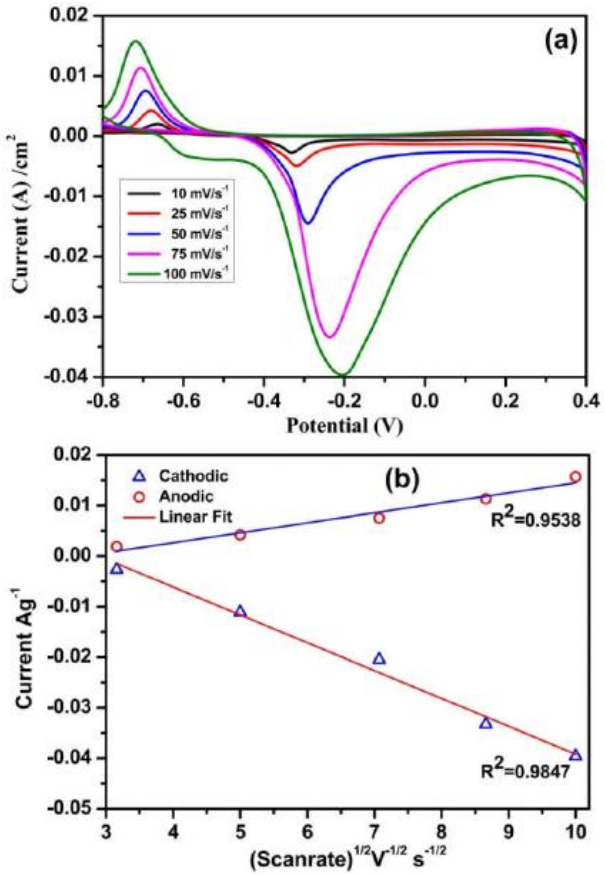

Fig.3. (a) Cyclic Voltamograme, (b) Peak current versus square root of scan rate plots of Monoclinic $BiVO_4$ sample

TABLE.1. SCAN RATE VS SPECIFIC CAPACITANCE

Sl.No	Scan rate	Specific capacitance (Fg^{-1})
1	10	139
2	25	109
3	50	75
4	75	70
5	100	64

IV. CONCLUSION

The flattened rice shaped $BiVO_4$ NPs were successfully prepared by sol-gel technique. From the structural analysis, the pure monoclinic $BiVO_4$ crystal structure was confirmed through PXRD pattern. The PXRD data sets were well matched with standard data. The surface morphology and sample purity were confirmed by SEM and EDX analysis. In addition the modified $BiVO_4$ NPs electrode was examined with cyclic voltametry analysis. Cyclic voltametry analysis showed the pseudocapacitance behaviour of $BiVO_4$ NPs. The electrode has obtained a maximum C_{SP} of 139 F g^{-1} at a scan rate 10 mV s^{-1}. These electrochemical study leads to flattened rice shaped $BiVO_4$ electrode is significant candidate for supercapacitor application.

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Medicinal Plants as Potent Power for Malaria Control: Review

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ABSTRACT

Mosquito vectors are living organisms that can transmit infectious diseases between human from animals to humans. It is bloodsucking insect that ingest disease-producing microorganisms during a blood meal from an infected host. This review aims to explain some medicinal plants that exhibit mosquitocidal agents on genus Anopheles mosquito species. Currently, the utilization of artificial chemicals to regulate insects and mosquitoes raises many considerations associated with environment and human health. Another aim is to use natural merchandise that possess sensible effectiveness, simple handiness, fewer adverse environmental impacts, and are environmentally friendly have crystal rectifier to the multiplied interest in plant origin chemical pesticides. It's supported original articles obtained by looking on major databases. Our literature review disclosed that 96 medicinal plants used in the all parts (leaves, root, bark and bud). This article envisaged to review the reports of ovicidal, oviposition deterrent, larvicidal, pupicidal and repellent activities, which might be employed in vector-borne disease management programs.

Keywords: *Mosquitocidal Activity, Medicinal Plants, Anopheles Species*.

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INTRODUCTION

Malaria

Mosquitoes have been accounted as the well-spring of different diseases influencing human. Including around 3500 species, mosquitoes are discovered past the tropical and subtropical locales of the world [1]. Malaria in people is brought on by contamination with one or a greater amount of a few types of *Plasmodium* (i.e., *Plasmodium falciparum, Plasmodium vivax, Plasmodium ovale, Plasmodium malariae*, and incidentally other *Plasmodium* species) parasites. The parasite is transmitted by the bite of an infective female Anopheles mosquito. *P. falciparum* and *P. vivax* species cause the most contaminations around the world. *P. falciparum* is the operators that most usually cause serious and possibly lethal malaria. As indicated by the most as of late accessible data, an expected 207 million clinical cases and 627,000 (0.3%) deaths were reported worldwide in 2012, for the most part among children matured five years living in sub-Saharan Africa. *P. vivax* and *P. ovale* have torpid liver stages, which can reactivate and cause malaria a while for years after the underlying disease. *P. malariae* can bring about durable diseases and if untreated or insufficiently treated, can continue asymptomatically in the human host for quite a long time, even a lifetime.

Malaria disease worldwide

Anopheles stephensi transmits of malaria fever in urban district of India. Among 53 Anopheline species present in India, 9 are vectors of malaria fever [2] and malaria is still the most critical reason for mortality with around a few million new cases emerging each year. A more effective approach to decrease mosquito populace is to focus on the hatchlings. The main arrangement is to keep the ailment conveying mosquito from

reproducing and gnawing people. In this situation, mosquito vector control is pivotal. *An. subpictus* Grassi is distributed throughout India, China, Afghanistan, Borneo, Malaysia, Sri Lanka, Philippines, Indonesia, and Java. It is a dominant species in Haryana and Uttaranchal states. Though it is a non-vector species, same infected specimens with malaria parasite have been reported from India, Indonesia, and Java. *An. subpictus* is a significant secondary vector in Sri Lanka [3, 4]. *An. subpictus* is recognized as the secondary vector of malaria in South East Asia, with a large number of cases being reported from India. India contributes 77% of the world's population [5, 6]. Members of the *An. gambiae* complex are the most important vectors of malaria in sub-Saharan Africa of which *An. gambiae* and *An. arabiensis* are the most widely distributed and most efficient vectors [7]. Mosquito larvae and pupae are normally focused on utilizing organophosphates, insect development controllers and microbial control operators. Inside lingering showering and insect spray treated bed net are likewise utilized to decrease transmission of malaria in tropical nations. Notwithstanding, engineered chemicals have solid negative consequences for human wellbeing and nature, and incite resistance in various mosquito species [8, 9].

Eruption of malaria disease in the world

Mosquitoes have been accounted for as the wellspring of different diseases influencing human, and are discovered in the tropical and subtropical locales of the world. In Asia, the mosquito is the most important vector of malaria, with 200 to 450 million infections annually worldwide; it sources up to 2.7 million deaths. WHO Malaria Report estimates that 3.3 billion people were at the peril of malaria in 2010, although of all geographical regions, populations living in sub-Saharan Africa (SSA) have the highest peril of obtaining malaria; among 216 million episodes of malaria in 2010, approximately 81%, or 174 million occasions, were noticed from the African region. There were estimated malaria demises of 655,000 in 2010, of which 91% were from Africa [7]. Malaria afflicts 36% of the world people i.e. 2020 million in 107 countries and territories position in the tropical and subtropical regions.

As indicated by the most recent evaluations, there were around 198 million cases with malaria fever in 2013 and an expected 584,000 passing. Most passing's happen among children living in Africa. Malaria death rates among children in Africa have been decreased by an expected percentage of 58% since 2000. Globally, 4.28 million deaths have been averted due to malaria control efforts between 2001 and 2013, of which 3.92 million (92%) are in children under 5 years of age in sub-Saharan Africa. They account for 20% of the 20 million child deaths; it has been estimated that it has been averted in sub-Saharan Africa between 2001 and 2013 through the overall reduction in child mortality rates [3]. In 2013, around 6.3 million children died before their fifth birthday, at a rate of around 17,000 per day. The risk of dying before age five varies enormously depending on where a child is born. In Luxembourg, the under-five mortality pace is fair 2 deaths per 1,000 live births; in Angola it is 167 per 1,000. Since 1990, 223 million children have died before their fifth birthday [9]. Malaria deaths dropped by 58% in African children under five years old from 2000-2013 [3]. As per the most recent evaluations from WHO, there were 214 million new instances of malaria worldwide in 2015 and there were an expected 438,000 malaria passing. Children under five are especially helpless to malaria ailment, disease and demise. Malaria slaughtered an expected 306,000 under-fives comprehensively, incorporating 292,000 kids in the African region [4]. Malaria incidence rate has declined by 41% since 2000, and 2010. Mortality has fallen by 61% since 2000 and 29% since 2015. Seventeen countries eliminated malaria between 2000 and 2015, with a further 13 countries "approaching elimination" [10]. According to the reports, India has 6% of all malaria cases in the world, 6% of the deaths, and 51% of the cases in global *P. vivax* cases. The report estimates the total cases in India as 1.31 million and the deaths as 194 [11]. History uncovers that the occurrence of malaria in India happened in the 1945s with an expected 75 million cases and 800,000 passed for each year. This makes female mosquito a perfect transmitter of different blood borne pathogens and operators of destroying human diseases. The year's malaria has been considered as one of the main sources of

deaths in India (http://nvbdcp.gov.in/malaria-new.html) (Table 1).

Medicinal plants based mosquito control

Medicinal plants might be elective wellsprings of mosquito control specialists since they constitute a rich wellspring of bioactive compounds that are biodegradable and conceivably suitable to control mosquitoes. Pesticides of organic inception might serve as suitable option for biocontrol procedures later on [12]. The use of synthetic chemicals for management of mosquitoes raises many issues connected to the environment and human health [13]. Eco-friendly tools to manage mosquito young instar populations in an IPM framework are urgently needed. Natural products are usually most preferred due to their less harmful nature and fast biodegradability. Plant products have been used traditionally by human communities in different rural areas worldwide against insect vectors and parasites [14]. Most of the mosquito control programs target the larval stage in their breeding sites with larvicides. Monoterpenes such as a-pinene, limonene, terpinolene, citronellol, citronellal, camphor and thymol are regular constituents of a number of essential oils of medicinal plants. The chemicals derived from plants have been projected as weapons in future mosquito control program as they are shown to act as general toxicant, growth and reproductive inhibitors, repellents and oviposition-deterrent [15]. Table 2 gives definite surveys of plant items reported for mosquitocides and insecticides tested against malarial species.

CONCLUSION

Plants as option for wellspring of larvicidal, ovicidal, oviposition deterrent, pupicidal and repellent activity reported in various ethnobotanical reviews. All the tried plants had diverse scope of larvicidal, ovicidal, oviposition deterrent, pupicidal and repellent action which might be utilized as traditional mosquito control operators. On the premise of the present examination results we presume that therapeutic plants extracts contains strong mosquitocidal bioactive standards which might be required further sanitizations to have its manufactured analogs, which will be done in future.

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Table 1. Incidence of Malaria surveillance data in India

Years	Total malaria cases million	*P. falciparum* million	Death due to malaria (Thousand)
1961	49,151	-	-
1962	59375	-	-
1963	87306	-	-
1964	112942	-	-
1965	99,667	-	-
1966	148012	-	-
1967	278214	-	-
1968	274634	-	-
1969	347975	-	-
1970	694017	-	-
1971	1322398	-	-
1972	1428649	-	-
1973	1930273	-	-
1974	3167658	-	-
1975	5166142	-	-
1976	6.47	0.75	59
1984	2.18	0.65	247
1985	1.86	0.54	213
1986	1.79	0.64	323
1987	1.66	0.62	188
1988	1.85	0.68	209
1989	2.05	0.76	268
1990	2.02	0.75	353
1991	2.12	0.92	421
1992	2.13	0.88	422
1993	2.21	0.85	354
1994	2.51	0.99	1122
1995	2.93	1.14	1151
1996	3.04	1.18	1010
1997	2.66	1.01	879
1998	2.22	1.03	664
1999	2.28	1.14	1048
2000	2.03	1.05	932
2001	2.09	1.01	1005
2002	1.84	0.90	973
2003	1.87	0.86	1006
2004	1.92	0.89	949
2005	1.82	0.81	963
2006	1.79	0.84	1707
2007	1.51	0.74	1311
2008	1.53	0.77	1055
2009	1.56	0.84	1144
2010	1.60	0.83	1018
2011	1.31	0.67	754
2012	1.07	0.53	519
2013	0.88	0.46	440
2014	1.10	0.72	561
2015	1.17	0.78	384
2016	1.09	0.71	331
2017	1.31	0.53	194
2018 (up to July)	172643	92436	14

Sources data (http://nvbdcp.gov.in/malaria-new.html)

Table 2. Plant reported for larvicidal, ovicidal, oviposition deterrent, pupicidal and repellent activity

Plant species	Family	Plant parts	Instars larvae / eggs / pupae / adults	Bioactivity	$LC_{50}$and LC_{90}values	References
Abrus precatorius	Fabaceae	Seed	IV instars	Larvicidal	Ethyl acetate extract was19.31 and 71.71 ppm, respectively	Bagavan and Rahuman [16]
Acalypha alnifolia	Euphorbiaceae	Leaf	NL	Ovicidal, repellent, adulticidal	Adulticidal activity of methanol extract was 274.76 and 495.88 ppm, respectively	Kovendan et al. [17]
Achras sapota	Sapotaceae	Leaf	III instars	Larvicidal, ovicidal, pupicidal and repellent	Methanol extract was 39.54 and 98.53 ppm, respectively	Krishnappa and Elumalai [18]
Adansonia digitata	Bornbacaceae	Leaf	III instars	Larvicidal and repellent	Methanol extract was 78.18 and 155.42 ppm, respectively	Krishnappa et al. [19]
Adhatoda vasica	Acanthaceae	Leaf	IV instars	Larvicidal	Acetone extract was 18.20 and 96.33 ppm, respectively	Kamaraj et al. [20]
Aloe vera	Asphodelaceae	leaves	I, II, III, IV instars	Larvicidal, pupicidal	Ethanol extract of LC_{50} value was 48.79, 59.09, 70.88, 83.58 ppm, respectively	Dinesh et al. [21]
Annona muricata	Annonaceae	Leaf	NL	Larvicidal	Aqueous extract of LC_{50} value was 61.38 and 156.55 ppm, respectively	Santhosh et al. [22]
Annona reticulate	Annonaceae	leaf	III instars	Larvicidal, ovicidal and pupicidal	Methanol extract of LC_{50} value was 74.36 and 93.85 ppm, respectively	Baluselvakumar et al. [23]
Annona sengalensis	Annonaceae	Leaf	IV instars	Larvicidal, ovicidal and pupicidal	N-hexane fraction of LC_{50} value was 298.8 and 572.9 ppm, respectively	Younoussa et al. [24]
Annona squamosa	Annonaceae	Leaf	IV instars	Larvicidal, oviposition deterrent and repellent	Petroleum ether extract was 118.4 and 213. 10 ppm, respectively	Vijaya kumar et al. [25]
Arachis hypogaea	Fabaceae	Leaves	IV instar	Larvicidal	Aqueous extract of LC_{50} value was 71.57 and 214.62 ppm, respectively	KuppanVelu et al. [26]
Artemisia nilagirica	Compositae	Leaves	I, II, III, IV instars	Larvicidal, pupicidal, adulticidal, repellent	Methanol extract was 272.50, 311.40, 361.51, 442.51 and 590.07, 688.81, 789.34, 901.59 ppm, respectively	Panneerselvam et al. [27]
Azadirachta indica	Meliaceae	Leaf, pod	IV instars	Larvicidal, repellent and smoke toxicity effect	Azadirachtin was 0.299 and 1.061 ppm, respectively	Murugan et al. [28]
Balanites aegyptiaca	Balanitaceae	Roots	IV instars	Larvicidal	Ethanolic extract of LC_{50} value was 6.61 ppm, respectively	Nganjiwa et al. [29]
Calotropis gigantean	Asclepiadaceae	Leaf	I, II, III, IV instars	Larvicidal, pupicidal	Methanol extract was 73.77, 89.64, 121.69,	Kovendan et al. [30]

					155.49 and 199.31, 245.07, 373.85, 415.31 ppm, respectively	
Catharanthus roseus	Apocynaceae	Leaves	IV instar	Larvicidal, pupicidal	Aqueous extract was 68.62 and 184.85 ppm, respectively	Subarani et al. [31]
Chlomolaena adorata	Asteraceae	Leaf	III, IV instars	Larvicidal	Methanolic extract was 1613 and 8306 ppm, respectively	Jagruti et al. [32]
Chomelia asiatica	Rubiaceae	Leaf	III instars	Larvicidal	Aqueous extract of LC_{50} value was 90.17 and 165.18 ppm, respectively	Muthu kumaran and Govindarajan [33]
Chrysanthemum indicum	Asteraceae	flower	I, II, III, IV instars	Larvicidal	Aqueous extract was 78.22, 110.63, 181.85, 298.79 and 417.30, 531.31, 675.76, 973.54 ppm, respectively	Arokiyaraj et al. [34]
Cissus quadrangularis	Vitaceae	Leaf	IV instars	Larvicidal and ovicidal	Methanol extract was 37.48 and 95.93 ppm, respectively	Krishnappa et al. [35]
Citrullus lanatus	Cucurbitaceae	Leaf	III, IV instars	Larvicidal	Methanolic extract was 84.23 and 989.39 ppm, respectively	Ajithadas aruna et al. [36]
Citrus sinensis	Rutaceae	Leaf	I, II, III, IV instars	Larvicidal	Ethanol extract was 182.24, 227.93, 291.69, 398.00 and 452.44, 544.72, 659.31, 858.92 ppm, respectively	Murugan et al. [37]
Clausena anisata	Rutaceae	Leaf oil	III instars	Larvicidal	Essential oil from119.59 and 209.96 ppm, respectively	Govindarajan [38]
Cleistanthus collinus	Euphorbiaceae	Leaf	III instars	Larvicidal	Ethyl acetate extract was 399.72 and 1251.76 ppm, respectively	Arivoli and Samuel [39]
Clerodendron inerme	Verbinaceae	Leaf	I, II, III, IV instars	Larvicidal	Crude extract was 55.04, 63.33, 73.05, 80.16 and 125.50, 137.16, 153.55, 156.93 ppm, respectively	Kovendan and Murugan [40]
Cochliobolus lunatus	Pleosporaceae	Leaves	II, III, IV instars	Larvicidal	Plant extract was 1.17, 1.30, 141 and 2.99, 3.13, 3.29 ppm, respectively	Rahul et al. [41]
Coleus aromaticus	Lamiaceae	Leaf	III instars	Larvicidal	Ethyl acetate extract was 28.88 and 65.35 ppm, respectively	Barani tharan et al. [42]
Commiphora caudata	Bursaceae	Leaf	III instars	Larvicidal	Ethyl acetate extract was 96.04 and 104.44 ppm, respectively	Barani tharan and Dhanasekaran [43]
Corchorus capsularis	Malvaceae	Leaf	III instars	Larvicidal, ovicidal	Methanol extract was 176.19 and 334.56 ppm, respectively	Elangovan et al. [44]
Cymbopogon citratus	Poaceae	Leaf	III instars	Larvicidal	Methanol extract was 74.02 and 158.20 ppm, respectively	Karunamoorthi and Ilango [45]
Cuminum cyminum	Apiaceae	Vegetables	III instars	Larvicidal (24, 48 and 72	Chloroform;methanol (1:1) extract of 0.15,	Singha, and Chandra [46]

				hours)	0.09, 0.04 and 1.21, 0.23, 0.12 ppm, respectively	
Datura metel	Solanaceae	Leaves	I, II, III, IV instars	Larvicidal	Aqueous extract was 34.69, 42.16, 47.16, 55.22 ppm and 105.47, 118.82, 133.25, 150.11 ppm, respectively	Murugan et al. [47]
Datura stramonium	Solanaceae	Leaf	IV instars	Larvicidal, repellent	Ethanolic extract of LD_{50} and LD_{90} values was 16.07 and 41.95 dose , respectively	Swathi et al. [48]
Derris elliptica	Fabaceae	Root, shoot	III instars	Larvicidal	Petroleum ether extract was 0.307 and 0.58 ppm, respectively	Dohutia et al. [49]
Elaeagnu indica	Elaeagnaceae	Leaf	IV instar	Larvicidal	Acetone extract of LC_{50} value was 3.30 and 6.94 ppm, respectively	Srinivasan et al. [50]
Eranthemum roseum	Acanthaceae	Leaf	III instars	Larvicidal, ovicidal, pupicidal	Acetone extract was 121.65 and 237.38 ppm, respectively	Elumalai et al. [51]
Ervatamia coronaria	Apocynaceae	Leaf	III instar	Larvicidal	Methanol extract was 86.47 and 159.59 ppm, respectively	Govindarajan et al. [52]
Euodia ridleyi	Rutaceae	Leaf		Larvicidal	Ethyl acetate extract was 120.0 and 178.20 ppm, respectively	Prathibha et al. [53]
Euphorbia hirta	Euphorbiaceae	Leaves	I, II, III, IV instars	Larvicidal, pupicidal	Methanol extract was 121.51, 145.40, 169.11, 197.40 and 236.44, 293.75, 331.42, 371.34 ppm, respectively	Agalya Priyadarshini et al. [54]
Exacum pedunculatum	Gentinaceae	Leaf	IV instars	Larvicidal and ovicidal	Ethanol extract was 121.24 240.57 ppm, respectively	Elangovan et al. [55]
Ficus benghalensis	Moraceae	Leaf	III instars	Larvicidal	Methanol extract of 56.66 and 100.88 ppm, respectively	Govindarajan et al. [56]
Gliricidia sepium	Leguminosae	Leaf	III instars	Larvicidal, ovicidal and pupicidal	Ethanol extract was 121.79 and 231. 98 ppm, respectively	Krishnappa et al. [57]
Gnetum ula	Gnetaceae	Leaf	III instars	Larvicidal, ovicidal and repellent	Ethanol extract of LC_{50} value was 82.86 ppm, respectively	Dhanasekaran et al. [58]
Gymnema sylvestre	Asclepiadaceae	Leaves	IV instar	Larvicidal	Petroleum ether extract of LC_{50} value was 166.28 ppm, respectively	Gopiesh Khanna et al. [59]
Hippophae rhamnoides	Elaeagnaceae	Leaf	IV instars	Larvicidal (24 hours only)	Ethanol extract was 1494.30 and 2605.78 ppm, respectively	Bazhir and Javid [60]
Hybanthus enneaspermus	Violaceae	Leaf	IV instar	Larvicidal	Aqueous extract of LC_{50} value was 117.83, respectively	Suman et al. [61]
Jatropha curcas	Euphorbiaceae	Leaf	III instars	Larvicidal	Methanol extract was 92.09 and 241.09 ppm, respectively	Zewdneh Tomass et al. [62]
Juniperus	Cupressaceae	Oil	III instars	Larvicidal	Essential oil was 14.42	Kaliyaperumal et

procera					and 24.65 ppm, respectively	al. [63]
Lantana camara	Verbenaceae	Flowers	III, IV instars	Larvicidal	Petroleum ether extract was 126.7 and 248.9 ppm, respectively	Babita et al. [64]
Leucas aspera	Lamiaceae	Leaf	III instars	Larvicidal	Ethyl acetate extract was 352.84 and 1033.60 ppm, respectively	Arivoli et al. [65]
Melissa officinalis	Lamiaceae	Oil	III instar	Larvicidal, ovicidal and repellent	Citronellal compound was 85.44 and 159.73 ppm	Barani tharan et al. [14]
Metarhizium anisopliae	Clavicipitaceae	Leaves	I, II, III, IV instars	Larvicidal, pupicidal	Ethanol extract was 1.40, 3.99, 5.56, 8.77 and 13.84, 17.62, 22.20, 25.71 ppm, respectively	Kovendan et al. [66]
Momordica charantia	Cucurbitaceae	Leaf	I, II, III, IV instars	Larvicidal, pupicidal	Methanol extract was 93.45, 123.74, 167.17, 216.15 and 454.96, 573.31, 630.66, 722.25 ppm, respectively	Subramaniam et al. [67]
Morinda citrifolia	Rubiaceae	Leaf	III instar	Larvicidal	Methanol extract was 261.96 and 505.06 ppm, respectively	Kovendan et al. [68]
Moringa oleifera	Moringaceae	Leaf	I, II, III and IV instars	Larvicidal, pupicidal and repellent	Methanol extract was 57.7, 63.9, 72.4, 78.9 and 125.9, 133.0, 139.8, 143.2 ppm, respectively	Prabhu et al. [69]
Murraya exotica	Rutaceae	Oil	IV instars	Larvicidal	Essential oil of LC_{50} value was 56.3 and 107.8 ppm, respectively	Krishnamoorthy et al. [70]
Nelumbo nucifera	Nelumbonaceae	Leaves	IV instar	Larvicidal	Methanol extract was 8.89 and 28.65 ppm, respectively	Santhoshkumar et al. [71]
Nepeta cataria	Lamiaceae	Leaf	IV instars	Larvicidal	Methanol extract of LC_{50} value was 0.93 ppm, respectively	Adewole et al. [72]
Nerium oleander	Apocynaceae	Leaf	I, II, III, IV instars	Larvicidal, pupicidal	Extract was 20.60, 24.90, 28.22, 33.55 and 41.62, 50.33, 57.78, 33.99 ppm, respectively	Mathath Roni et al. [73]
Nigella sativa	Ranunculaceae	Seed	IV instars	Larvicidal	Essential oil of LC_{50} value was 88.1 and 272.4 ppm, respectively	Adaikala Raj et al. [74]
Nymphaea lotus	Nymphaeaceae	Leaf	III instars	Larvicidal	Ethanolic extract of LC_{50} value was 62.8 ppm, respectively	Iman and Tajuddeen [75]
Ocimum basilicum	Lamiaceae	Leaf	III instars	Larvicidal	Methanol extract of 0.01 and 3.13 ppm, respectively	Mauryaet al. [76]
Ocimum gratissimum	Lamiaceae	Leaf	Instars larvae	Larvicidal	Ethanolic extract was 60.9 and 464.4 mg/ml, respectively	Pearl et al. [77]
Ocimum sanctum	Lamiaceae	Leaf	III instars	Larvicidal	Methanol extract was 115.32 and 209.25 ppm, respectively	Gokulakrishnan et al. [78]
Orthosiphon	Labiatae	Leaf	III instars	Larvicidal	Methanol extract of	Kovendan et al.

thymiflorus					118.74 and 377.09 ppm, respectively	[79]
Oxystelma esculentum	Apocynaceae	Leaf	III instars	Larvicidal	Methanol extract was 63.84 and 122.48 ppm, respectively	Balu et al. [80]
Pavonia odorata	Malvaceae	Leaf	III instars	Larvicidal and repellent	Methanol extract was 58.22 and 239.82 ppm, respectively	Balu et al. [81]
Pedalium murex	Pedaliaceae	Leaf	III instar	Larvicidal, ovicidal and repellent	Methanol extract was 111.66 and 200.01 ppm	Gokulakrishnan et al. [82]
Pemphis acidula	Lythraceae	Leaf	III instars	Larvicidal and ovicidal	Methanol extract of 22.1 and 43.71 ppm, respectively	Samidurai [83]
Pergularia daemia	Asclepiadaceae	Leaves	I, II, III, IV	Larvicidal	Crude aqueous extract was 81.47 and 159.51 ppm, respectively	Chandrashekhar et al. [84]
Piper nigrum	Piperaceae	Seed	III instars	Larvicidal and repellent	Ethyl acetate was 24.54 and 108.03 ppm, respectively	Kamaraj et al. [85]
Pisonia alba	Nyctaginaceae	Leaves	III instars	Larvicidal and ovicidal	Petrolium ether 1.0 ppm 2.0	Tamizhazhagan et al [86]
Pithecellobium dulce	Fabaceae	Leaf, seed	III instars	Larvicidal and ovicidal	Methanol extract was 145.43 and 251.23 ppm, respectively	Govindarajan et al. [87]
Polianthes tuberose	Agavaceae	Bud	I, II, III and IV instars	Larvicidal and repellent (24 hours only)	Crude plant extract of 0.18, 0.51, 0.05, 0.30 and 1.38, 0.51, 0.29, 7.03 ppm, respectively	Rawani et al. [88]
Polygala arvensis	Polygalaceae	Leaf	III instars	Larvicidal, ovicidal and repellent	Methanol extract was 46.37 and 189.82 ppm, respectively	Deepa et al. [89] and Lakshmanan et al.[90]
Pongamia pinnata	Fabaceae	Bark	IV instars	Larvicidal	Methanol extract was 151.7 and 299.4 ppm, respectively	Guna et al. [91]
Prosopis juliflora	Fabaceae	Leaf	III instars	Larvicidal	Leaf extracts was 37.55 and 514.35 ppm, respectively	Tyagi Varun et al. [92]
Quassia africana	Simaroubaceae	Leaf, stem, roots	IV instars	Larvicidal	Methanol extract of LC_{50} value was 17.58 ppm, respectively	Woquan Sama et al. [93]
Ricinus communis	Euphorbiaceae	Leaf	III instars	Larvicidal	Ethanol extract was 282.70 and 501.23 ppm, respectively	Awad et al. [94]
Sesamum indicum	Pedaliaceae	Leaf	III instar	Larvicidal	Methanol extract was 338.27 and 538.50 ppm, respectively	Barani tharan et al. [95]
Sida acuta	Malvaceae	Leaf	III instars	Larvicidal and repellent	Methanol extract was 38.64 and 74.78 ppm, respectively	Govindarajan [96]
Solanum trilobatum	Solanaceae	Leaf		Toxicity	116.64 ppm	Premalatha et al. [97]
Spondias mombin	Anacardiaceae	Leaf	IV instars	Larvicidal	Hexane fraction was 92.2 and 245.37 ppm, respectively	Elijah et al. [98]

Tinospora cordifolia	Menisperma-ceae	Leaf	IV instars	Larvicidal	Aqueous extract of LC_{50} was 53.93 ppm, respec-tively	Jayaseelan et al. [99]
Tridax pro-cumbens	Compositae	Whole	IV instars	Larvicidal (24 hours only)	Methanol extract was 57.991 and 172.280 ppm, respectively	Devan Elumalai et al. [100]
Vernonia anthelmintica	Asteraceae	Seed	III, IV instars	Larvicidal	Ethanol extract was 1.95 and 10.49 ppm, respec-tively	Alina et al. [101]
Zanthoxylum gilletii	Rutaceae	Oil	III instars	Larvicidal	Essential oil LC_{50} value was 57.73 and140.24 ppm, respectively	Ombito et al. [102]

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Punica Granatum-Based Green Ethanolic Extract as Highly Effective and Eco-Friendly Larvicide, Repellent against Medically Important Mosquito Vectors

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ABSTRACT

Background & objective: Mosquito vectors are living organisms that can transmit infectious diseases between human from animals to humans. It is bloodsucking insect that ingest disease-producing microorganisms during a blood meal from an infected host. The present investigation discovered that the larvicidal activity of ethanol extract of P. granatum showed most mortality among the opposite crude extracts. Methods: The chemical composition of P. granatum ethanolic extract was analyzed by gas chromatography-mass spectroscopy. A total of twenty five III instar larvae of An. stephensi and Cx. quinquefasciatus were exposed to various concentrations (50-250 ppm) in the laboratory by using the standard protocol described by WHO (2005). The repellent activity of P. granatum chemical compositions tested against 100 blood starved female mosquitoes of An. stephensi and Cx. quinquefasciatus using the protocol of WHO (1996). Results: In GC-MS analysis, a total of seven compounds were identified in the ethanolic extract composition, the main component was Methyl 4-piperidineacetate. Further, the LC_{50} and LC_{90} values were found to be 110.36 and 212.28 mg/L against Cx. quinquefasciatus. The repellent activity to be best and therefore the most activity was ascertained at 3.5 mg/cm^2 concentration provided 100% protection up to 240 min against Cx. quinquefasciatus. Conclusion: The results clearly show confirmed that the presence of active compounds in leaf of P. granatum.

Keywords: *Punica granatum, Anopheles stephensi, Culex quinquefasciatus, FT-IR, GC-MS.*

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INTRODUCTION

Malaria is one in all the grave scourges inflicted upon human beings and causes human mortality alongside giant economic loss [1-6]. In line with the newest estimates, there have been regarding 198 million cases of malaria in 2013 and a calculable 584,000 deaths. Most deaths occur among youngsters living in continent, wherever a baby dies each minute from malaria [7] and killed an expected 3,06,000 under-fives widely, including 2,92,000 children in African countries [8]. Death rates have fallen by 61 per cent for 2000 and 2015, with a more 13 countries "approaching elimination" reported WHO, 2016. Presence of the report, India statement for 6 per cent of all malaria cases in the world, 6 per cent deaths, and 51 per cent of the cases in world. The statement estimates the total cases in India found in 1.31 million and deaths at 194 reported WHO, 2017. *Culex quinquefasciatus* is a crucial feature inflicting filariasis, St. Louis encephalitis, Avion malaria and West nile virus. It's extensive-

ly studied, because it transmits crucial diseases [3, 9, 10]. In 2014, estimate is impure with lymphatic filariasis parasites and over 20 per cent of the planet population is at hazard of getting roundworm infection. In Asian nation, it's calculable that regarding 554.2 million folks area unit at hazard of humor disease unhealthiness in a pair of 43 districts [11, 12]. Around the world, 25 million men clumsy person with sex organ sickness and over 15 million folks are afflicted with lymphedema [13].

About 40% of the world's population is at risk from mosquito-borne diseases. In 2015, 2.35 million cases of dengue were reported in the Americas, of which 10 200 cases were identified as severe dengue causing 1181 deaths [14-17]. The year 2015 was characterized by abundant global dengue outbreak, then Philippines reporting more than 169 000 cases and Malaysia exceeding 111 000 doubtful cases of dengue, defining a 59.5% and 16% increase in case numbers to the previous year, respectively. Brazil separately documented over 1.5 million cases in 2015, approximately 3 times higher than in 2014 [18-23].

Punica granatum is one in every of the oldest cultivated plants within the world [24]. It's a crucial crop proverbial by its style and organic process and medicinal properties [25-31]. Many studies have reported the antimicrobial and antifungal [32, 33], molluscicidal [34] and isecticidal [35] activities of extracts from completely different tree components, like bark, leaves, fruit and fruit peel. Biopesticides are alternative to synthetic pesticides because of their generally low environmental pollution, low toxicity to humans and other applications [36]. The chemicals constituents had been isolated and identified from flowers and fruits of pomegranate. The bark and stem contain a number of alkaloids. This research would be helpful to foster research aimed at the identification of novel and safer plant-borne mosquitocides. Further, present study investigated larvicidal and repellent activity of *P. granatum* ethanol extract on important mosquitoes.

MATERIALS AND METHODS

Sample collection and preparation

P. granatum leaves were collected from around Velankanni (10º40'N to 11º12'N latitude and 79º50'E to 80º72'E longitude), Nagapattinam District, Tamilnadu in India. The dried leaves (100g) were powdered by electrical stainless-steel liquidizer and extracted with hexane, ethyl acetate, chloroform and ethanol by Soxhlet equipment. The extract was collected by reduced pressure 22–26 mmHg at 45°C by 'Rotavapour' and therefore the residue obtained was hold on at 4°C. The condensed crude leaves extract was hold on in refrigerator till needed for investigation for larvicidal and repellent activities.

Larvicidal activity

The larvicidal activity of crude *P. granatum* extracts were evaluated based on the method described previously [36]. In view of the wide range and thin range tests, all concentrates from 50-250 ppm were readied and were tried against the newly shed (0-6 shrs) third instar hatchlings of *An. stephensi* and *Cx. quinquefasciatus*. The plants concentrates were added to 1 ml DMSO (Dimethyl sulfoxide) and afterward diluted in 249 ml of dechlorinated faucet water. The control was prepared utilizing 1ml of DMSO as a part of 249 ml of dechlorinated water. The hatchlings of test species (25) were placed in 250 ml plastic glass with 250 ml of fluid medium (249 ml of dechlorinated water + 1ml of Dimethyl Sulfoxide) and the required measure of compound syntheses was included. The larval mortality was inspected and recorded after 24 h post treatment. For every examination, five recreates were kept up at once. Percent mortality was rectified for control mortality according to [37].

Repellent activity

The repellency was evaluated by victimization of the minutes of protection in respect to dose technique was utilized by World Health Organization [38]. Three day old blood-starved mosquitoes (100) were unbroken in a very web cage (45cm × 30cm × 45cm). The volunteer had no contact with lotion, perfumes or perfumed soaps on the day of the assay. The arms of volunteer, solely 25 cm^2 dorsal facet of the skin on every arms were exposed and therefore the remaining space lined by rubber gloves. The crude extracts were applied at 3.5 mg/cm^2 on an individual basis within the exposed space of the fore arm. The time of the take a look at obsessed with whether or not are the target mosquitoes day or

night biters. *An. stephensi* and *Cx. quinquefasciatus* were tested in dark from 20:00 to 4:00. The management and treated arm were introduced at the same time in to the experimental cages, and the mosquitoes were activated. Every take a look at concentration was perennial five times. The volunteer conducted their take a look at of every concentration by inserting the treated and management arm in to an equivalent cage for one full minute for each 5 minutes. The mosquitoes that landed on the hand were recorded and so jolted off before uptake of any blood; creating out a five minutes protection. The proportion of repellency was calculated by the subsequent formula.

$$\% \ Repellency = [(T_a\text{-}T_b)/T_a] \times 100$$

Where T_a is the quantity of mosquitoes in the control group and T_b is the quantity of mosquitoes in the treated group.

Statistical analysis

The examination program probit [39] was utilized for the determination of LC_{50}, LC_{90} and different insights at mean, slope, regression, chi-square qualities were figured utilizing the SPSS 16.0 programming.

RESULTS AND DISCUSSION

The larvicidal activity of crude ethanol, ethyl acetate, chloroform and hexane solvent extracts of *P. granatum* against *An. stephensi* and *Cx. quinquefasciatus* were studied. The ethanol extract of *P. granatum* reported in the present study showed the mosquitocidal properties in the plant, suggestive of their use in mosquito population control (Table 1). *Cx. quinquefasciatus* was more vulnerable followed by *An. stephensi*. Plant extracts exhibited the maximum larvicidal activity with LC_{50} and LC_{90} values of 125.78 and 225.98 mg/L against the larvae of *An. stephensi*, followed by, the ethyl acetate, chloroform and hexane extract with LC_{50} and LC_{90} values are 134.71, 171.27, 198.07 and 232.83, 271.49, 299.88 mg/L. Ethanol, ethyl acetate, chloroform and hexane extracts of *P. granatum* against *Cx. quinquefasciatus* with LC_{50} and LC_{90} values are 110.36, 126.68, 151.06, 176.49 and 212.28, 224.34, 256.75, 275.71 mg/L, respectively (Table 2). The repellent action of the *P. granatum* extract showed important repellent against *An. stephensi* and *Cx. quinquefasciatus*. It showed that repellency depends on the potency of the 3.5 mg/cm^2 provided 100% protection up to 200 and 240 min against *An. stephensi* and *Cx. quinquefasciatus,* respectively (Table 3).

The mass spectral analysis of seven compounds, concentration of percentage (%) and retention indices were summarized in Table 4 and the mass chromatogram was shown in Figure 2. Among all, cardanolide ($C_{23}H_{36}O_2$), n-Boc-4-piperidineacetaldehyde ($C_{12}H_{21}NO_3$), 4-Cyclopropylbenzaldehyde ($C_{10}H_{10}O$), 3,5-Dimethylcyclohexanone ($C_8H_{14}O$), Digoxigenin ($C_{23}H_{34}O_5$), Methyl 4-piperidineacetate ($C_8H_{15}N0_2$), 2',6'-Dihydroxyacetophenone ($C_8H_8O_3$).

Table 1. Percentage mortality of mosquito larvae of *An. stephensi* and *Cx. quinquefasciatus* exposed to different concentrations of different solvent leaf extracts of *P. granatum*.

Extracts	*An. stephensi*		*Cx. quinquefasciatus*	
	Concentration (ppm)	mortality±SD[a]	Concentration (ppm)	mortality±SD[a]
Hexane	Control	0.00±0.0[f]	Control	0.00±0.0
	50	4.2±1.7[a]	50	5.6±1.8[a]
	100	10.6±2.6[a]	100	16.2±2.2[a]
	150	25.8±2.7[ab]	150	37.8±1.7[ab]
	200	48.6±2.6[bc]	200	56.8±2.2[c]
	250	76.8±2.7[cd]	250	85.8±1.7[d]
Ethyl acetate	Control	0.00±0.0	Control	0.00±0.0
	50	15.8±2.4[a]	50	18.8±3.8[a]
	100	32.6±3.1[ab]	100	36.2±2.1[ab]
	150	54.4±2.3[bc]	150	57.8±3.6[c]
	200	76.4±2.3[cd]	200	78.6±3.5[cd]
	250	97.2±1.7[e]	250	99.2±0.8[e]
Chloroform	Control	0.00±0.0	Control	0.00±0.0

	50	6.8±2.6[e]	50	12.2±2.6[a]
	100	17.8±2.4[a]	100	26.4±3.2[ab]
	150	39.6±2.5[ab]	150	48.8±2.2[bc]
	200	60.8±2.5[c]	200	69.4±2.5[cd]
	250	86.6±2.1[d]	250	90.8±2.1[de]
Ethanol	Control	0.00±0.0	Control	0.00±0.0
	50	19.2±3.2[a]	50	24.8±2.7[a]
	100	37.4±2.5[ab]	100	42.6±2.6[b]
	150	58.2±2.6[c]	150	64.2±3.8[c]
	200	78.8±3.9[cd]	200	86.6±1.9[d]
	250	98.4±1.6[e]	250	100±0.00[e]

[a] Values are mean ± SD of four replicates. Within each row, different letters indicate significant differences (ANOVA, Duncan's new multiple range method test)

Table 2. LC_{50}, LC_{90}, slope, regression and chi squire analysis of larvicidal activity of *P. granatum* extracts against *An. stephensi* and *Cx. quinquefaciatus*

Species	Extracts	LC_{50} (mg/L)	95% Confidence limits		LC_{90} (mg/L)	Slope	Regression	χ^2
			LCL	UCL				
An. stephensi	Hexane	198.07	187.40	210.27	299.88	3.356793	y=0.919x+1.829	1.04[a]
	Ethyl acetate	134.71	124.65	144.46	232.83	3.775939	y=3.864x+1.140	4.30[a]
	Chloroform	171.27	161.33	181.78	271.49	3.52617	y=1.625x+1.604	1.05[a]
	Ethanol	125.78	115.25	135.71	225.98	3.832026	y=4.732x+1.022	5.35[a]
Cx. quinquefaciatus	Hexane	176.49	166.57	187.12	275.71	3.578278	y=1.354x+1.709	1.82[a]
	Ethyl acetate	126.68	116.43	136.42	224.34	4.100648	y=4.592x+1.041	7.00[a]
	Chloroform	151.06	140.70	161.49	256.75	3.366214	y=2.955x+1.266	1.13[a]
	Ethanol	110.36	106.34	129.18	212.28	3.987976	y=6.053x+0.889	6.04[a]

Values represent mean of five replications. Mortality of the after 24 h of exposure period LC_{50}= Lethal Concentration brings out 50% mortality and LC_{90}= Lethal Concentration brings out 90% mortality. LCL= Lower Confidence Limit, UCL= Upper Confidence Limit, χ^2 = Chi-squire,

[a] Significant at $p<0.05$

Table 3. Repellent activity of the *P. granatum* extracts against *An. stephensi* and *Cx. quinquefasciatus* at 3.5 mg/cm^2

Species	Extract	% of repellency					
		Time post application of repellent (min)					
		40	80	120	160	200	240
An. stephensi	Hexane	100±0.00	100±0.00	96.6±1.94	87.6±2.30	77.2±2.16	66.2±1.48
	Ethyl acetate	100±0.00	100±0.00	100±0.00	100±0.00	100±0.00	96.8±2.48
	Chloroform	100±0.00	100±0.00	100±0.00	96.8±1.30	86.2±3.27	75.2±1.64
	Ethanol	100±0.00	100±0.00	100±0.00	100±0.00	100±0.00	100±0.00
Cx. quinquefasciatus	Hexane	100±0.00	92.8±2.16	83.2±2.68	72.2±2.16	61.6±2.30	49.2±2.28
	Ethyl acetate	100±0.00	100±0.00	100±0.00	100±0.00	100±0.00	92.8±2.38
	Chloroform	100±0.00	100±0.00	94.4±1.81	84.2±2.38	72.6±2.60	61.8±2.48
	Ethanol	100±0.00	100±0.00	100±0.00	100±0.00	100±0.00	100±0.00

Values represent mean ± SD of the five replications

Table 4. List of identified phytocompounds in the *P. granatum* ethanol leaf extract

Peak	Compounds	MF	MW	RT(min)*	Concentration (%)
1	Cardanolide	$C_{23}H_{36}O_2$	344.539	12.17	9.75
2	N-Boc-4-piperidineacetaldehyde	$C_{12}H_{21}NO_3$	227.304	13.26	2.43

3	4-Cyclopropylbenzaldehyde	$C_{10}H_{10}O$	157.213	14.2	26.82
4	3,5-Dimethylcyclohexanone	$C_8H_{14}O$	126.199	14.45	21.95
5	Digoxigenin	$C_{23}H_{34}O_5$	390.52	15.33	2.12
6	Methyl 4-piperidineacetate	$C_8H_{15}N0_2$	146.189	16.19	34.14
7	2',6'-Dihydroxyacetophenone	$C_8H_8O_3$	152.149	17.9	4.87

*RT- Retention Time (min), MF- Molecular Formula, MW- Molecular Weight.

The parasite is transmitted by the bite of an infective female Anopheles mosquito. *P. falciparum* and *P. vivax* species cause the most contaminations around the world [40]. The results of present investigation showed that pure compound Methyl 4-piperidineacetate was more than 2-fold highest active than ethanolic leaf extract of *P. granatum* in larvicides, and repellent against *An. stephensi* and *Cx. quinquefasciatus*. The present investigation is comparable with some of other reports that the LC_{50} and LC_{90} values of 85.44 and 159.73 mg/L, from citronella component from *Melissa officinalis* were tested against *An. stephensi*. In the same way, highest larvicidal activity (LC_{50} values) were 136.75, 140.56, 144.90 and 149.89 mg/L for *Ageratina adenophora* ethyl acetate extract with I, II, III, IV instar larvae of *Cx. quinquefasciatus* [39]. Further, ethanol fractions of *Eichhornia crassipes* displayed the larvicidal and pupicidal activity against *Cx. quinquefasciatus* analyzed to solvent extracts and fractionates with LC_{50} values were 71.43, 94.68, 120.42, 152.15 and 173.35 ppm for first, second, third, fourth instars and pupae respectively. Presences of metabolites like flavonoides, alkaloids, anthroquinones and anthocyanins in the proved extracts might be the reason for the larvicidal and pupicidal action of the plant extracts and fractions of water hyacinth. The plant realm is considered as an asset for various kinds of potential drugs. In ancient days, many of the diseases were cured using plant products [39, 41-43]. Repellent action was not exhibited by these extracts at the tested concentrations. In potential, *Eichhornia crassipes* aquatic extract was successful in the control of the filarial vector, *Cx. quinquefasciatus* [44]. The bioactive compounds have been utilized to the development of environmentally safe vector managing agents. The extracts from aromatic plants are rising as possible mosquito vector control agents, since there are cheap, easy to administer and with hazard free properties [45, 46]. The compounds were eugenol, α-pinene and β-caryophyllene from *Plectranthus barbatus*. It is appeared to be most effective against *An. subpictus* (LC_{50}= 25.45, 32.09 and 41.66 µg/ml), followed by *Ae. albopictus* (LC_{50}= 28.14, 34.09 and 44.77 µg/ml) and *Cx. tritaeniorhynchus* (LC_{50}= 30.80, 36.75 and 48.17 µg/ml) [47].

Among the tested compounds, eucalyptol (1,8-cineole) and α-terpinyl acetate were considered to be inactive as the LC_{50}>50.0 mg L^{-1} [48]. Larvicidal leaf extract of *Gymnema sylvestre* showed the highest mortality in the concentration of 1000 ppm against *An. subpictus* (LC_{50}=166.28 ppm) and the maximum efficacy was observed in gymnemagenol compound isolated from *Gymnema sylvestre* petroleum ether extract with LC_{50} values against *An. subpictus* at 22.99 ppm and *Cx. quinquefasciatus* at 15.92 ppm, respectively [49]. The investigated compounds were β-pinene, sabinene, germacrene D, estragole and linalool in *Clausena anisata* against *An. stephensi, Ae. aegypti* and *Cx. quinquefasciatus* with LC_{50} values range from 11.01 to 42.28 µg/ml [50].

The phytochemical components and larvicidal activity to confirm the presence of various photochemical was studied for glycosidase, saponin, fixed oil & fats, protein, carbohydrates and tannin. The most effective larvicidal activity with concentrations 0.4% *Cassia tora* extracts gave 80% mortality in the larvae of *An. Stephensi* [51]. The larvicidal action of components of essential oils against mosquito species is due to the monoterpenes β-asatone, ρ-cymene, (+)-limonene, linaly acetate, myrcene, α-phellandrene, (+)-β-pinene, (-)-β-pinene, γ-terpinene and terpinolene, α-terpinene, phenylpropenes safrole and eugenol, and the sulfur containing compound daillyl disulfide on one or more species of mosquitoes[52]. Compounds were limonene, cis-carveol and carvone from *Mentha spicata* against *Cx. tritaeniorhynchus, Ae. albopictus, An. subpictus* and LC_{50} values range from 9.82 to 36.33 µg/ml [52-54] reported that major phytochemical compound, phytol isomer in chloroform extract of

Terminalia chebula leaf, which have potential mosquito larvicides and pupicides on *Cx. Quinquefasciatus* [55-57].

In conclusion, generally, this research provides useful information for the safer mosquito control properties and development of newer ones. Concerning the composition of the *P. granatum* ethanol extract, it was mainly composed by Methyl 4-piperidineacetate compound. Mosquitocidal activity clearly noticed the toxicity of *P. granatum* ethanol extract against *Cx. quinquefasciatus* larvae, even at low dosages. Further studies needed to validate and develop efficient mosquito larvae and adults with least impact on human health and environment.

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Citrus limetta (Risso) - borne compound as novel mosquitocides: Effectiveness against medical pest and acute toxicity on non-target fauna

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ABSTRACT

In the present study, *Citrus limetta* major phyto-compound (MPC) was identified from leaf methanolic extract (LME) through different spectral analysis such as GC–MS, Mass spectrum and FT-IR. The MPC of Corynan-17-0l,18,19-didehydro-10-methoxy-,acelate (ester) isolated from LME was tested with various concentrations (10–90 μg/mL) against 3rd instars mosquito larvae of *Aedes albopictus, Anopheles maculatus* and *Culex mimulus* under laboratory condition by using the standard protocol. GC–MS analysis of LME revealed the presence of six phyto-compounds of which, Corynan-17-0l,18,19-didehydro-10-methoxy-,acelate (ester) ($C_{22}H_{28}N_2O_3$) was found as MPC (39.01%). MS analysis supports the existence of MPC. Further, FTIR spectral data was used to find and confirm the exact functional group(s). The LC_{50} values of LME and MPC were recorded as 15.56, 13.72, 11.45 and 88.21, 86.49 and 79.72 μg/mL respectively. Moreover, less susceptible and higher suitability index were noticed on aquatic non-target fauna (NTF). We hypothesized that *C. limetta* LME and MPC of Corynan-17-0l,18,19-didehydro-10-methoxy-,acelate (ester) could be the important element, responsible for the larval toxicity. This suty promises an eco-friendly approach for the control of human vector mosquitoes (HVMs) and least toxic effects on non-target fauna (NTF).

1. Introduction

Many species of mosquitoes spread different types of pathogenic diseases to animals and humans, they are very serious and invariably disease-transmitting small arthropodan enemy to higher blood yielding vertebrates which cause millions of deaths every year (WHO, 2014). Mosquito bites induce different types of hypersensitivity responses in humans including skin irritation, itching sensation and swellings. In a tropical country such as India, the population of 40 million people, are greatly affected by mosquito bites and mosquito born diseases (MBDs) (Ghosh et al., 2012; Krishnappa et al., 2012; Baranitharan et al., 2019). MBDs are more endemic in 100 countries including India. Particularly, in India MBDs are increasing every year, the major reasons are rapid growth of human population, lack of knowledge/poor awareness about mosquito and MBDs, drastic environmental changes, limited funds allocation for mosquito control program (MCP), and on top of that mosquito develops resistance to the synthetic chemical pesticides (SCPs) (Billingsley et al., 2008). *Aedes albopictus* is a significant vector and transmission of many human diseases (chikungunya, dengue yellow fever and zika) are widespread in many tropical and sub-tropical countries (Pancharoen et al., 2002; Chen and Wilson, 2010; WHO, 2016). WHO estimated about 50 million people were seriously impacted by dengue fever across the world in 2008 (WHO, 2009). Anopheles maculatus is a primary vector of malaria and is widespread on the Indian subcontinent as well as Southeast Asia to Taiwan. This mosquito prefers areas around human settlements and they predominately lay their eggs in clean water bodies like lakes, swamps, ditches, wells, ponds, pools, streams, seepages, agricultural fields, foot and wheel prints and tree holes (WHO, 2010, 2014; Mathalaimuthu 2015, 2016; Mathalaimuthu 2017; Damrongpan et al., 2018). *Culex vishnui* is a important vector of Japanese encephalitis (JE) which breed prolifically in India and other tropical countries and they prefer to lay eggs in different types of freshwater containers (Colless, 1957;

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Das et al., 2006; Haoues et al., 2007; Someshwar et al., 2012). Around 3 billion of the world populations are seriously affected by JE, In India 50,000 cases and 15,000 deaths were recorded annually (Kabilan et al., 2004). Certain natural hosts such as domestic and wild animals act as reservoir of the deadliest virus (Banerjee and Chandra, 2004; Keiser et al., 2005).

Continuous exposure of SCPs causes more side effects on human health, such as immune dysfunction, cancer and birth defect (Rajasingh et al., 2017). The uses of biodegradable PCs are best alternatives medicine for HVMs control programmes and less harmful effect on natural ecosystem (Baranitharan and Dhanasekaran, 2014; Baluselva et al., 2012; Rawani et al., 2010). The phytoproducts have been used as traditional methods in several countries to solve the various types of MBDs/ parasites diseases which can be treated by different parts of plants origin like rhizome, bark, leaf, flower, seed, fruit etc., (Krishnappa and Elumalai, 2014 & 2015) as well as the phyto-compounds have varieties of entomotoxic values (Elumalai et al., 2012 & 2013). *Citrus limetta* (Rutaceae) essential oil (EO) utilized for industrial advantages and which has uses in food preparation (Anwar et al., 2008) and its chemical compounds (CCs) has therapeutic value and is used in many countries as remedies of human diseases (Nostro et al., 2000). The *C. limetta* seed CCs consist of α-thujene, α-pinene, camphere, sabinnene myrcene, α-teroinene, ρ-cymene, β-pinene, linalool, D-limonene and last terpene was the most common components. The rich CCs of flavonoids, vitamin C, coumarins and bergapten are used to sensitise the skin to sunlight (Mondello et al., 2003). To the best of our knowledge there was no information is available on mosquitocidal and suitability index of different NTF of *C. limetta* and its MPC.

2. Materials and methods

2.1. Plants collection and extraction method

The fresh plant leaf sampling was carried out during the growing season of July - October 2016 in and around Pagasalai village (11° 17′N 11° 24′N latitude and 79° 71′E to 80° 45′E longitude), Sirkali Taluk, Nagapattinam District, India. Plant leaves were air-dried at room temperature 28±2 °C.A sample was kept in a dark place and another sample was ground to a fine powder with the help of an electrical blender. The leaf powder was loaded in Soxhlet apparatus and extracted with high polarity methanol solvent. Later, collected extract was allowed to condense by Rotary vacuum evaporator. The condensed leafs extract was stored in refrigerator (<4 °C) until required for investigation for bioassays.

2.2. Mass spectrum analysis

Gas Chromatography-Mass Spectroscopy (GC–MS) was performed using a mass detector Turbo mass gold-Perkin Elmer with Elite-5MS (5% Diphenyl/ 95% Dimethy poly siloxane) slender segment. The stove temperature was modified from 50 to 280 °C at the rate of 5 °C min^{-1} and blocked at this temperature for 36 min. The delta and interface temperatures were 250 and 280 °C, respectively. The carrier gas was raised to a stream rate of 1.0 mL min^{-1} (consistent stream). The sample (2 μl) was inserted at a split of 10:1. Electron sway mass spectrometry was passing on at 70 eV. Elements source and fourfold temperature were kept up at 250 and 200 °C separately (Kumaravel et al., 2010). Mass spectra were reported at the Department of Instrumentation, Indian Institute of Technology using a Manchester Micromass PLATFORM II (ES) and Termo Finnigan MAT95XP (Accurate mass) instruments. Mass spectrometry gives the pair molecular weight and fragmentation arrangement of the components. It depends on the making of ions from a parent compound and the subsequent characterization of the pattern that are produced.

2.3. Fourier transform-infrared spectroscopy

FT-IR is used to probe bond vibrations and bending in molecules and to reveal the types of functional groups present in compound. Functional group region is in the range of 3600–650 cm^{-1} and finger print region is from 1625–650 cm^{-1} (Vivek et al., 2011).

2.4. Larval toxicity

The larvicidal activity of *C. limetta* LME and MPC was evaluated as per standard method (WHO, 2005). The whole bioassay test was analyzed between the doses ranged of 10 - 120 μg/mL and the selected doses were tested on early third instars larvae (0–6 h old) of the target mosquitoes. The phyto-compound were dissolved in 1 mL dimethyl sulfoxide (DMSO) and taken in 249 mL of dechlorinated water. Each test species, 25 larvae used for single concentration and replicated five times, larval mortality was recorded after 24 hrs of post treatment. Percent mortality was rectified for control mortality and calculated by using the Abbott's (1925) formula. The LC_{50} values, chi-square values and other statistical values were calculated by using probit analysis described by Finney (1971).

2.5. Target medical vectors

The eggs/eggs raft, larvae/pupae of target medical vectors, such as *Ae. albopictus, An. maculates* and *Cx. mimulus* were collected from the Kodaikanal Wildlife Sanctuary (protected area), Theni District, Tamilnadu, India. The collected eggs/larvae/ pupae of mosquitoes were identified by ICMR-centre for Research in Medical Entomology, Chinna Chokkikulam, Madurai, Tamil Nadu 625,002 and brought to laboratory for continuous rearing. The larvae were fed on dog biscuits and yeast powder in the 3:1 ratio. Adults were provided with 10% sucrose solution and 1-week-old chick for blood meal. Mosquitoes were held at 27 ± 2 °C, 75±5% RH, with a photo period of 12L: 12D.

2.6. Biotoxicity on non-target organisms

The effect of non-target fauna (NTF) was assessed by following the method of Sivagnaname and Kalyanasundaram (2004). The toxicity of *C. limetta* LME and MPC were tested against aquatic NTF like *Acilius sulcatus, Diplonychus indicus* and *Anisops bouvieri* that were carefully collected and separately maintained in the aerated transparent glass container (100 cm diameter and 30 cm depth) containing water at 27 ± 2 °C; the external relative humidity was 75±5%. The test was replicated five times, NTF were observed for mortality and other abnormalities such as sluggishness, reduced swimming activity and insects flying activity after 48 h exposure. The exposed NTF were also observed continuously for 15 days to understand the post-treatment effect of LME and MPC.

2.7. Statistical analysis

The average mortality data were subjected to probit analysis for calculating LC_{50}, LC_{90} and other statistics at 95% confidence Limit by using IBM -SPSS Statistics version 25.0, results with $p \leq 0.05$ were considered to be statistically significant. In experiments evaluating the biotoxicity of NTF, the Suitability Index was calculated for each NTF using the following formula (Deo et al., 1988).

3. Results

3.1. GC–MS analysis and percentage of PCCs

GC–MS spectral analysis of LME was identified seven PCCs and their % concentrations were displayed in (Table 1 and 2). A total of 7 PCCs representing 100% the MPC of *C. limetta* was Corynan-17-ol,18,

19-didehydro-10-methoxy-,acelate (ester) (39.01%) followed by Pentadecanoic acid, 14- ethyl-,methylester (25.27), Cyclohexanone,2,2-dimethyl1-5-(methoyloxinyl)-,(2á(R^x),3 á]-(+-)- (17.58), Curan, 16,17,19,20-tetradehydro- (5.49), á –Patchoulane (4.39), 14-Octacenoic acid, methyl ester (6.07%) and Isopropyl stearate (2.19). The observed m/z value 368.47 for Corynan-17-ol,18,19-didehydro-10-methoxy-,acelate (ester) (39.01%) compound is consistency with the proposed molecular formula ($C_{22}H_{28}N_2O_3$), Besides, the mass spectral studies also confirmed the MPC (Figs. 1 and 2). The oxygen, hydrogen and carbon atoms of the MPC were indicated the different colors (Red, greenish blue and gray) in Figs. 1 and 2 represent the two dimensional (2D) structure of the MPC. Whereas, Fig. 3a and b shows the three dimensional (3D) structure of MPC identified from the selected plant.

Table 1
Phyto-components identified in the *C. limetta* LME using GC–MS.

Peak	CN	RT	CP	Mode of Identification
1	Cyclohexanone,2,2-dimethyl1-5-(methoyloxinyl)-,(2á(R^x),3 á]-(+-)-	16.32	17.58	RI, MS
2	á -Patchoulane	16.77	4.39	RI, MS
3	Pentadecanoic acid, 14-methyl-, methylester	17.47	25.27	RI, MS
4	14-Octacenoic acid, methyl ester	19.3	6.07	RI, MS
5	Isopropyl stearate	21.13	2.19	RI, MS
6	Curan, 16,17,19,20-tetradehydro-	23.4	5.49	RI, MS
7	Corynan-17-0l,18,19-didehydro-10-methoxy-,acelate (ester)	27.8	39.01	RI, MS

CN: Compounds Name; RT: Retention Time; CP: Composition Percentage; RI: Retention Index; MS: Mass Spectra.

Table 2
Mass spectra values of Corynan-17-ol,18,19-didehydro-10-methoxy-,acelate (ester).

Phyto-compound	MF	MW	*m/z*
Corynan-17-ol,18,19-didehydro-10-methoxy-,acelate (ester)	$C_{22}H_{28}N_2O_3$	368.21	368.47

MF: Molecular Formula; MW: Molecular Weight.

3.2. FT-IR analysis of LME

FT-IR analysis was accomplished to identify the functional groups of LME and spectrum indicated the clear peaks with (3372, 2919, 1631, 1436, 1317, 1230, 1099, and 634 cm^{-1}) different values. In spectrum, strong and broad absorption band checked at 3300 cm^{-1} in the IR spectrum is commonly accredited to N—H mode of the secondary amine. In the compounds *C. limetta*, characteristic IR band that appeared at 3422 cm^{-1} (strong and sharp) is due to the N—H stretching of the indole ring. The absorption bands in the region 2555–2924 cm^{-1} (strong) are due to aromatic and aliphatic C—H stretching frequencies. IR spectra of the compound *C. limetta* shown the presence of carboxylic acid, alkenyl groups C=O, C=C stretching (strong) frequency observed at 1735, 1625 cm^{-1}. A strong absorption band that appeared at 1065 and 1096 cm^{-1} was due to furan C—O stretching. The observed amine, aliphatic and aromatic C—H stretching frequencies are evidences for the presence of compound in the *C. limetta* (Fig. 4). The functional groups such as alcohols, phenols, 1*,2* amines, amides, aromatic, aliphatic, carboxylic acid, alkenyl, esters, ethers, amine confirmed their presence in methanol extract.

3.3. Larvicidal activity of LME and MPC

The larvicidal activity of LME and MPC were tested against three different vector mosquitoes *Ae. albopictus, An. maculates* and *Cx. mimulus*. Furthermore, LC_{50} values of LME showed the higher activity represented against *Cx. mimulus* followed by *An. maculates* and *Ae. albopictus* were 79.72, 86.49 and 88.21 μg/mL respectively. The LC_{50} values of MPC against freshly moulted 3rd instar larvae of *Ae. albopictus, An. maculates* and *Cx. mimulus* were 15.56, 13.72 and 11.45 μg/mL respectively. The chi-square values are statically significant at $p \leq 0.05$ level and LC_{90} LCL, UCL, Regression value and degrees of freedom of larvicidal activity were clearly displayed in Table 3.

3.4. Biotoxicity on aquatic NTF

The toxicity of LME and MPC against aquatic NTF are shown in Table 4. *A. sulcatus, D. indicus* and *A. bouvieri* were the least susceptibility, with LC_{50} values ranging from 1520.42 to 4133.63 μg/mL. LME

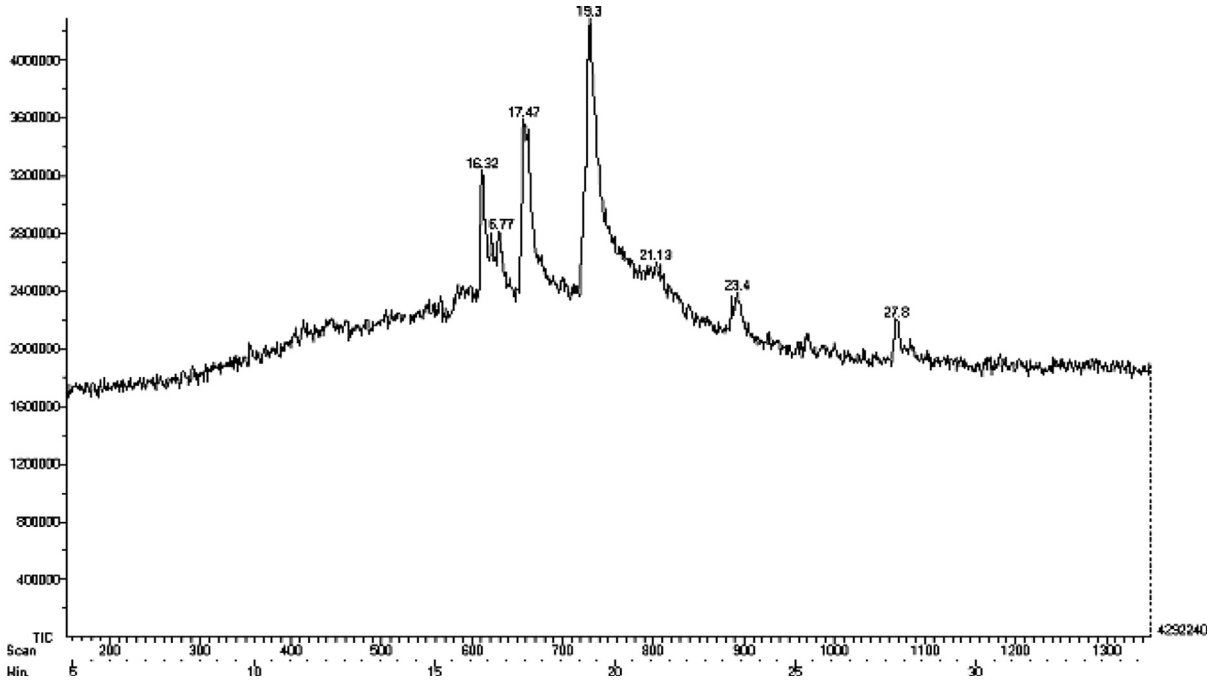

Fig. 1. GC–MS chromatogram of *C. limetta* LME. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

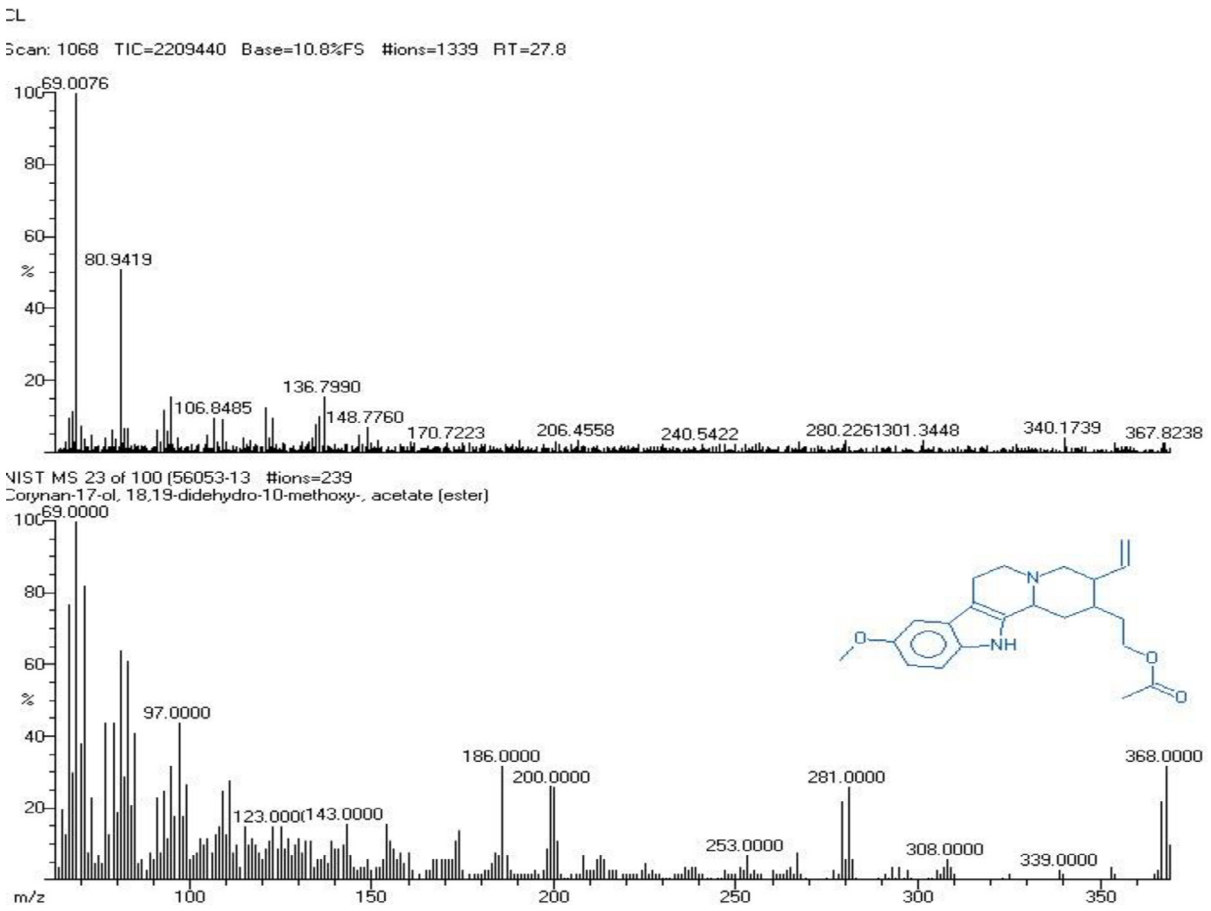

Fig. 2. Mass spectrum and structure of phyto-compound (Corynan-17-ol,18,19-didehydro-10-methoxy-,acelate(ester)) identified by GC–MS in the *C. limetta* LME. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

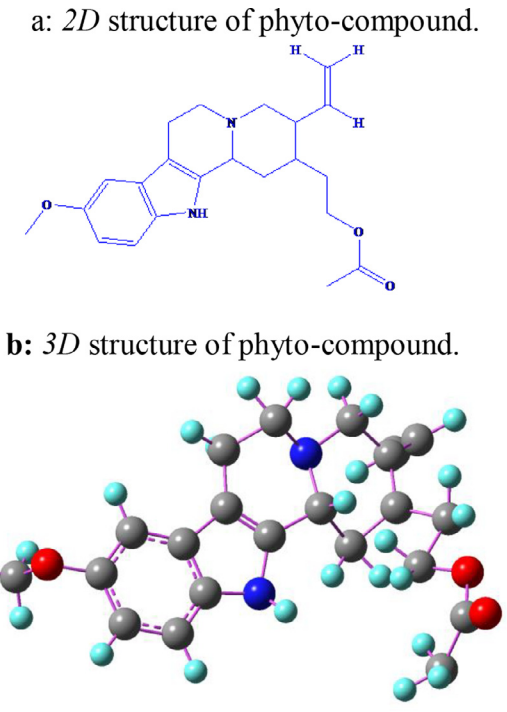

Fig. 3. a and b The phyto-compound Corynan-17-ol,18,19-didehydro-10-methoxy-, acelate (ester) identified in *C. limetta* LME. (Exact mass: 368.21; Molecular Weight: 368.47). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

had less susceptible to NTF than *C. limetta* MPC, Corynan-17-ol,18,19-didehydro-10-methoxy,acelate(ester).

3.5. Suitability index of NTF

The suitability index of different aquatic NTF over young instars of selected HVMs, exposed to phyto-compounds (PCs). The suitability index indicated that LME and MPC are less harmful to the NTF (Tables 5). Survival, swimming and flying activity of the test species were not significantly changed. Moreover, same PCs were exposed to HVMs which produced maximum LC_{50} and LC_{90} values. Our findings threw more light on the possible utilization of LME and MPC as mosquitocidal agents against the *Ae. albopictus, An. maculates* and *Cx. mimulus*. More precisely, the isolated and identified MPC, Corynan-17-0l,18,19-didehydro-10-methoxy-,acelate (ester) exerts threefold activity than the LME against the selected HVMs.

4. Discussion

The present investigation results are comparable with some of the earlier reports. Earlier, Sarita et al. (2012) *C. limetta* peel hexane extract reported higher larvicidal potential against *An. stephensi* and the phytochemical study of said extract showed the presence of terpenoids and flavonoids . Correspondingly, the results of the present studies are on par with previous reports that the 15 percent *C. limetta* oil exhibited highest percentage of larval mortality and knockdown effects as elucidated by Prakash Rao et al. (2016). Rosalinda et al. (2016) reported hexane extract of *C. grandis* peel tested for 3rd and 4th instar larvae of *Ae. aegypti* provided maximum activity. *C. limetta* oils also had prospective larval killing activity and have knockdown

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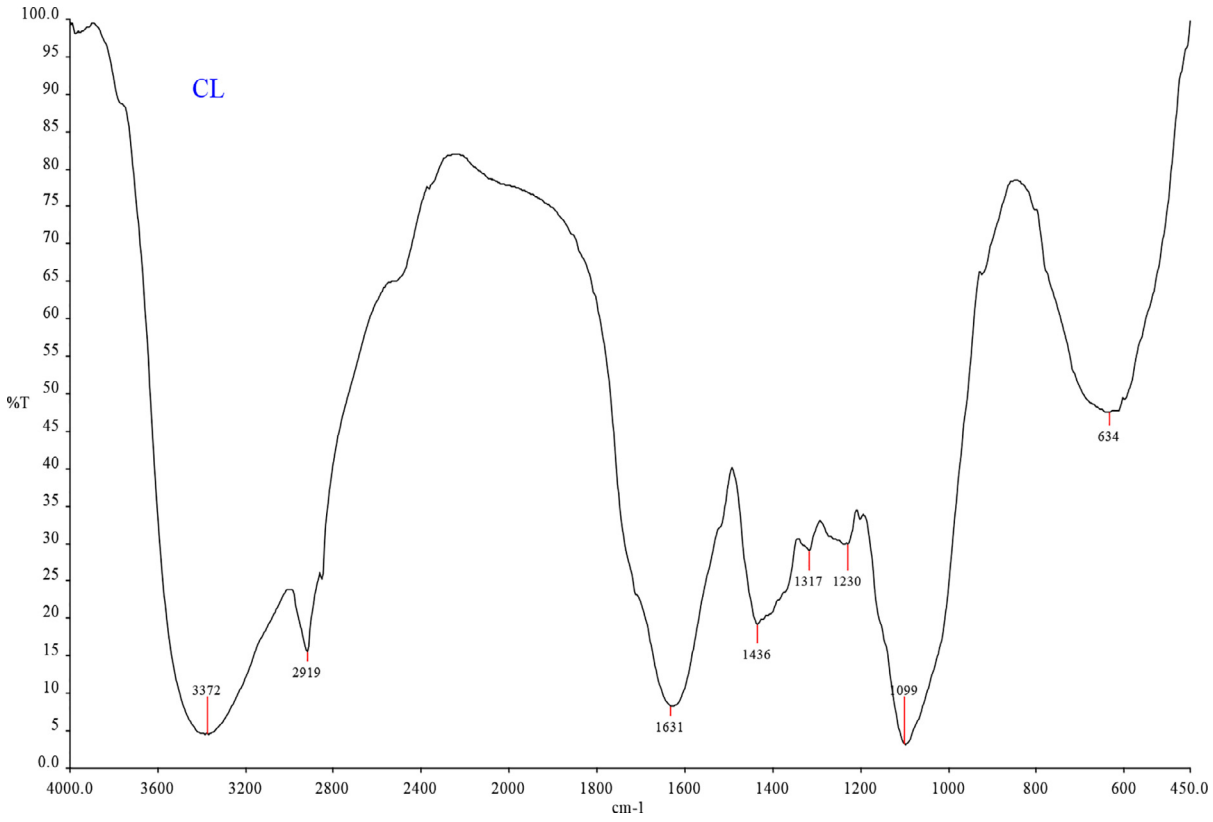

Fig. 4. Fourier transfer-infra red (FT-IR) spectrum of *C. limetta* LME. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 3
Larvicidal activity of *C. limetta* LME and its MPC against 0 - 6 h old third instar larvae of vector mosquitoes.

Target medical pests	LC_{50} (μg/mL)	95% Fiducial limit (μg/mL)		LC_{90} (μg/mL)	Slope	R value	df	χ^2
		LCL	UCL					
C. limetta LME								
Ae. albopictus	88.21	83.64	93.29	115.77	4.17592	$y = 2.2521x + 0.1248$	4	10.5371
An. maculatus	86.49	81.12	92.86	113.54	4.30679	$y = 2.6250x + 0.0352$	4	11.8204
Cx. mimulus	79.72	74.55	91.18	110.25	4.14210	$y = 2.2016x + 1.2673$	4	14.3723
C. limetta MPC, Corynan-17-ol,18,19-didehydro-10-methoxy-,acelate (ester)								
Ae. albopictus	15.56	12.72	17.38	26.30	4. 97,105	$y = 2.3260x + 1.3216$	4	8.7102
An. maculatus	13.72	11.91	16.32	25.75	4. 26,801	$y = 2.3045x + 1.4890$	4	7.6430
Cx. mimulus	11.45	10.82	14.60	21.21	4. 80,432	$y = 2.4231x + 0.5231$	4	8.4218

No mortality was observed in the control (without phytochemical), LC_{50} lethal concentration that kills 50% of the exposed organisms, LC_{90} lethal concentration that kills 90% of the exposed organisms, UCL 95% Upper Confidence Limit, LCL 95% Lower Confidence Limit, χ^2 Chi Square, d.f. degrees of freedom.

Table 4
Toxicity of *C. limetta* LME and its MPC against NTF sharing same ecological niche of selected HVMs.

Non –target fauna	LC_{50} (μg/mL)	95% Fiducial limit (μg/mL)		LC_{90} (μg/mL)	Slope	R value	df	χ^2
		LCL	UCL					
C. limetta LME								
A. sulcatus,	2678.32	2358.21	2895.83	4575.81	6.61373	$y = 3.5210x+1.5217$	4	9.8342
D. indicus	3664.81	2942.90	3871.88	6584.18	6.26040	$y = 3.2835x+1.3454$	4	10.6704
A. bouvieri	4133.63	3782.22	4425.83	8270.56	6.94721	$y = 3.6122x+1.6323$	4	11.7291
C. limetta MPC, Corynan-17-ol,18,19-didehydro-10-methoxy-,acelate (ester)								
A. sulcatus,	1520.42	1127.46	1741.58	3194.28	5.77308	$y = 3.7123x + 1.8291$	4	11.6822
D. indicus	1819.59	1526.84	1941.33	3485.32	4.84686	$y = 3.4305x + 1.7203$	4	10.6350
A. bouvieri	2146.83	1877.26	2364.26	4058.33	5.41974	$y = 3.2681x + 1.3542$	4	10.3022

No mortality was observed in the control (without phytochemical), LC_{50} lethal concentration that kills 50% of the exposed organisms, LC_{90} lethal concentration that kills 90% of the exposed organisms, UCL 95% Upper Confidence Limit, LCL 95% Lower Confidence Limit, χ^2 Chi Square, d.f. degrees of freedom.

fects (Manimaran et al., 2012; Mallik et al., 2016). The maximum mosquito larvicidal activity of n-hexane and petroleum ether solvent peel extracts of *C. limetta* against 4th instar larvae of *An. stephensi* and *Ae. aegypti* was reported by Kumar et al. (2012). The results of the present study is in agreement with the findings of Simon Oke et al. (2017) who have reported that, *Citrus* fruit peel and seeds were found to contain important phytogroups such as tannins, cardiac glycosides, saponins, flavonoids and terpenoid . Moreover, *Citrus* peel ethanolic

Table 5
Suitability index of different NTF over young instars of selected HVMs, exposed to *C. limetta* LME and its MPC.

Non –target fauna	*Ae. albopictus*	*An. maculatus*	*Cx. mimulus*
C. limetta LME			
A. sulcatus,	76.22	86.84	81.53
D. indicus	65.56	77.54	85.91
A. bouvieri	79.60	82.68	74.15
C. limetta MPC, Corynan-17-ol,18,19-didehydro-10-methoxy-,acelate (ester)			
A. sulcatus,	64.18	88.55	85.78
D. indicus	86.49	76.81	88.00
A. bouvieri	73.84	96.90	77.56

extracts found statically significant mosquitoicidal activity against *Anopheles* mosquitoes (Samta et al., 2013; Ihemanma et al., 2014). The MPC of limonene presence of different *citrus* plants like *C. aurantifolia, C. reticulate, C. limon* and *C. sinensis* (94.92%) and indicated 100% larval mortality were recorded (Azzous et al., 1976; Haro and Faas, 1985; Patrick Ntoinga et al., 2015; Manorenjitha et al., 2017). The similar study were observed from different activities of phytocompound 11-octadecenoic acid, methyl ester extracted from *C. aromaticus* leaf extract, citronellal component from *M. officinalis,* Saponin from methanol extracts of *Chlorophylum borivilianum,* phytol, 3,7,11-trimethyldodeca-1,6,10-trien-3-ol and 3,7,11-trimethyldodeca-6,10-dien-3-ol were isolated from *J. officinale* leaf extracts and *S. campanulata* leaf acetone extract were showed maximum mosquitocidal activities (Baranitharan et al., 2016; 2017; Deore and Khadabadi, 2009; Feng et al., 2015; Pravin et al., 2015).

Mosquito larvicidal effectiveness of LME and MPC were tested against selected HVMs which given excellent larvicidal effectiveness besides which a very low toxicity was observed on listed NTF and we achieved high suitability index and entirely safer for NTF. Earlier, some authors have been studied the toxicity PCs on aquatic NTF. Conti et al., (2014) reported that the *M. alternifolia* was tested against toxicity on aquatic NTF water flea, *Daphnia magna* which share same ecological niche of *A. albopictus*. The MPCs of *M. alternifolia* was investigated significant acute toxicity also towards adults of the non-target arthropod. Govindarajan et al. (2016) studied that the Indian medicinal plant *O. scabrum* and its MPCs tested against medically important HVMs which provided more toxic effect on mosquitoes larvae and less toxic effects were observed on aquatic NTF. The various studies have been reported on neem extracts and its PCs tested against aquatic NTF like *D. magna, I. bicolor rufa, Tipula* species, *P. dorsata, H. argus, Daphnia* species, *D. pulex, H. azteca,* and *C. riparius* (Scott and Kaushik, 1998; Kreutzweiser, 1997; Kreutzweiser et al., 2004; Saucke and Schmutterer, 1992; Stark, 2001). Recently, Roman Pavela (2018) reported that the F. vulgare EO tested against important polyphagous pest Myzus persicae and non target soil organism (natural predator) H. axyridis and E. fetida. The EO and its MPCs were provided excellent effectiveness against M. persicae though not causing any significant mortality/side effect on tested NTF. Since there was no previous research works and literature available about the larvicidal and suitability of NTF of *C. limetta* LME and MPC of these present investigations serve as firsthand information.

5. Conclusion

The results of the present study revealed that, the *C. limetta* LME and MPC Corynan-17-ol,18,19-didehydro-10-methoxy,acelate (ester) induced maximum larval mortality against selected medical pests. Consequently, the present approach should be encouraged in the vector control programme since it displayed meager harmful effects on ecologically important NTF. It is important to control the selected mosquitoes in the wild and around human settlements as they are serious threat the health of animals and humans

Declaration of competing interest

The authors declare that there is no conflict of interest

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Supplementary materials

Supplementary material associated with this article can be found in the online version at doi:10.1016/j.sajb.2019.11.014.

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THE DUTCH WINDMILL $D_3^{(M)}$ OF GALLAI FUZZY GRAPHS ON DOMINATION NUMBER

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Abstract - *In this paper, we discussed about the Dutch windmill graphs, the complete fuzzy graphs, the wheel of Gallai fuzzy graphs. Also analyses some theorems and domination number of that graphs.*

Index Terms: The Dutch windmill fuzzy, the complete fuzzy graph and the wheel of Gallai fuzzy graph, fuzzy domination number.

AMS Classification: 03E72, 05C99**.**

1. INTRODUCTION

. In 1965, L.A. Zadeh [7] introduced a mathematical structure to describe the concepts of vagueness in real life throughout the periodical of a determining paper. In 1975, A. Rosenfeld [4] introduced the information of fuzzy graph theoretic perception such as paths, cycles and connectedness. In 1996, Van Bang Le [6] discussed regarding the Gallai graphs and anti-Gallai graphs. Correspondingly, S. Aparna Lakshmanan and S.B. Rao [1] also deliberated the Gallai graphs and anti-Gallai graphs. Further, A. Somasundram and S. Somasundram [5] have explored the domination in fuzzy graphs. In accumulation, the domination, independent and irredundance numbers were discussed by A. Nagoorgani and P. Vadivel [2]. In [8], in our previous effort we have discussed the concept of Gallai-type theorems in Gallai Fuzzy Graphs on Domination parameters. The idea of Dutch windmill graph was discussed by M.R.Rajesh kanna, R.Pradeep kumar, and R.Jagadeesh [3]

In this manuscript, we discussed about the Dutch windmill, the wheel of Gallai fuzzy graphs and some theorems of this graphs. Also we analyses the structures and fuzzy domination numbers.

2. PRELIMINARIES

A fuzzy graph with G as the underlying set is a finite non-empty unordered pair of G= (σ, μ), where σ : V →[0,1] is a fuzzy subset, μ: E→[0,1] is a fuzzy relation on the fuzzy subset σ such that μ(u, v) ≤ σ(u) ∧ σ(v) for all u, v∈ V where ∧ and ∨ stands for minimum and maximum. The underlying crisp fuzzy graph of G=(σ, μ) is denoted by G^*=(V, E), where V ={ u ∈ V : σ(u) > 0}andE = {(u, v)∈ VxV : μ(u, v) > 0 },the fuzzy order P and fuzzy size of the fuzzy graph G=(σ, μ) are defined by p = $\Sigma_{v \in V}$ σ(u) and q =$\Sigma_{u, v \in E}$ μ(u, v). Each pair μ = u, v of fuzzy vertices in σ is a fuzzy edge of G and μ is said to join u and v are fuzzy adjacent vertices, fuzzy vertex u and fuzzy edge μ are fuzzy incident with each other as are σ and μ if two distinct fuzzy edges are incident with a common fuzzy vertex, then they are called fuzzy adjacent edges. A fuzzy edge e = uv of a fuzzy graph is an fuzzy edge if μ(u, v) = σ(u) ∧ (v). N (u) ={v ∈ V / μ (u, v) = σ(u) ∧ σ(v) } is called the open fuzzy neighborhood of u and N[u]= N(u)∪{u}is the closed fuzzy neighborhood of u.

Definition: 2.1

Let G be a fuzzy graph and u be a fuzzy vertex in G then there exists a fuzzy vertex v such that (u, v) is a fuzzy edge then we say that u dominates v.

Definition: 2.2

Let G = (σ, μ) be a fuzzy graph. A subset D of V is said to be fuzzy dominating set of G if for every v ∈ V-D, there exists u ∈ D such that u dominates v.

Definition: 2.3

A fuzzy dominating set D of a fuzzy graph G is called minimal fuzzy dominating set of G, if for every fuzzy vertex v ∈ D, D-{v} is not a fuzzy dominating set.

Definition: 2.4

The Minimum fuzzy cardinality among all minimal fuzzy dominating sets in Γ(G) is called fuzzy domination number of Γ(G) and is denoted by γ(Γ(G)).

Definition: 2.5

A fuzzy graph G is said to be connected if every pair of its fuzzy vertices are connected. Otherwise it is disconnected.

Definition: 2.6

Let G_i denote the induced fuzzy sub graph of G with fuzzy vertex set V_i .Clearly the sub graphs G_1, G_2,….G_n are connected and are called the fuzzy components of G

Definition: 2.7

A fuzzy graph G = (σ, μ) is complete fuzzy graph if μ(x, y) = σ(x) ∧ (y) for all x, y ∈ V.

Definition: 2.8

Let G : (σ, μ) be a fuzzy graph on G* (V,E). If $d_G(v) = k$ for all $v \in V$, that is if each vertex has same degree k, then G is said to be a regular fuzzy graph of degree k or k-regular fuzzy graph.

Definition: 2.9

In the mathematical discipline of graph theory, a wheel graph W_n is a graph with n vertices ($n \geq 4$) formed by connecting a single vertex to all vertices of an n-1 cycle. A wheel graph with fuzzy labelling is called a fuzzy wheel graph

Definition: 2.10

A fuzzy graph whose fuzzy edge set is empty, is called a null fuzzy graph or a totally disconnected fuzzy graphs.

3. THE DUTCH WINDMILL GRAPHS OF GALLAI FUZZY GRAPHS.

In this division, we describe on the Dutch windmill fuzzy graph, a wheel fuzzy graph of Gallai fuzzy graphs, and some significant theorems are analysed.

Definition: 3.1

The Dutch windmill fuzzy graph is denoted by $D_n^{(m)}$ and it is the fuzzy graph obtained by taking m copies of the fuzzy cycle C_n with a fuzzy vertex in common. The Dutch windmill fuzzy graph is also called as friendship fuzzy graph if n=3.Dutch windmill fuzzy graph $D_n^{(m)}$ contains (n-1) m+1 fuzzy vertices and mn fuzzy edges.

Definition: 3.2

The Gallai fuzzy graph Γ(G) of a fuzzy graph G has the fuzzy edges of G as its fuzzy vertices and two distinct fuzzy edges of G are fuzzy incident in G, but do not span a fuzzy triangle in G. The line fuzzy graph L (G) of a fuzzy graph G has the fuzzy edges of G as its fuzzy vertices and two distinct fuzzy edges of G are adjacent in L (G) if they are fuzzy incident in G. This concept of Gallai fuzzy graph will be apply to the Dutch windmill $D_3^{(n)}$, a wheel fuzzy and complete fuzzy graph . Also to analyses the construction of structures and fuzzy domination number of Gallai fuzzy graph.

Example: 3.3

Let us consider $D_3^{(2)}$ be a the Dutch windmill graphs or friendship graph which is called a butterfly fuzzy graphs.

Let us consider $D_3^{(2)}$ be a the Dutch windmill graphs or friendship graph which is called a butterfly fuzzy graphs.

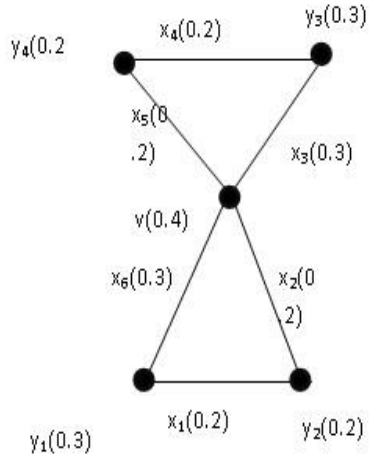


Fig: 3.3.1 ($D_3^{(m)}$), m=2 .

The domination number

$\gamma(D_3^{(m)}) = 0.4$

By using the definition of $\Gamma(D_3^{(2)})$ we have the following

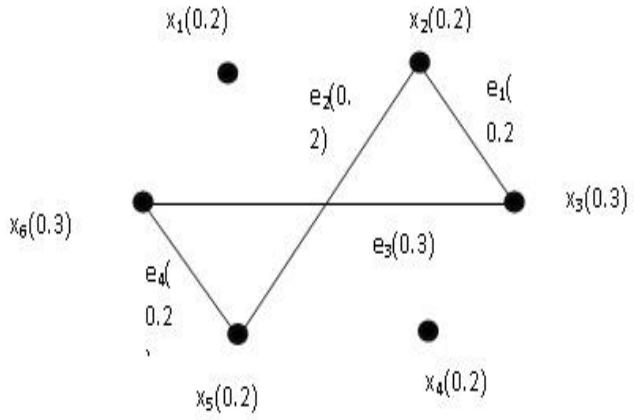


Fig: 3.3.2 Dutch windmill graph of Gallai fuzzy graph $\Gamma(D_3^{(2)})$.

The domination number $\gamma(\Gamma(D_3^{(2)})) = 0.8$

Since $\Gamma(D_3^{(m)}) = \frac{1}{2}\sum d_i^2 - x - 3t$, is the number of fuzzy edges, where $\sum d_i$ is degree of the vertex, t is the total triangle and x is number of edges in $D_3^{(m)}$, m=2 as shown in figure 3.3.1. Then $\Gamma(D_3^{(2)}) = (2^2+2^2+2^2+2^2+4^2) - 6 - 3(2) = 4$ fuzzy edges (0.2+0.2+0.3+0.2 = 0.9) .Here $\Gamma(D_3^{(2)})$ is disconnected graphs of Gallai fuzzy graph with 3 components.

Example: 3.4

We consider a wheel fuzzy graph w_4 is as shown below

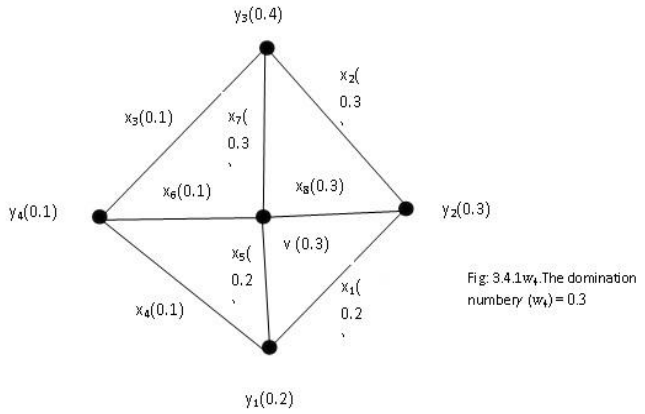


Fig: 3.4.1w_4.The domination numbery (w_4) = 0.3

By using the definition of $\Gamma(w_4)$ we have the following

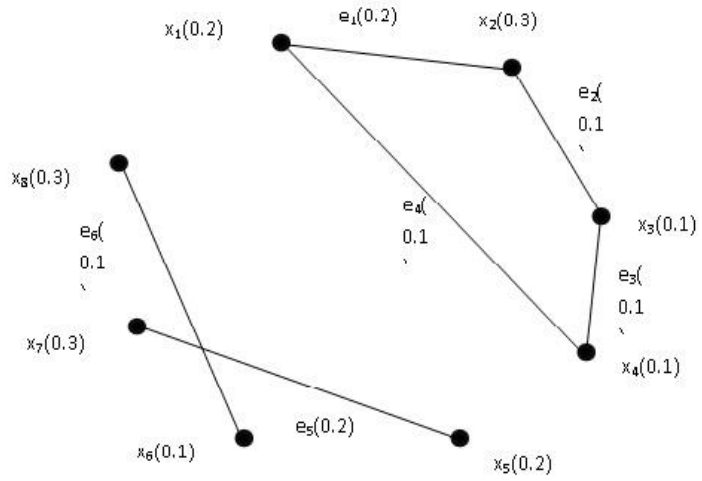


Fig: 3.4.2 $\Gamma(w_4)$ .The domination numbery ($\Gamma(w_4)$) = 0.5

Theorem: 3.5

Let $D_3^{(m)}$ be a Dutch windmill fuzzy graph contains m fuzzy triangles of G. Then $\Gamma(D_3^{(m)})$ is disconnected of Gallai fuzzy graph with m+1 number of components.

Proof : Let $D_3^{(m)}$ be a Dutch windmill fuzzy graph contains m fuzzy triangles of G.The fuzzy vertex set V ={v, y_1, y_2, …….y_{2m}}, and the fuzzy edge set E= {$x_1, x_2, \ldots x_{3m}$}. $D_3^{(m)}$ contains 2m +1 fuzzy vertices and 3m fuzzy edges, where m is a copies of the triangles.

Since every pair of its fuzzy vertices has exactly one common fuzzy neighbor in $D_3^{(m)}$.By the definition of the Gallai fuzzy graphs $\Gamma(D_3^{(m)})$,the number of edges in $\Gamma(D_3^{(m)})$ is $\frac{1}{2}\sum d_i^2 - x - 3t$, where $\sum d_i$ is degree of the vertex , t is the total triangle and x is number of edges in $D_3^{(m)}$. suppose $D_3^{(m)}$ contains m triangles, then its has m+1 components . Out of this m+1

component, one component is regular fuzzy graph and remaining m are null fuzzy graphs. Hence $\Gamma(D_3^{(m)})$ is disconnected of Gallai fuzzy graphs with m+1 numbers of components

Example: 3.6

The Dutch windmill fuzzy graphs $D_3^{(m)}$ $m \geq 2$ is as shown below

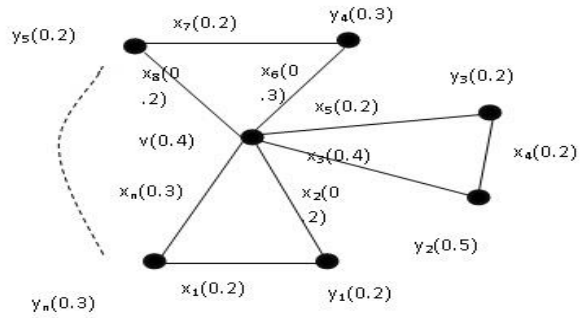


Fig: 3.6.1$D_3^{(m)}$. The domination number
$\gamma(D_3^{(m)}) = 0.4$

The Dutch windmill of Gallai fuzzy graph $\Gamma(D_3^{(m)})$ is as shown below

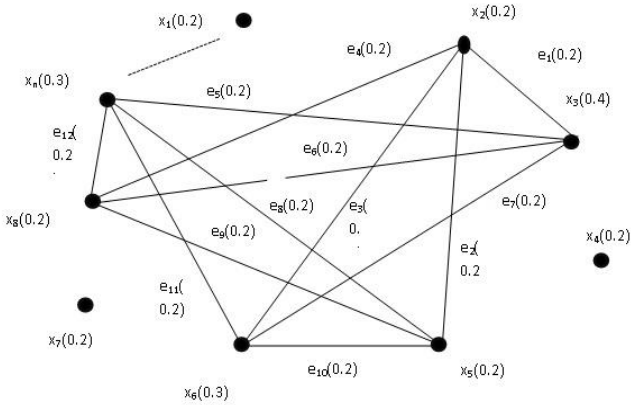


Fig: 3.6.2. Here $\Gamma(D_3^{(m)})$ is disconnected fuzzy graphs with m+1 number of components .
The domination number$\gamma(\Gamma(D_3^{(m)})) = 1.0$.

Theorem: 3.7

Every friendship fuzzy graph is a windmill of Gallai fuzzy graph but the converse is not true.

Theorem: 3.8

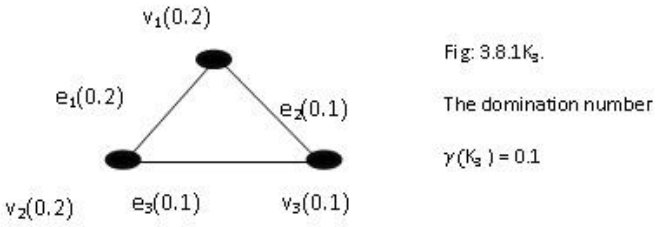
Let K_n be a complete fuzzy graph contains triangles of G. Then $\Gamma(K_n)$ is a totally disconnected fuzzy graph with n numbers of components.

Proof:

Let K_n be a complete fuzzy graphs of G. Prove that $\Gamma(K_n)$ is a totally disconnected fuzzy graph with n numbers of components. Every pair of edge are adjacent in K_n. But each edge V_iV_j in K_n is the triangle of Kn. By the definition of the Gallai fuzzy graph $\Gamma(K_n)$, $|\Gamma(V(Kn))| = |E(Kn)|$ are n vertices. The number of edges in $\Gamma(k_n)$ is $\frac{1}{2}\sum d_i^2 - x - 3t$, where $\sum d_i$ is degree of the vertex, t is the total triangle and x is number of edges in k_n. Since each fuzzy edge of the graph K_n forms triangles. Hence $\Gamma(Kn)$ is a totally disconnected of fuzzy graph with n numbers of components.

Example: 3.9

The complete fuzzy graph K_n, $n \geq 3$ with n=3 vertices is as shown below



The complete Gallai fuzzy graph is as shown below

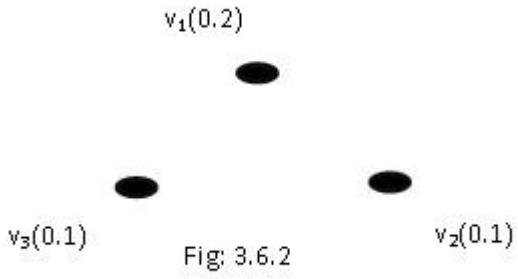


Fig: 3.9.1 Here $\Gamma(K_3)$ is a totally disconnected fuzzy graphs with n components. The domination number γ $(\Gamma(K_3))$ = $\overline{K}_3$ = 0.4.

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Anti Gallai Fuzzy Graphs in Corona Graphs

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Abstract

In this paper, we investigate about the anti Gallai fuzzy graphs ***(Δ(G))*** *in corona graphs. Also we have analyzed some theorems, structure and domination number of that anti - Gallai graphs.*

Key words**:** *Corona fuzzy graphs; Anti –Gallai fuzzy graphs; Domination numbers*.

AMS Classification: *03E72, 05C99***.**

1. Introduction

In 1965, L.A. Zadeh [7] introduced a mathematical structure to describe the concepts of vagueness in real life throughout the periodical of a determining paper. In 1975, A. Rosenfeld [4] introduced the information of fuzzy graph theoretic perception such as paths, cycles and connectedness. In 1996, Van Bang Le [6] discussed regarding the Gallai graphs and anti-Gallai graphs. Correspondingly, S. Aparna Lakshmanan and S.B. Rao [1] also deliberated the Gallai graphs and anti-Gallai graphs. Further, A. Somasundram and S. Somasundram [5] have explored the domination in fuzzy graphs. In accumulation, the domination, independent and irredundance numbers were discussed by A. Nagoorgani and P. Vadivel [2]. In [8], in our previous effort we have discussed the concept of Gallai-type theorems in Gallai Fuzzy Graphs on Domination parameters and Also discussed the Dutch windmill $D_3^{(m)}$ of Gallai fuzzy graphs on domination number[9]. The concept on corona product of two fuzzy graphs was discussed by Ozge Colakoglu Havare and Hamza Menken [3]. In this paper, we shall investigate about the anti Gallai fuzzy graphs in corona graphs.

2. Preliminaries

Definition: 2.1[6]

The Gallai fuzzy graph Γ(G) of a fuzzy graph G has the fuzzy edges of G as its fuzzy vertices and two distinct fuzzy edges of G are fuzzy incident in G, but do not span a fuzzy triangle in G. The line fuzzy graph L (G) of a fuzzy graph G has the fuzzy edges of G as its fuzzy vertices and two distinct fuzzy edges of G are adjacent in L (G) if they are fuzzy incident in G.

Definition 2.2[6]

A Gallai set of fuzzy vertices which covers all the fuzzy edges of a Gallai fuzzy graph Γ(G) is called a fuzzy vertex cover of Γ(G).The smallest number of fuzzy vertices in

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any fuzzy vertex cover for Γ(G) is called its fuzzy vertex covering number and is denoted by α_o(Γ(G)).

Definition 2.3[6]

Two fuzzy vertices in Γ (G) are said to be fuzzy independent if they are not fuzzy adjacent. A subset S of V is a fuzzy independent set if no two fuzzy vertices on S are adjacent. The maximum number of fuzzy independent vertices in a Gallai fuzzy graph Γ (G) is called fuzzy independence number of Γ (G) and is denoted by β_o (Γ (G)).

Definition 2.4[6]

Let Γ (G) = (σ, μ) be a Gallai fuzzy graph. A fuzzy dominating set S of Γ (G) is said to be a minimal fuzzy dominating set for γ(Γ (G)), if no proper fuzzy subset of S is a fuzzy dominating set.

Definition 2.5[6]

The fuzzy domination number of a Gallai fuzzy graph Γ(G) denoted by γ(Γ(G)) is defined by γ(Γ(G)) = $\wedge\{|S|$: S is a fuzzy dominating set of Γ(G), that is γ(Γ(G)) the minimum fuzzy cardinality of a set in the set of minimal fuzzy dominating set of Γ(G).

3. Anti - Gallai Fuzzy Graph in Corona Graphs.

In this section, we derive the definition, theorems and examples of anti - Gallai fuzzy graph in corona graphs.

Definition: 3.1

Let G_1 and G_2 be two fuzzy components. The corona graph $G = G_1oG_2$ obtained by taking one copy of G_1, $|V(G1)|$ copies of G_2, andthen each fuzzy vertices of G_1 are adjacent to every fuzzy vertex in the copyof G_2.

Definition: 3.2

A tour of a connected fuzzy graph G is a closed walk that traverses each fuzzy edge of G at least once, and an Euler tour one that traverses each fuzzy edge exactly once. A fuzzy graph is Eulerian if it admits an Euler tour.

Definition: 3.3

The anti - Gallai fuzzy graph Δ (G = G_1 o G_2) of a fuzzy graph G has the fuzzy edges of G as its fuzzy vertices and two distinct fuzzy edges of G are fuzzy adjacent in Δ(G_1 o G_2), if they are incident in G and lie on a triangle in G. This concept of anti - Gallai fuzzy graph will be apply to the corona graphs, G = G_1 o G_2.

Example: 3.4

Let us consider P_2 o P_2, n >1 be a corona graphs. Also, the domination number we have the following

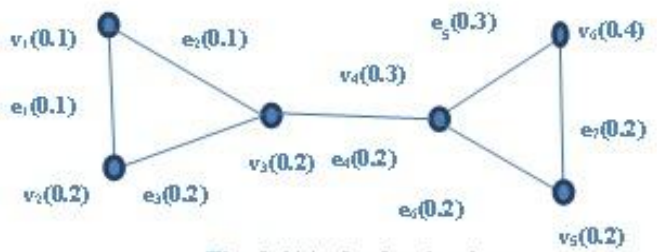

Fig: 3.4(a), the domination number $\gamma(P_2 \text{ o } P_n, n=2) = 0.3$

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By using the definition of anti - Gallai corona fuzzy graphs, $\Delta[P_2 \text{ o } P_n]$, n = 2 and the domination number, we have the following.

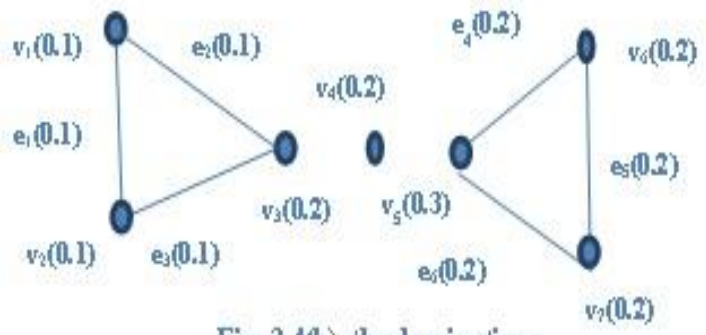

Fig: 3.4(b), the domination number $\gamma(\Delta(P_2 \text{ o } P_n, n=2)) = 0.5$

Theorem: 3.5

Suppose P_2 o P_n , n > 1, is a corona fuzzy graph of G. Then $\Delta(P_2 \text{ o } P_n) = G_1[C_1]$ o $G_2[C_2]$ o $G_3[P_1]$ is disconnected fuzzy graphs with exactly three components.

Proof:

Let P_2 and P_n , n > 1, be two distinct fuzzy paths.
Claim $\Delta(P_2 \text{ o } P_n) = G_1[C_1] \cup G_2[C_2] \cup G_3[P_1]$ is disconnected fuzzy graphs with exactly three components. Clearly $|E(P_2 \text{o } P_n)| = |V(\Delta(P_2 \text{o } P_n))|$ and if any fuzzy edge form a triangle in P_2 o P_n then take that triangle to anti - Gallai fuzzy graph,$\Delta(P_2 \text{ o } P_n)$. There is e+7 vertices, where e = 0,4,8…. and e+6 edges, where e = 0,6,12… in this fuzzy graphs. If P_2 o P_n, n>1 contains at least two fuzzy triangles then $\Delta (P_2 \text{ o } P_n) = G_1[C_1] \cup G_2[C_2] \cup G_3[P_1]$ is disconnected fuzzy graph with exactly three components

Corollary: 3.6

Prove that anti-Gallai fuzzy graph,$\Delta(P_2 \text{ o } P_n) = G_1[C_1] \cup G_2[C_2] \cup G_3[P_1]$ is an even degree.

Proof:

Let $\Delta(P_2 \text{ o } P_n)$ be disconnected fuzzy graphs of anti-Gallai fuzzy graph.
Claim $\Delta (P_2 \text{ o } P_n)$ is an even degree. If $G_1[C_1] \cup G_2[C_2] \cup G_3[P_1]$is disconnected fuzzy graphs. Then these graph exactly two components are Eulerian fuzzy graphs and one component is isolated fuzzy graph. Hence $\Delta(P_2 \text{ o } P_n)$ is an even degree.

Example : 3.7

Let us consider C_3 o P_n,n=2 be a corona graphs.

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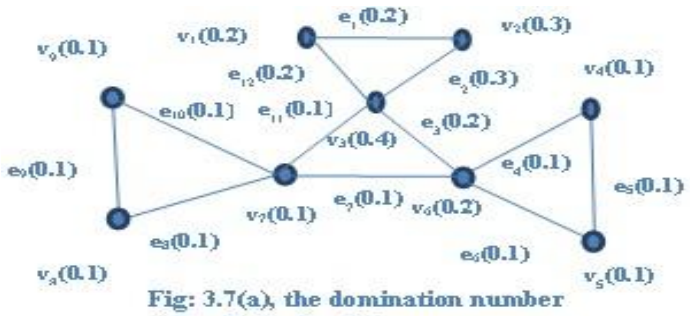

Fig: 3.7(a), the domination number
$\gamma(C_3 \text{ o } P_n, n=2) = 0.4$

By using the definition of anti-Gallai corona fuzzy graph, $\Delta[P_2 \text{ o } P_n]$, n =2 and the domination number, we have the following.

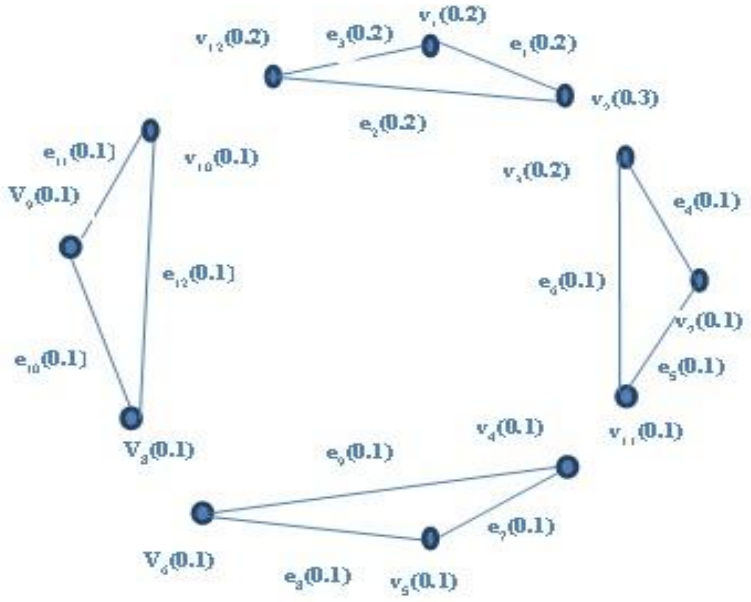

Fig: 3.7(6), the domination number
$\gamma(\Delta(C_3 \text{ o } P_n, n=2)) = 0.5$

Theorem: 3.6

Let C_3 o P_n, n > 1 be a corona fuzzy graph G. Then $\Delta(C_3 \text{ o } P_n) = G_1[C_1] \cup G_2[C_2] \cup G_3[C_3] \cup G_4[C_4]$ is disconnected fuzzy graphs with exactly four components.

Proof:

Let C_3 o P_n, n > 1 be a corona fuzzy graph G. Claim $\Delta(C_3 \text{ o } P_n) = G_1[C_1] \cup G_2[C_2] \cup G_3[C_3] \cup G_4[C_4]$ is disconnected fuzzy graphs with exactly four components.
By the definition of anti-Gallai fuzzy, $\Delta(C_3 \text{ o } P_n)$ $|E(C_3 \text{ o } P_n)| = |V[\Delta(C_3 \text{ o } P_n)]|$ and if any fuzzy edge form a triangle in P_2 o P_n then take that triangle to anti - Gallai fuzzy graph, $\Delta(C_3 \text{ o } P_n)$.There is e+12 vertices, where e = 0,6,12,… and e+12 edges, where e = 0,9,18,…. in this fuzzy graphs. Hence $\Delta(C_3 \text{ o } P_n) = G_1[C_1] \cup G_2[C_2] \cup G_3[C_3] \cup G_4[C_4]$ is disconnected fuzzy graphs with exactly four components.

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Structural, morphological and electrochemical studies of nanostructured $BiVO_4$ for supercapacitor application

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ABSTRACT

A facile and cost-effective stratagem has been applied in the synthesizing of multifunctional Bismuth Vanadate ($BiVO_4$) nanoparticles (NPs) by using one-step hydrothermal method. The synthesized $BiVO_4$ NPs under three different temperatures such as 140, 160 and 180 °C led different morphologies. Further they have been subjected to various characterizations such as XRD, FTIR, SEM, RAMAN, UV and electrochemical analyses. XRD pattern showed the combined monoclinic and tetragonal crystal structures which may be due to various temperatures (140, 160 and 180 °C). FTIR is proved with metal oxide (V–O) vibrations and also substantiated the purity of the samples. The optical band gap values were studied by UV spectral results for different reaction temperatures. The electrochemical evaluation and supercapattery studies were carried out for all the samples. The maximum specific capacitance (C_{sp}) of 1451 F g^{-1} at a current density of 1 A g^{-1} was observed to $BiVO_4$ NPs synthesized at 180 °C.

1. Introduction

In the modern era, energy requirement is considered as a major issue across the world. As a developing energy storage device, supercapacitors (SCs) have received huge attention because of their peculiar ability to shorten the space between the batteries and conventional capacitors [1,2]. Interestingly, SCs play an promising role in electrical system which produces high power energy in a short period. Furthermore, there is the availability of two varieties of SCs such as an electric double-layer capacitor (EDLC) and pseudocapacitor (PCs) [3,4]. Generally, EDLC devices take place the ion transfer mechanism involves at the interface between the electrolyte and electrode surface which results in providing low specific capacitance. For the PCs system, a faradaic redox reaction has happened during the electrochemical process and poses a high specific capacitance [4,5]. In PCs, nanostructured transition metal oxides (TMOs) such as RuO_2, MnO_2, MoO_3, $Ni(OH)_2$, etc., have been widely utilized as an electrode material owing to their peculiar properties of inexpensive cost, earth abundant, high surface area and excellent redox properties [6–8]. However, TMOs greatly suffers from their practical utilization because of their poor cyclic stability and rate capability. To ensure the aforementioned problem, the exploitation of superior and highly stable electrode material is a crucial concern in research analysis [9–11].

Continuing, binary semiconducting transition metal oxides (BSTMOs) have been gained with immense attention as an electrode material due to their superior electrical conductivity, thermal stability and large active surface area when compared to primary TMOs [12–14]. In particular, bismuth and vanadium combined oxides ($BiVO_4$) have a wide variety of applications including photocatalysis, photo-electrocatalysis, energy storage devices and photo-electrochemical water splitting owing to their narrow band gap, low onset-potential, good redox capacity and prolonged stability [5,15–18]. Vanadium (IV) oxide based symmetric supercapacitors showed better electrochemical performance because of its properties such as variable oxidation state, lower cost, high level capacitance, good rate capability and improved storage ability [19]. $BiVO_4$ is considered as one of the acted as best n-type semiconducting materials because of forming the bandgap energy (2.4–2.5eV), high reduction potential, leading absorption ability, great capacitive ability, etc [20]. However, $BiVO_4$ NPs has been varied into three types such as zircon structured tetragonal, Scheelite structured tetragonal and Scheelite structured monoclinic phase depending upon their lattice crystal facets [21]. Among them, monoclinic phase $BiVO_4$ demonstrates superior photocatalytic and electrocatalytic properties because of their strong distortions between

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the BiO_6 octahedron and VO_4 tetrahedron [22]. For these reasons, the synthesis of the pure m-$BiVO_4$ phase through a simple technique is still challenging task for the researchers. Up to now, plenty of techniques have been available for the preparation of m-$BiVO_4$ which includes including microwave-assisted route, flame spray pyrolysis, solid-state reaction, co-precipitation, hydrothermal, solvothermal and solution combustion method [23–29]. Among them, hydrothermal route could offers environment-friendliness and uncomplicated experimental condition, and the obtained products were uniform in size, well-defined self-assembled structure, peculiar morphology and high phase purity [30]. Moreover, It has extraordinary electrochemical behaviour owing to their high surface to volume ratio [31].

Here, we report a facile and simple hydrothermal synthesis with three different elevated temp (140, 160 and 180 °C) nanostructured m-$BiVO_4$ NPs and its characterization in detail. This is the first report of the investigation on the phase structure, morphology and electrochemical properties of $BiVO_4$ NPs at three various temperatures (140, 160 and 180 °C) of the hydrothermal method. Based on our experimental results, the diverse functioning temperatures and morphologies spurred us to evaluate the electrochemical behaviour of the prepared semiconducting $BiVO_4$ materials as an electrode material for supercapacitor applications.

2. Experimental details

2.1. Materials

The synthetic precursors utilized in this preparation were taken as analytical grade chemicals and subsequently utilized without any further purification. The starting precursors are bismuth nitrate pentahydrate (Bi $(NO_3)_3{\cdot}5H_2O$), ammonium metavanadate (NH_4VO_3), sodium hydroxide (NaOH), hexamethylenetetramine (HMT), ethanol (Merck) and double distilled (DD) water. The entire reaction was performed in an aqueous medium, in a hydrothermal reaction using stainless steel (SS) Teflon-lined autoclave. Nickel foam (1.5 mm thickness-pore size 0.5 mm) was purchased in MTI Corporation for electrochemical studies.

2.2. Synthesis of $BiVO_4$ NPs

In this typical recipe, 2 mM Bi $(NO_3)_3.5H_2O$ and 2 mM NH_4VO_3 were separately dissolved in 50 ml DD water, respectively (named as solution-A and solution-B). Additionally, 2 ml $HNO_3$was added to solution-A. The ammonium metavanadate (NH_4VO_3) was dissolved in H_2O at 50 °C. After that, the solution A and solution B were mixed dropwise under constant stirring. Consequently, a certain amount of 0.01 M of NaOH in 10 ml and 100 mg of hexamethylenetetramine (HMT) in 10 ml was added to the above suspension and the obtained yellow-colored suspensions 120 ml were transferred in to 150 ml volume Teflon-lined autoclave and further allowed to the hydrothermal reaction in different operating temperature at 140, 160 and 180 °C for 12 h. The final precipitates were cooled at room temperature (RT) then washed five times with DD water and followed by ethanol then collected using a centrifuge and dried at 70 °C for 5 h.

2.3. Characterization

The crystalline structure of all the prepared $BiVO_4$ materials was recorded by XRD using Bruker (D8) advance spectrometer with 1D SSD160 detector and Cu-Kα radiation at the wavelength range of 1.541 Å. Functional group analysis has been done by Shimadzu FTIR TENZOR-27 spectrophotometer with the range of 4000–400 cm^{-1}. Diffuse Reflectance Spectra (DRS) of samples and the absorption spectra of $BiVO_4$ NPs were analysed on a JASCOUV–Vis–NIR Spectrophotometer Model V–770PC.ZEISS EVO-18 equipped BRUKER-X Flash-6130 scanning electron microscopy (SEM) was used to observe the surface topology and elemental composition of the prepared NPs. HRTEM measurements were probed using a JEOL JEM-2100 instrument. Raman spectrum was carried out on a BRUKER RFS-27 FT-Raman Spectrometer with a range of 50–1000 cm^{-1}. Electrochemical measurements of modified electrodes were performed using an electrochemical workstation (CHI6008E, USA) with a three electrode cell setup at room temperature.

2.4. Electrode fabrication and electrochemical analysis

The $BiVO_4$ NPs modified working electrodes were made as follows, the combination of active material (sample), conducting material (activated carbon) and binder (PVDF) with the ratio of 80:15:5, respectively, were grounded using an agate mortar and *N*-methyl pyrrolidone was added drop wise until a homogeneous slurry was obtained. The resulting mucilage was coated on 1 × 1 cm^2 Ni-foam and dried in a hot air oven at 80 °C for 12 h. Eventually, the dried electrodes were pressed at 10 MPa employing pelletizer later were used as working electrodes. Herein, saturated Ag/AgCl and Pt wire were used as the reference and counter (auxiliary) electrode, respectively. The 2 M KOH solution was used as the electrolyte for all three samples. The compatible specific capacitance values were calculated from the following equations [32,33]:

$$C = \frac{i * \Delta t}{\Delta V * m} \quad (1)$$

$$C = \frac{\int i dV}{s * m * \Delta V} \quad (2)$$

where, C-specific capacitance in (F g^{-1}), i-discharge current in ampere, Δt-discharge time in a sec, ΔV (V)-potential window, s-scan rate (V/s) and m-a mass of the active material (g).

3. Results and discussion

3.1. Structural, morphological and optical characterization of samples

Fig. 1 shows X-ray diffraction (XRD) pattern of the prepared $BiVO_4$ NPs at three different hydrothermal temperatures. For the samples $BiVO_4$-140 °C and $BiVO_4$-160 °C, diffraction pattern are in perfect consistency with their standard JCPDS data which exhibits heterophase structure including tetragonal type zircon phase (JCPDS No.14–0133) and monoclinic scheelite phase (JCPDS No.14–0688) structure of $BiVO_4$. The operating temperature at 180 °C, the diffraction peaks of $BiVO_4$-180 °C is obtained in complete agreement with that of pure monoclinic

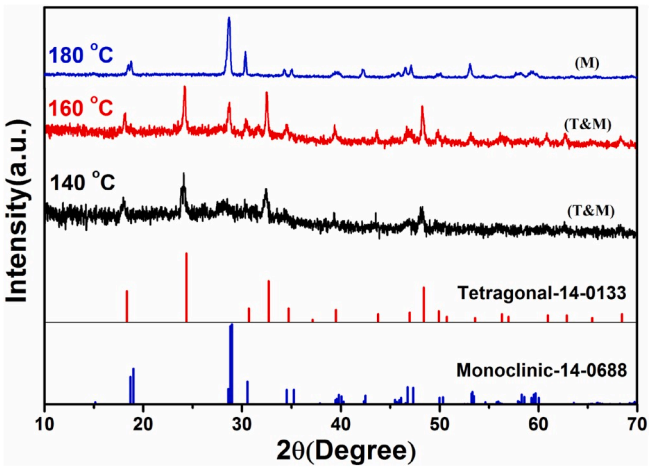

Fig. 1. XRD pattern of $BiVO_4$ at different hydrothermal temperatures such as 140 °C, 160 °C and 180 °C.

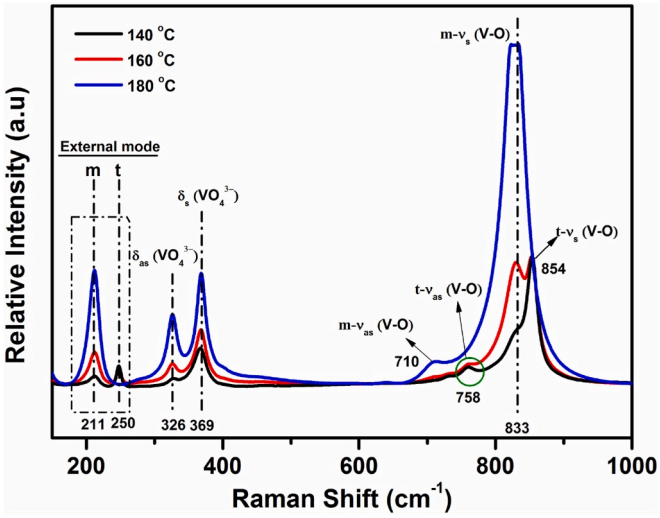

Fig. 2. Raman spectra of $BiVO_4$ samples with different hydrothermal temperatures such as 140 °C, 160 °C and 180 °C.

scheelite-crystal structure $BiVO_4$with space group (15) I2/a (JCPDS No.14–0688). Because, from literature, the minimum hydrothermal temperature required to form pure monoclinic phase $BiVO_4$ is 180 °C [34].

Furthermore, Raman studies were performed in order to gain the detailed structural insights of as-prepared materials and presented in Fig. 2. It can be seen that the intensity of peaks increases gradually while increasing the temperature. All the three (140, 160 and 180 °C) $BiVO_4$ materials showed the peaks at the lower regions such as 369 and 326 cm^{-1} were ascribed to the symmetric and asymmetric bending vibrations modes of VO_4^{3-}, respectively. The external twisting modes in $BiVO_4$ were observed at the 211 and 250 cm^{-1} which can be corresponding to the formation of monoclinic and tetragonal phase, respectively (Fig. 2) [35,36]. However, $BiVO_4$ materials exhibit the Raman bands at 833 and 854 cm^{-1} corresponding to the symmetric stretching vibrations of monoclinic and tetragonal V–O bonds, respectively. Besides, asymmetric stretching vibration of tetragonal V–O bonds was observed at 758 cm^{-1} for operating temperatures of 140 °C and 160 °C whereas the asymmetric stretching vibration of the V–O bond at monoclinic was appeared at 710 cm^{-1} for 180 °C of $BiVO_4$. Hence, at 180 °C $BiVO_4$, it was clearly portrayed the monoclinic phase due to predominant high intense peaks

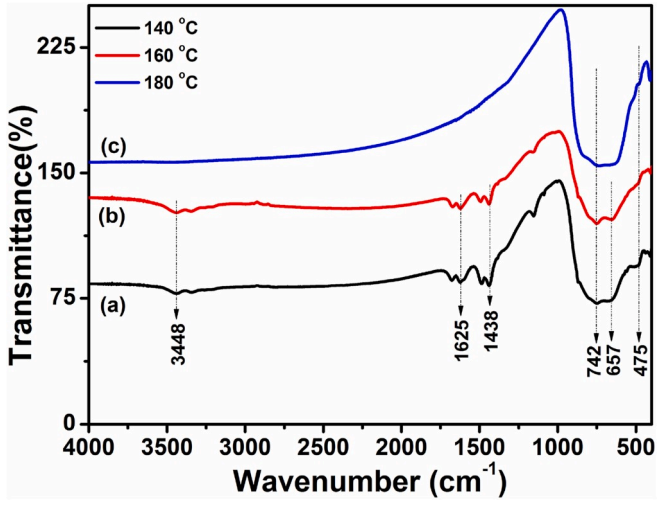

Fig. 3. FTIR pattern of $BiVO_4$ at different hydrothermal temperatures such as **(a)**140 °C, **(b)** 160 °C and **(c)**180 °C.

at 833 cm^{-1} and 710 cm^{-1} [37]. Moreover, no other redundant peaks at tetragonal $BiVO_4$ are implied that the confirmation of pure monoclinic $BiVO_4$ at 180 °C which is well coincide with the XRD results.

The contribution of functional groups for the synthesis of $BiVO_4$ NPs was studied by FTIR studies. FTIR spectrum in Fig. 3 (curve a & b) illustrate the peaks at 3448 and 1625 cm^{-1} were ascribed to the symmetric stretching and bending vibrations of H–O–H molecules adsorbed on the material surface atmosphere [38]. The band at 1438 cm^{-1} was attributed to the $–CH_3$ bending vibrations of HMT in $BiVO_4$ [39]. The band at 742 cm^{-1} was correspond to the asymmetric stretching vibration of metal – oxide due to V–O group and its shoulder peak at 657 cm^{-1} was respect to the shorter V–O bond which might be due to the presence of both tetragonal and monoclinic phase structures in (140 °C and 160 °C) $BiVO_4$ materials. At higher temperatures (180 °C), the reduction of the distinctive peak at 742 cm^{-1}clearly suggested that the presence of longer asymmetric V–O bond of pure monoclinic phase and these results are well consistent with the raman report [40].

Optical studies are one of the important tools for identifying the semiconducting based NPs. Fig. 4(a) shows the optical UV-diffuse reflectance (DRS) spectra of as-prepared semiconducting $BiVO_4$ NPs at three different temperatures in the hydrothermal process. It can be seen that m-$BiVO_4$ (180 °C) illustrates the red-shift in the visible light region when compared to the other two tetragonal mingled monoclinic phases. By using Tauc's equation, the determined energy gap (E_g) values are 2.43, 2.42 and 2.40eV for 140, 160 and 180 °C, respectively. The results revealed that the pure m-$BiVO_4$ phase portrayed a lower energy gap (2.40 eV) which could be more beneficial to catalytic activity and solar light absorption performances [41].

As synthesized $BiVO_4$ NPs at different hydrothermal conditions have been subjected to examine the morphology and purity by SEM equipped with EDX and HRTEM techniques as shown in (Fig. 5 & Fig. 6). The images have demonstrated the impact of temperature on morphology. Fig. 5 [a1, b1] show the sphere which is tightly composed with tiny cubes. Increase of temperature from 140 °C to 160 °C causes the growth of tiny cubes thereby the tightly bound tiny cubes tends to become free as shown in Fig. 5 (a2&b2). This may be supposed due to the increase of kinetic energy led by temperature. The further increase of temperature to 180 °C surges the kinetic growth of rod-like structure and it is noticed in Fig. 5 (a3, b3). In results of the XRD pattern of $BiVO_4$ at 180 °C, a pure monoclinic crystal structure is observed. Altogether, a complete formation of rod like structure is also observed at the same operating temperature. EDX spectra are showed the existence of Bi, V and O elements only which implies the purity of the samples as shown in Fig. 5 (c1, c2 and c3).

HRTEM images (Fig. 6) strongly demonstrated the cubic shape and nanosized spherical particles for the $BiVO_4$ sample synthesized at 140 & 160 °C. The rod-like shape (180 °C) of $BiVO_4$ NPs is appeared as shown in Fig. 6 (180-a&b). Selected area electron diffraction (SAED) patterns of 180 °C have displayed circles that indicate the nanocrystalline nature of the sample. For the illustrated circles of SAED pattern of the 180 °C sample, the d-spacing values can be estimated using the following equation [42].

$$L\lambda = dR \tag{3}$$

where, L-length of the instrument camera (120 mm), λ – (0.04965 Å) is the wavelength of the high-resolution electron beam, R-radius measured from diffracted circles. The estimated d-spacing values good agreement concerning with respect to hkl planes of (110), (011), (−121), (040), (200) and (002), which they were well accordance with XRD results.

XPS spectroscopy is a significant tool to analyze the chemical state of the as-prepared nanomaterials and identified the presenting elements of the prepared elements. Fig. 7(a) shows the survey spectrum of the as synthesized m-$BiVO_4$ sample (@180 °C) and which perspicuously shows the characteristics peaks of Bi 5 d, 4f, 4 d and 4p, O 1s and V 2p states. The peak presented at 285 eV denoted C 1s as reference for calibration.

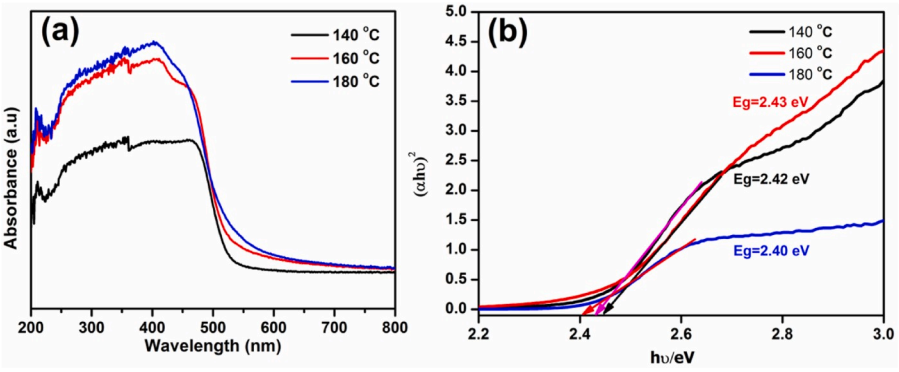

Fig. 4. **(a)** UV plot and **(b)** Tauc plot of as-prepared $BiVO_4$ at different hydrothermal temperatures such as 140 °C, 160 °C and 180 °C.

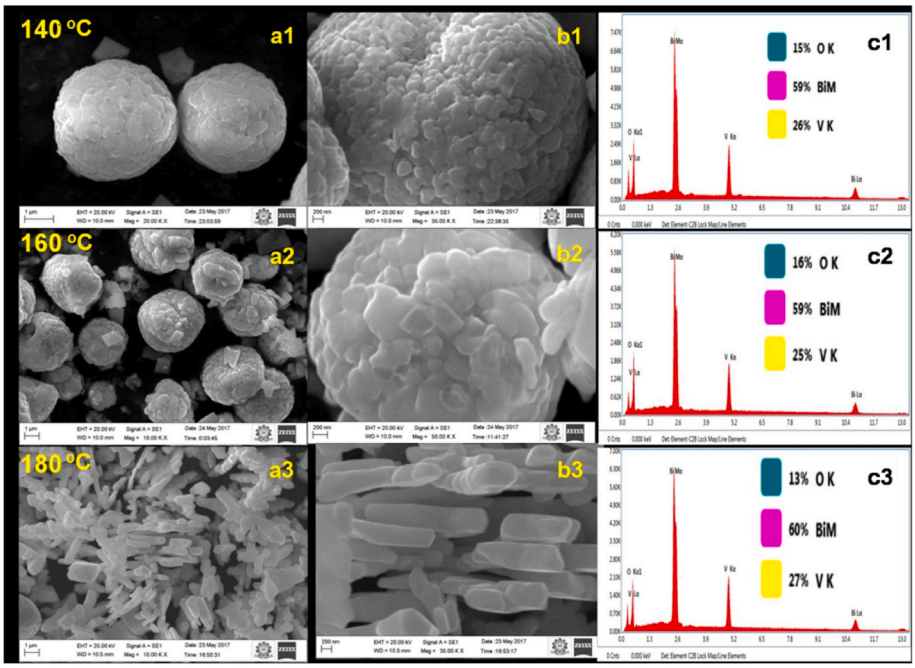

Fig. 5. The SEM and EDX images of $BiVO_4$ NPs prepared at different hydrothermal temperatures such as 140 °C (a1, b1, c1), 160 °C (a2, b2, c2) and 180 °C (a3, b3, c3).

Fig. 7(b–d) shows the high resolution element spectra of Bi, V and O of m-$BiVO_4$. Fig. 7 (b) depicts the binding energy peak at 159 eV and 164 eV which was ascribed to the Bi $4f_{7/2}$ and $4f_{5/2}$ core spin level. The binding energy peaks located at 517 and 524.8 eV can be attributed to V $2P_{3/2}$ and V $2P_{1/2}$ states as shown in Fig. 7 (c) [43]. The curve fitting in the O1s region was contributed by two peaks having peak position O═C–OH, C═O at 529.7 eV (lattice oxygen in $BiVO_4$), and 531.2 eV (attributed to H_2O or chemisorbed oxygen ions) respectively. The observed XPS binding energy values clearly enunciated that the chemical elemental composition and oxidation states of prepared m-$BiVO_4$ materials [44,45].

3.2. *Electrochemical properties of $BiVO_4$ electrodes*

The electrochemical properties of all the prepared samples are studied by cyclic voltammetry (CV) technique in 2 M KOH in aqueous medium using three electrode cell setup between the potential range of 0.1–0.5 V with various scan rates such as 5, 10, 25, 50, 75 and 100 mV/s. Fig. 8(a–c) represented the CVs of as-prepared samples ($BiVO_4$- 140, 160 and 180 °C). It clearly observed that the distinctive strong anodic and cathodic peaks confirmed electrochemically active and is governed by Faradaic redox reaction. The observation from Fig.(8a-8c) indicated that the direct proportionality of scan rates and peak currents attributing the direct diffusion of hydroxyl radical anions (OH^-) at the reactive sites of the electrode surface [46]. Moreover, a linear plot was visualized between the square root of scan rate versus peak current with the correlation co-efficient (R^2) of 0.9995 for cathodic and 0.9994 for anodic peak, implied that the whole electrode reaction occupies the diffusion-controlled process (Fig. 9) [47]. Fig. 7(d) displays the comparison of CV curves of all the three samples at 50 mV/s. The maximum current density observed for $BiVO_4$ (180 °C) NPs is due to the small particle size compared to other samples. The maximum current density observed for $BiVO_4$ (@180 °C) NPs when compared than other others. As we known that, the electrochemical capacitance behaviour mainly related to the intercalation as well as de-intercalation of cations of electrode material, however, only the crystalline phase structures might provide the surplus amount of ion transfer at the electrode surface which are more beneficial for the capacitance performances. Therefore, the pure monoclinic phase of the $BiVO_4$ demonstrated superior capacitance performances in this present study [48].

Based on the electrochemical reaction, the intercalation of electrolyte ions onto the electrode surface is considerably high since the rod-

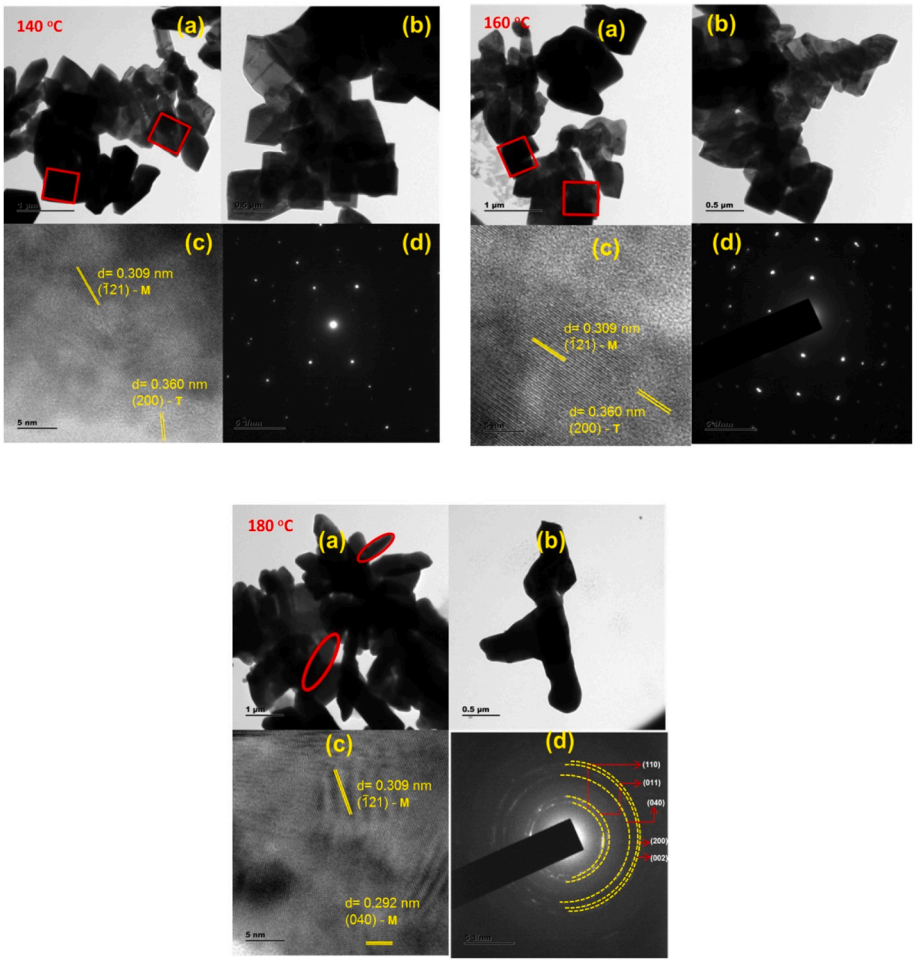

Fig. 6. HRTEM images (a–c) of the $BiVO_4$ NPs with SAED pattern (d) prepared at different hydrothermal temperatures - 140 °C, 160 °C and 180 °C with different magnifications.

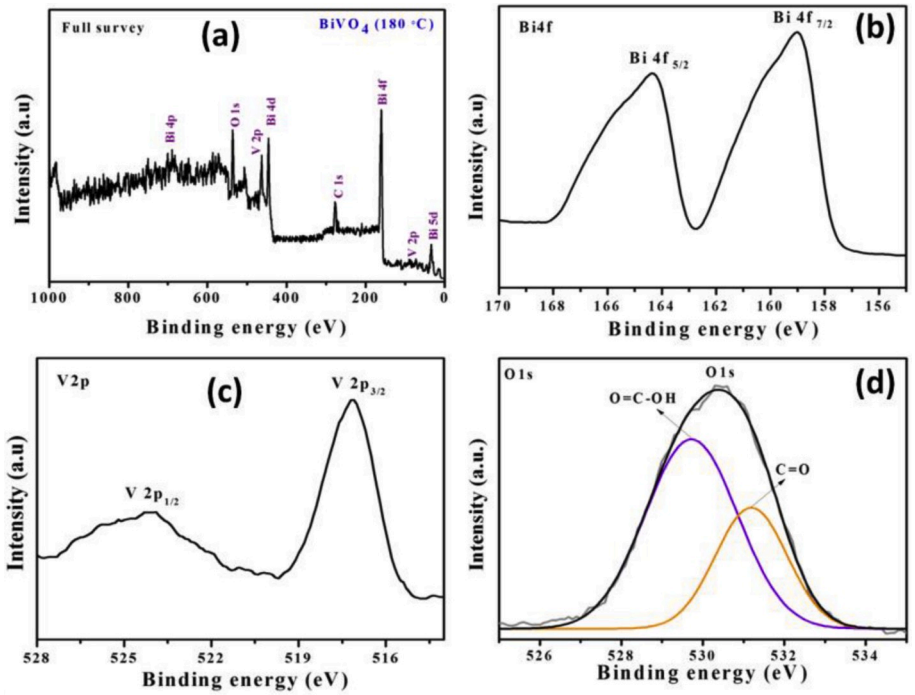

Fig. 7. XPS spectra of prepared at 180 °C-$BiVO_4$ sample.

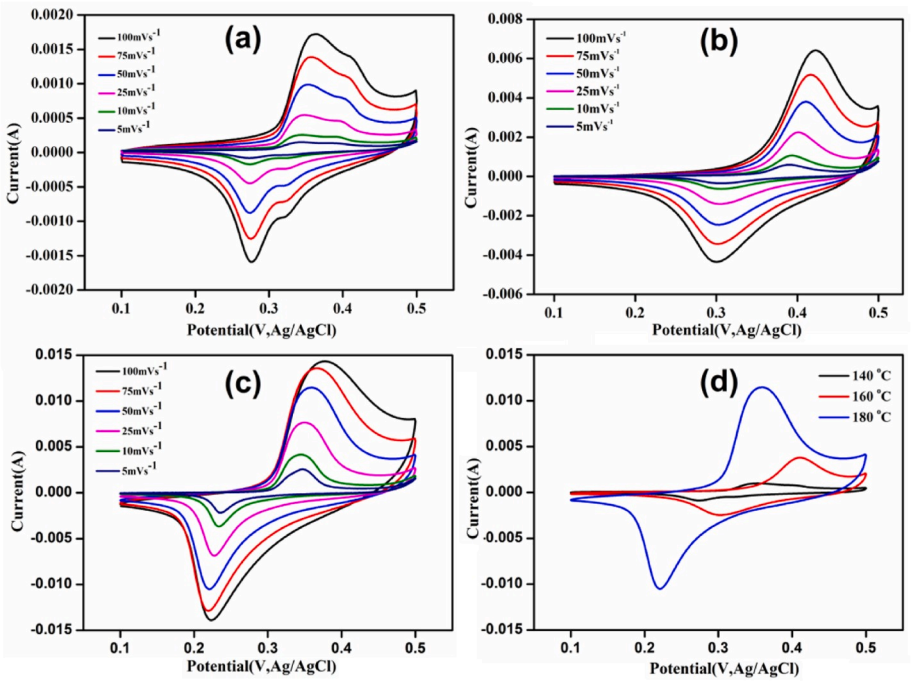

Fig. 8. (a, b, c) Cyclic Voltammograms curves of $BiVO_4$ -140 °C, 160 °C and 180 °C electrodes at different scan rates from 5 to 100 mVs^{-1} (d) comparison CV curves of different hydrothermal temperatures 140 °C, 160 °C and 180 °C at 50 mVs^{-1}..

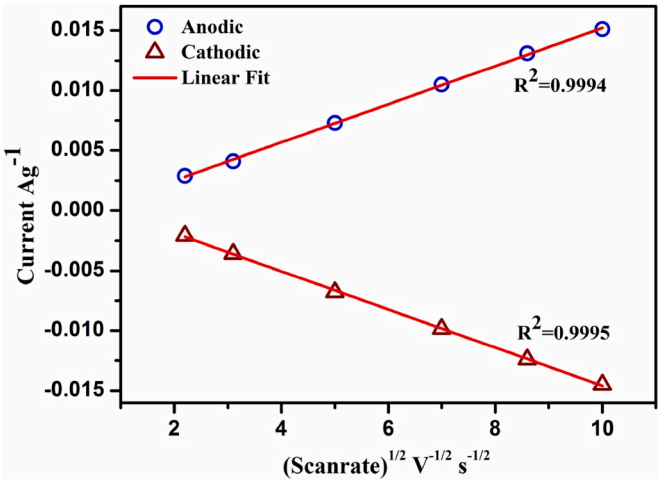

Fig. 9. Peak current versus square root of scan rate plots of $BiVO_4$ -180 °Csample

Table 1
Specific capacitance data of $BiVO_4$-180 °C sample obtained from CV and GCD curves.

Specific capacitance of $BiVO_4$-180 °C sample			
Specific capacitance from CV graph		Specific capacitance from GCD graph	
Scan rate m Vs^{-1}	Specific capacitance (F g^{-1})	Current density A g^{-1}	Specific capacitance (F g^{-1})
5	317	1	1451
10	302	2	1386
25	273	3	1066
50	195	4	311
75	102		
100	88		

like $BiVO_4$ NPs shaped nanoparticles have a large surface area. All the $BiVO_4$ NPs have a similar tendency of the occurrence faradaic redox reaction mechanism [49].

The specific capacitance (C_{sp}) values for all the samples were calculated using equation (1) and tabulated in Table 1.

$$BiVO_4 + xK^+ + Xe^- \leftrightarrow K_xBiVO_4 \quad (4)$$

GCD curve of the prepared samples ($BiVO_4$-140, 160 and 180 °C) in a potential window range between 0 to 0.45 V against the Ag/AgCl electrode has been studied at various current densities ranging from 1 to 4 A g^{-1} and presented in Fig. 10(a–c) respectively. All the GCD curves depict the pseudocapacitance nature of the materials. All the curves are not linear and have two curves. Further it is noticed that there is a small potential drop at a low current density and a huge potential drop at a high current density which is about pertaining to fast redox mechanism at higher current density. Fig. 10 (d) depicts the displays the comparison of GCD curves of all the three samples at 1 mA/cm^2. Interestingly, the $BiVO_4$ (180 °C) sample demonstrated the maximum specific capacitance (Csp) value and the Csp of all the three ($BiVO_4$-140, 160 and 180 °C) electrode materials were tabulated in Table 1.

Cyclic stability, repeatability and reproducibility are the vital parameter of the electrode for its real time utilizations. Therefore, $BiVO_4$ electrode probed cyclic stability for 5000 continuous charge discharge cycles in 2 M KOH solution at the current density 4 A g^{-1} and the results are illustrated in Fig. 11. As can be seen, the proposed electrode material portrayed 98.97% and 97.4% retention efficiency even up to 3000 and 5000 repeated cycles, respectively. The observed results unambiguously revealed that the developed electrode material is more capable of real-time practical applications.

The impedance plots of the prepared $BiVO_4$-140, 160 and 180 °C based electrodes in the frequency range from 0.01 to 100 kHz are shown in Fig. 12. The inset in Fig. 12 shows the equivalent fitting circuit (Z-view) of $BiVO_4$-(140, 160 and 180 °C) electrodes. R_s represent the solution resistance and R_{ct} denotes the charge transfer resistance. The semicircle part implies the charge transfer resistance which is 0.09, 0.09 and 0.08 Ω for the $BiVO_4$-140,160 °C and 180 °C electrodes, respectively. The low R_{ct} value of the $BiVO_4$ electrode possesses an enhanced

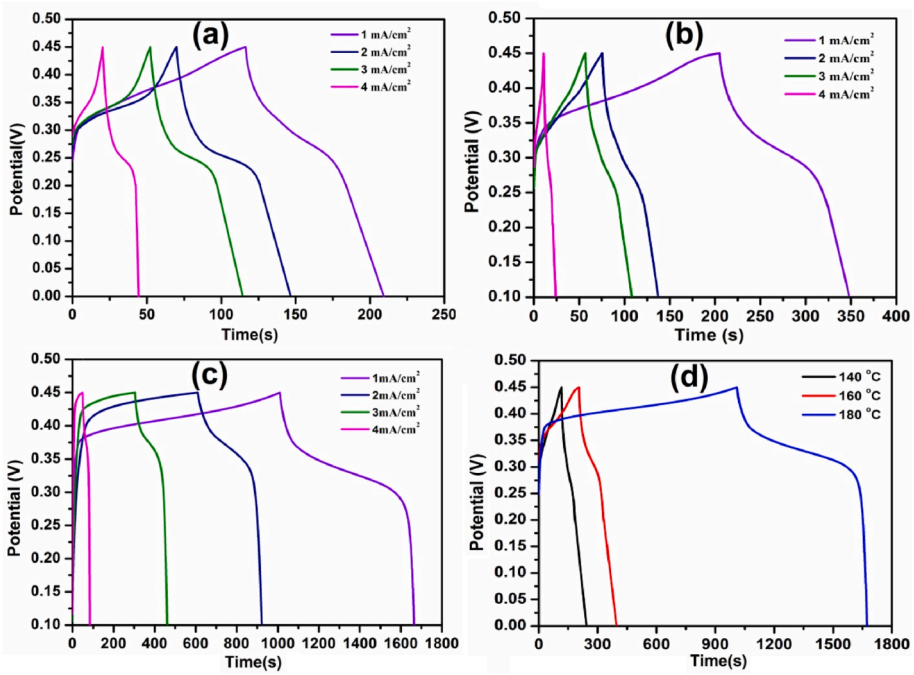

Fig. 10. (a, b, c) Charge discharge curves of $BiVO_4$ -140 °C, 160 °C and 180 °C electrodes at different current densities from 1 to 4 mA/cm^2 (d) comparison GCD curves of different hydrothermal temperatures such as 140 °C, 160 °C and 180 °C at 1 mA/cm^2.

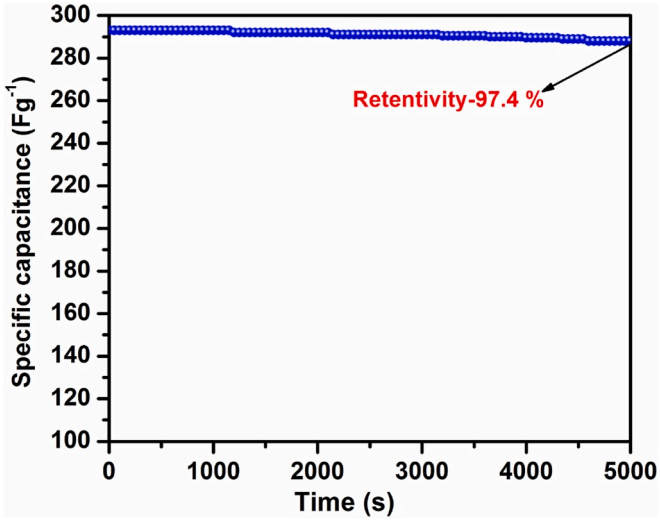

Fig. 11. Cyclic stability graph of $BiVO_4$ - 180 °C sample.

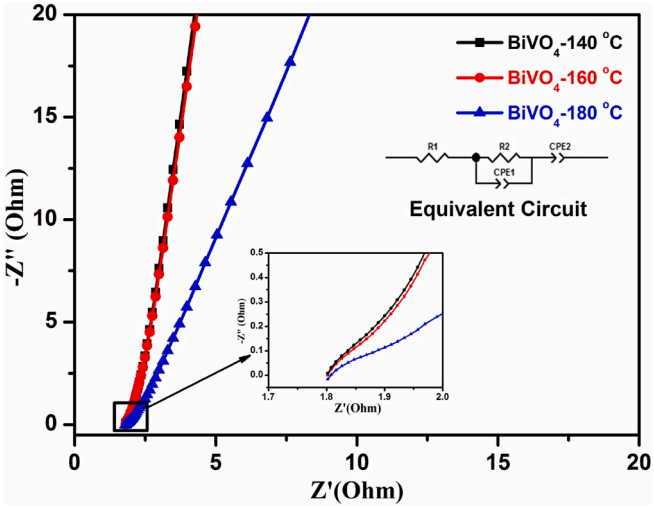

Fig. 12. Electrochemical impedance spectra of $BiVO_4$ NPs at different hydrothermal temperatures such as (140 °C, 160 °C and 180 °C).

diffusivity of OH^- ions [50]. The calculated internal resistance values of the electrodes are 1.79, 1.79 and 1.8 Ω for the $BiVO_4$-140, 160 and 180 °C electrodes, respectively. The low internal resistance values are indicating the good ionic conductivity of the samples [51].

4. Conclusion

To sum up, spheres and rod-like $BiVO_4$ NPs were developed through the three different elevated temperatures (140, 160 and 180 °C) assisted by the hydrothermal process. The structural studies of XRD results confirmed the heterophase of tetragonal-zircon and monoclinic-scheelite phase at 140 °C & 160 °C and pure monoclinic scheelite phase at 180 °C. Moreover, the morphological features were illustrated spherical and nanorods shaped morphology due to its identical operating temperatures. The enhanced electrochemical performances could be ascribed to the well-defined one-dimensional rod-like structure with pure monoclinic phase of $BiVO_4$. Interestingly, $BiVO_4$ NPs synthesized at 180 °C temperature demonstrated superior electrochemical behaviour with a maximum specific capacitance value of 1451 Fg^{-1} at a current density of 1 Ag^{-1}. This results leads to open up for enhanced energy storage devices by simple binary metal oxides with cost-effective.

Declaration of competing interest

We all author declare that, there is no conflict of interest for publishing research article entitled "**Structural, Morphological and Electrochemical Studies of Nanostructured $BiVO_4$ for Supercapacitor Application**" for the publication in Materials Science in Semiconductor Processing.

CRediT authorship contribution statement

R. Packiaraj: Conceptualization. **K.S. Venkatesh:** Formal analysis. **P. Devendran:** Data curation. **S. Asath Bahadur:** Validation. **N. Nallamuthu:** Supervision.

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