



## Synthesis and Identification of Some Schiff Base and Chalcone Compounds and Studying the Effectiveness as Flame Retardant of Some Thermosetting Polymers

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### Abstract

In this work, seven additives were used, namely: (5-(6-methylbenzothiazol-2-yl)diazenyl)-2-hydroxybenzaldehyde(Additive I), (5-(6-methylbenzothiazol-2-yl)diazenyl)-2-hydroxyphenyl)-1-(4-hydroxy-3-methoxyphenyl)prop-2-en-1-one (Additive II), 5,6-(methylbenzothiazol-2-yl)diazenyl)-2-hydroxyphenyl)-1-(4-hydroxyphenyl) prop-2-en-1-one (Additive III), 5-(6-methylbenzothiazol-2-yl) diazenyl)-2-hydroxyphenyl)-1-(4-methoxyphenyl)prop-2-en-1-one (Additive IV), 4-(6-methylbenzothiazol-2-yl) diazenyl)-2-(2-hydroxy-5-methylphenyl)imino)methyl)phenol(Additive V), 4-(6-methylbenzothiazol-2-yl) diazenyl)-2-(2-hydroxy-5-ethylphenyl)imino)methyl) phenol (Additive VI) and (1E)-N-(5-((6-methylbenzo [d] thiazol-2-yl)diazenyl)-2-hydroxybenzylidene)-N'-(4-chlorophenyl)formimidamide(Additive VII). The effects of these additives on flammability of some of the thermosetting polymers (unsaturated polyester and epoxy resins), have been studied. Sheets of the specimens (resins with different weight percentages of additives), in dimensions (150X150X3) mm were prepared. Two standard test methods used to measure the flame retardation which are: (ASTM: D-2863) and (ASTM: D-635). Results are obtained from these tests indicated that, additive 6B has high efficiency as flame retardant, self-extinguishing (S.E.) at the percentage (0.1 %) for unsaturated polyester and the epoxy resins. Also self-extinguishing (S.E.) at the percentage (0.1%) for additives 1B, 4B and 5B, in unsaturated polyester resin and non-burning occurs in 0.15% with Additive 6B and 5B in unsaturated polyester resin but it occurs in 0.2% with additives 6B and 5B in epoxy resin. Additive B shows low effect on flammability in both resins.

**Keywords:** Flame retardant, Flammability, Polyester resin, Unsaturated polyester resin, Epoxy resin, Schiff base, Chalcones, Synthesis of Schiff base, Identification of Schiff base, Synthesis of chalcone, Identification of chalcone.

### Introduction

The history of all polymer materials was traced by the success of their applications in replacing traditional materials like wood, leather and metals [1]. The rapid expansion of combustion in courage many researches to use many additives to retard flammability of the polymers [2]. A large number of synthetic polymeric materials were used these days, with various different properties are available for medical applications and engineering matrices.

Most of the common materials have sufficient mechanical stability and elasticity as well as desired stability towards degradation, and are non-toxic. [3, 4]. Flame retardants are used

to reduce flammability of polymeric materials, these chemical compounds are capable of imparting flame resistance to the materials and they can be classified into two general types [5]: those which do not react chemically with the polymer and the other type which are those incorporated chemically into the basic polymer structure. A good flame-retardant additive must meet following requirements [6, 7]: thermally stable up to the processing temperature of the polymer and stable to light, not interact with main chain of the polymer, should not be poisonous and should not inversely affect physical properties of polymer. Many inorganic compounds [8, 9] were used as flame

retardants, such as antimony, phosphorus and halogen compounds, but organic compounds were not used as flame retardants because they needed certain preparation conditions and their high efficacy was discouraged inhibition. In this work, the effectiveness of some organic aromatic Schiff bases and chalcones compounds was studied.

## Experimental part

### Materials

- All chemicals were used in this work analytical grade.
- Preparation of flame-retardant additives

### Preparation Additive B

Preparation compound (6-bromobenzothiazol-2-amine) <sup>[10]</sup>

A mixture (4.3 g, 0.25 mole) of para-bromoaniline and (3.80 g, 0.5 mole) of potassium thiocyanate in 70 ml of glacial acetic acid and then refrigerate the mixture to (0-5 C°) and on the other hand, a mixture of 1.3 ml of bromine dissolved in 30 ml of glacial acetic acid was added with a continuous stirring until the addition was completed.

Leave the mixture for an hour to settle at (0-5 C°) and then gradually cool the distilled water. A cold, concentrated solution of sodium hydroxide was gradually added to the formed solution and a yellow light-colored trace was observed, filtered and washed with water several times and then recrystallization with ethanol. The reaction process was followed using TLC technology.

### Preparation of Additive (B) <sup>[11]</sup> [5-(6-methylbenzothiazol-2-yl) diazenyl)-2-hydroxybenzaldehyde]

**Solution (I)** Preparation of the azo compound (B) from dissolving the Thiazole derivative which contain methyl compound by taking (1.18 g, 0.004 mole) and dissolving it in 5 ml of concentrated hydrochloric acid and 20 ml of distilled water in a round bottom flask and then return the mixture in a snow bath at (0-5 C°).

**Solution (II)** dissolved (0.6 g) of sodium nitrite in (5 ml) of distilled water and stirred the mixture for 20 min. at (0-5 C°).

**Solution (III)** This solution was prepared by added solution (II) to the solution (I) in form of droplets gradually added to obtain the diazonium salt at (0-5 C°).

**Solution (IV)** Added 0.6 ml of salicyldehyde to a solution which component (1.0 g of sodium hydroxide dissolved in 50 ml of distilled water and 5 ml of ethanol) and cool the mixture at (0-5 C°). The solution (IV) was then added to the solution (III) and the reaction was left for one hour at pH 6-7 and at (0-5 C°), the product was filtered, the precipitate was collected and dried at 35 C° in the vacuum oven and then recrystallization by using ethanol.

### Preparation of Additive 1B <sup>[12, 13]</sup>

### Methylbenzothiazol-2-yl) diazenyl)-2-hydroxyphenyl)-1-(4-hydroxy-3-methoxyphenyl) prop-2-en-1-one

This derivative was prepared by condensation (1.78 g, 0.006 mole) of azo compound (B) with (0.6 g, 0.004 mole) of (4-hydroxy-3-methoxy acetophenone) in a 100 ml of round bottom flask by dissolved it in 50 ml ethanol and stirred for 20 min.; A 10% (10 ml) of sodium hydroxide solution was gradually added to the droplet for 2-3 min. under continuous stirring.

The mixture was refluxed at 35 C° for 8-16 hr. and pH was equal to 7 by adding 10 ml of concentrated hydrochloric acid (10% M). The product was filtered and washed with distilled water and recrystallization with ethanol. The reaction process was followed using TLC technology with a percentage (1:9) of a mixture of (benzene: methanol).

### Preparation of additive 2B <sup>[12, 13]</sup>

### Methylbenzothiazol-2-yl) diazenyl)-2-hydroxyphenyl)-1-(4-hydroxyphenyl) prop-2-en-1-one

This derivative was prepared by condensation (1.78 g, 0.006 mole) of azo compound (B) with (0.54 g, 0.004 mole) of (4-hydroxy acetophenone) in a 100 ml of round bottom flask by dissolved it in 50 ml ethanol and stirred for 20 min.

A 10% (10 ml) of sodium hydroxide solution was gradually added to the droplet for 2-3 min. under continuous stirring. The mixture was refluxed at 35 C° for 8-16 hr. and pH was equal to 7 by adding 10 ml of concentrated hydrochloric acid (10% M). The product was filtered and washed with distilled water and recrystallization with ethanol. The reaction process was followed using TLC technology with a percentage (1:9) of a mixture of (benzene: methanol).

### Preparation of Additive 3B [12, 13]

#### Methylbenzothiazol-2-yl) diazenyl)-2-hydroxyphenyl)-1-(4-methoxyphenyl) prop-2-en-1-one

This derivative was prepared by condensation (1.78 g, 0.006 mole) of azo compound (B) with (0.6 g, 0.004 mole) of (4-methoxyacetophenone) in a 100 ml of round bottom flask by dissolved it in 50 ml ethanol and stirred for 20 min.; (10 ml) of sodium hydroxide solution 10% was gradually added to the droplet for 2-3 min. under continuous stirring. The mixture was refluxed at 35 °C for 8-16 hr. and pH was equal to 7 by adding 10 ml of concentrated hydrochloric acid (10% M). The product was filtered and washed with distilled water and recrystallization with ethanol. The reaction process was followed using TLC technology with a percentage (1:9) of a mixture of (benzene: methanol).

### Preparation of additive 4B [14, 15]

#### methylbenzothiazol-2-yl) diazenyl)-2-2-hydroxy-5-methylphenyl) imino) methyl phenol

The derivative (4 B) prepared by condensation of azo compound (B) by dissolving (2.67 g, 0.009 mole) from (B) in 20 ml of absolute ethanol in a round flask (100 ml) and stirred for 10 min. A solution of (0.004 mole : 0.69 g) from (2-amino-6-methyl pyridine) which dissolved in 15 ml of ethanol was added and mixed until the mixture is homogenized, then added 3 drops from glacial acetic acid to solution that formed which be dark color after several minutes from stirring and leaving the reaction to 3 hr. with continues stirring. The reaction process was followed using TLC technology with a percentage (1:9) of a mixture of (benzene: methanol) and kept the product 24 hr. then recrystallize by using ethanol.

### Preparation of Additive 5B [14, 15]

#### Methylbenzothiazol-2-yl) diazenyl)-2-2-hydroxy-5-methylphenyl) imino) methyl phenol

The derivative (5 B) prepared by condensation of azo compound (B) by dissolving (2.67 g, 0.009 mole) from (B) in 20 ml of absolute ethanol in a round flask (100 ml) and stirred for 10 min. A solution of (0.004 mole : 0.69 g) from (2-amino-6-methyl pyridine) which dissolved in 15 ml of ethanol

was added and mixed until the mixture is homogenized, then added 3 drops from glacial acetic acid to solution that formed which be dark color after several minutes from stirring and leaving the reaction to 3 hr. with continues stirring. The reaction process was followed using TLC technology with a percentage (1:9) of a mixture of (benzene: methanol) and kept the product 24 hr. then recrystallize by using ethanol.

### Preparation of additive 6B [14, 15]

#### (1E)-N-(5-((6-methylbenzo[d]thiazol-2-yl) diazenyl)-2-hydroxybenzylidene)-N'-(4-chlorophenyl) formimidamide

The derivative (6 B) prepared by condensation of azo compound (B) by dissolving (2.67 g, 0.009 mole) from (B) in 20 ml of absolute ethanol in a round flask (100 ml) and stirred for 10 min. A solution of (0.004 mole : 0.51 g) from (4-chloroaniline) which dissolved in 15 ml of ethanol was added and mixed until the mixture is homogenized, then added 3 drops from glacial acetic acid to solution that formed which be dark color after several minutes from stirring and leaving the reaction to 3 hr. with continues stirring. The reaction process was followed using TLC technology with a percentage (1:9) of a mixture of (benzene: methanol) and kept the product 24 hr. then recrystallize by using ethanol.

### Polymers

- Unsaturated polyester resin, hardener type (MEKP), imported from United Arab Emirate (U.A.E).
- Epoxy resin, type (CY223), hardener type (HY 956), imported from Ciba-Geigy Co.

### Standard Tests

#### ASTM: D-2863

The measurement of limiting Oxygen Index (LOI), is widely used for measuring flammability of polymers [16].

#### ASTM: D-635

The measurement of rate of burning (R.B), average extent of burning (A.E.B), average time of burning (A.T.B), Self - Extinguishing (S.E) and Non - burning (N.B.) [17].

### Preparation of Polymeric Specimens

The specimens of polymeric material containing additives were prepared in

dimensions (150X150X3) mm; three sheets were prepared from each percentage weight (0.1, 0.15, 0.2, 0.25 & 0.3 %); of flame retardant materials (as additives) and using the hardener for each resin. These sheets were cut as samples according to ASTM standard were used in this work.

## Results and Discussion

### Preparation Additive B

#### First Step

Preparation compound (6-bromobenzothiazol-2-amine), Figure (1), FT-IR spectrum <sup>[18]</sup> showed, the peak appearance at (3271, 448)  $\text{cm}^{-1}$  returns to the  $\text{NH}_2$  group, and the appearance of the peak at the frequencies (3093)  $\text{cm}^{-1}$  to alkyl aromatic C-H stretching, while the peak at (1303)  $\text{cm}^{-1}$  is returned to the aldehyde C-N. The frequency band at (810)  $\text{cm}^{-1}$  indicates the presence of bromine in the Thiazole ring and the appearance of the peak at the frequencies (694)  $\text{cm}^{-1}$  to C-S bond stretching, also the frequency band at (1635)  $\text{cm}^{-1}$  for C=N. We also note that there are two peaks that return to the presence of C=C group at frequencies (1527, 1442)  $\text{cm}^{-1}$ . Equation (1), represent this reaction.

#### Second Step

Preparation of additive(B), Figure (2), The FT-IR spectrum showed<sup>[18]</sup>; The peak appearance at (3124)  $\text{cm}^{-1}$  returns to the OH phenol group, and the appearance of the peak at the frequencies (3093)  $\text{cm}^{-1}$  to alkyl aromatic C-H stretching, while the peak at (1866)  $\text{cm}^{-1}$  is returned to the aldehyde C=O. The frequency band at (1280)  $\text{cm}^{-1}$  indicates the presence of methyl in the Thiazole ring and the frequency band at (1627)  $\text{cm}^{-1}$  for C=N. We also note that there are two peaks that return to the presence of the azo group at frequencies (1458, 1542)  $\text{cm}^{-1}$ .

Figure(3), The spectrum of  $^1\text{H-NMR}$ <sup>[19]</sup>, showed; A single beam at the displacement of (10.7) ppm is attributable to the proton phenolic hydroxyl group associated with the ring, two single beam at (6.8, 6.9) due to CH=CH group; single beam at (3.32) ppm; another triplet beam at (2.28) ppm due to (OCH<sub>3</sub>) group; a double signal beam at displacement of (10.25) ppm is due to the aldehyde group (CHO) and another multiple signal at (7.1-7.4) ppm, return to the proton in the aromatic group (CH). Figure(4), The spectrum of  $^{13}\text{C-NMR}$ <sup>[20]</sup> showed; The signal at (165) ppm returns to C18, the signal at

(150) ppm returns to C4 and the signal at (160) ppm returns to C14, also the signal at displacement (191) ppm returns to C10, Signals at displacement at (112-123) ppm returns to carbon aromatic. Equation (2), represent this reaction. Preparation of additive 1B; Figure(5), The spectrum of the FT-IR<sup>[18]</sup> of the derivative (1B) showed; The peak appearance at (3402)  $\text{cm}^{-1}$  to the stretching (OH), the peak at (3039)  $\text{cm}^{-1}$  returns to the stretching (=CH) aromatic in the ring, the peak at frequency (1797)  $\text{cm}^{-1}$  is due to (C=O) ketone, and the frequency band at (1643)  $\text{cm}^{-1}$  of the (C=N), we also note that there are two peaks due to the presence of the azo group at (1465, 1535)  $\text{cm}^{-1}$ .

Figure(6), The spectrum of  $^1\text{H-NMR}$ <sup>[19]</sup> showed; Single signal at (10.02) ppm is return to the proton of phenolic hydroxyl group of the ring, the other signal at displacement (3.8) ppm return to the proton in the (OCH<sub>3</sub>) group, and two singles at the displacement (6.9, 6.8) ppm of the alkenes group (CH=CH), and another signal at displacement (3.32) ppm return to the proton in the (CH-N) group, and multiple signal at (7.1-7.4) ppm of the protons aromatic group. Figure(7), The spectrum of  $^{13}\text{C-NMR}$ <sup>[20]</sup> was also showed; Signals at (126.2) ppm return to C4, signal at (165) ppm return to C10 and signal at (129.7) ppm return to C14, signal also at displacement (55) ppm return to C32, signal at (150) ppm return to C27, signal at (169) ppm return to C29, signal at (130) ppm return to C18, signals at displacement at (111-123) ppm returns to aromatic carbon.

Equation (3), represent this reaction. Preparation of additive 2B; Figure (8), The spectrum of the FT-IR<sup>[18]</sup> of the derivative (2 B) showed; The peak appearance at (3417)  $\text{cm}^{-1}$  to the stretching (OH), the peak at (3249)  $\text{cm}^{-1}$  returns to the stretching (=CH) aromatic in the ring, the peak at frequency (1743)  $\text{cm}^{-1}$  is due to (C=O) ketone, and the band at (1280)  $\text{cm}^{-1}$  indicates the presence of methyl group in the Thiazole ring and the frequency band at (1620)  $\text{cm}^{-1}$  of the (C=N), we also note that is peak due to the presence of the azo group at (1535)  $\text{cm}^{-1}$ .

Figure (9), The spectrum of  $^1\text{H-NMR}$ <sup>[19]</sup> showed; Single signal at (10.7) ppm is return to the proton of phenolic hydroxyl group of the ring, the other single at the displacement (6.84, 6.82) ppm of the alkenes group (CH=CH), signal at (3.9) ppm is return to the

proton of (HC-N) and another multiple signal at (7.04-8.05) ppm of the protons aromatic carbon. Figure (10), The spectrum of  $^{13}\text{C}$ -NMR<sup>[20]</sup> was also showed; Signals at (131) ppm return to C5, signal at (21) ppm return to C7 and signal at (166) ppm return to C10, signal also at displacement (131) ppm return to C14, signal at (162) ppm return to C27, signal at (150) ppm return to C27, signal at (196) ppm return to C29, signal at (147) ppm return to C18, signals at displacement at (115-128) ppm returns to aromatic carbon.

Equation (4), represent this reaction. Preparation of additive 3B; Figure (11), The spectrum of the FT-IR<sup>[18]</sup> of the derivative (2 A) showed; The peak appearance at (3409)  $\text{cm}^{-1}$  to the stretching (OH), the peak at (3031)  $\text{cm}^{-1}$  returns to the stretching (=CH) aromatic in the ring, the peak at frequency (1743)  $\text{cm}^{-1}$  is due to (C=O) ketone, and the band at (1249)  $\text{cm}^{-1}$  indicates the presence of methyl group in the Thiazole ring and the frequency band at (1643)  $\text{cm}^{-1}$  of the (C=N), we also note that there are two peaks due to the presence of the azo group at (1458, 1535)  $\text{cm}^{-1}$ .

Figure (12), The spectrum of  $^1\text{H}$ -NMR<sup>[19]</sup> showed; Single signal at (9.5) ppm is return to the proton of phenolic hydroxyl group of the ring, the other single at the displacement (7.004, 7.01) ppm of the alkenes group (CH=CH), also triplet signal at (3.8) ppm return to the proton of (OCH<sub>3</sub>) and another signal at (2.8) ppm return to methyl group that bonded with Thiazole ring, and multiple signal at (7.02-8.4) ppm of the protons aromatic group. Figure (13), The spectrum of  $^{13}\text{C}$ -NMR<sup>[20]</sup> was also showed; Signals at (21) ppm return to C<sub>7</sub>, signal at (130) ppm return to C<sub>4</sub> and signal at (144) ppm return to C<sub>18</sub>, signal also at displacement (163) ppm return to C<sub>31</sub>, signal at (167) ppm return to C<sub>14</sub>, signal at (162) ppm return to C<sub>10</sub>, signal at (150) ppm return to C<sub>27</sub>, signal at (196) ppm return to C<sub>28</sub>, signal at (55) ppm return to C<sub>23</sub>, signals at displacement at (114.9-129) ppm returns to aromatic carbon.

Equation (5), represent this reaction. Preparation of additive 4B; Figure (14), The spectrum of  $^1\text{H}$ -NMR<sup>[19]</sup> showed; Single at (9.3) ppm is return to the proton of phenolic hydroxyl group of the ring, the other single at the displacement (2.2) ppm of the (N-C-Methyl), and singlet at displacement (1.8)

ppm return to (CH<sub>3</sub>), singlet at displacement (5.8) ppm return to (CH=N), and another multiple signal at (6.2-7.4) ppm of the protons aromatic group. Figure (15), The spectrum of  $^{13}\text{C}$ -NMR<sup>[20]</sup> was also showed; Two Signals at (21.1) ppm return to C<sub>7</sub>, signal at (131) ppm return to C<sub>4</sub> and signal at (138) ppm return to C<sub>18</sub>, signal also at displacement (159) ppm return to C<sub>28</sub>, signal at (166) ppm return to C<sub>10</sub>, signal at (172) ppm return to C<sub>24</sub> signals at displacement at (105-130) ppm returns to aromatic carbon.

Equation (6), represent this reaction. Preparation of additive 5B; Figure (16), The spectrum of  $^1\text{H}$ -NMR showed, Single at (9.3) ppm is return to the proton of phenolic hydroxyl group of the ring, the other single at the displacement (2.2) ppm of the (N-C-Methyl), and singlet at displacement (1.8) ppm return to (CH<sub>3</sub>), singlet at displacement (5.8) ppm return to (CH=N), and another multiple signal at (6.2-7.4) ppm of the protons aromatic group.

Figure (17), The spectrum of  $^{13}\text{C}$ -NMR was also showed (7) (Figure 15); Two Signals at (21.1) ppm return to C<sub>7</sub>, signal at (131) ppm return to C<sub>4</sub> and signal at (138) ppm return to C<sub>18</sub>, signal also at displacement (159) ppm return to C<sub>28</sub>, signal at (166) ppm return to C<sub>10</sub>, signal at (172) ppm return to C<sub>24</sub> signals at displacement at (105-130) ppm returns to aromatic carbon. Equation (7), represent this reaction. Preparation of additive 6B; Figure (18), The spectrum of the FT-IR of the derivative (4 B) showed, the peak appearance at (3417)  $\text{cm}^{-1}$  to the stretching (OH), the peak at (3100)  $\text{cm}^{-1}$  returns to the stretching (=CH) aromatic in the ring, the peak at frequency (1701)  $\text{cm}^{-1}$  is due to (C=O) ketone, and the frequency band at (1641)  $\text{cm}^{-1}$  of the (C=N), we also note that there are two peaks due to the presence of the azo group at (1555, 1541)  $\text{cm}^{-1}$ .

Figure (19), The spectrum of  $^1\text{H}$ -NMR showed, Single at (10.16) ppm is return to the proton of phenolic hydroxyl group of the ring, and singlet at displacement (1.74) ppm return to (CH<sub>3</sub>), singlet at displacement (8.3) ppm return to (CH=N), and another multiple signal at (6.2-8.9) ppm of the protons aromatic group. Figure (20), The spectrum of  $^{13}\text{C}$ -NMR was also showed, Two Signals at (21.1) ppm return to C<sub>19</sub>, signal at (148) ppm return to C<sub>1</sub> and signal at (131) ppm return to



C<sub>9</sub>, signal also at displacement (129.8) ppm return to C<sub>16</sub>, signal at (129.4) ppm return to C<sub>13</sub>, signals at displacement at (99.99-128) ppm returns to aromatic carbon. Equation (8), represent this reaction. Table (1) showed Physical properties of synthesis compounds.

### Measurement of LOI Using ASTM: D - 2863

The limiting oxygen Index (LOI) for unsaturated polyester resin without additives is (20.4) <sup>[18]</sup> and for epoxy resin without additives is (19.7) <sup>[19]</sup>; Tables (2&3) and Figures (21&22) respectively, indicated that, Oxygen concentration required to support a candle-like in unsaturated polyester and epoxy resins samples were increased with increasing the weight percentage of additives. The efficiency of additives studied was in the following order: 6B > 5B > 4B > 1B > 3B > 2B > B

### Measurement of Rate of Burning (R.B) Using ASTM: D-635:

The rate of burning (R.B) for unsaturated polyester resin without additives is (1.44) <sup>[20]</sup> and for epoxy resin without additives is (1.95) <sup>[21]</sup>; Results obtained from these tests showed that the rate of burning (R.B) of these resins with additives are of inversely proportional with the percentage weight of additives, as indicated in Tables (4&5), Figures (23&24) respectively, showed the flame speed curves of flame retardation for the resins.

The results obtained from these measurements correspond to the results obtained from the limiting oxygen index measurements. The efficiency of additives studied was in the following order: 6B > 5B > 4B > 1B > 3B > 2B > B.

Table 1: Physical properties of synthesis compound

No.	Compound	M.wt g.mol <sup>-1</sup>	m.p C°	Yield %	Color	R <sub>f</sub>
1	6-bromobenzothiazol-2-amine	697.10	203	73	Yellow	0.43
2	B	297.3	118.8	80	Dark brown	-
3	1B	445.49	Solution	66	Dark brown	0.38
4	2B	415.46	Solution	79	Black	0.42
5	3B	429.49	66.6	73	Dark brown	0.56
6	4B	467.34	solution	82	Black	0.57
7	5B	402.12	81	87	Dark brown	0.46
8	6B	467.34	solution	82	Black	0.57

Table 2: (LOI) for unsaturated polyester resin with group B of additives

LOI Additives	LOI %					
	Non	0.1	0.15	0.2	0.25	0.3
B	20.60	21.00	21.57	22.15	22.71	23.09
1B	20.60	22.25	22.97	23.78	24.49	24.75
2B	20.60	21.63	22.55	23.01	23.65	24.10
3B	20.60	21.80	22.62	23.15	23.78	24.21
4B	20.60	23.26	24.05	24.80	25.61	25.85
5B	20.60	23.55	24.43	25.32	26.04	26.32
6B	20.60	23.81	24.65	25.70	26.80	27.11

Table 3: (LOI) for epoxy resin with group B of additives

LOI Additives	LOI %					
	Non	0.1	0.15	0.2	0.25	0.3
B	19.54	20.44	21.02	21.66	22.53	23.07
1B	19.54	21.03	21.96	23.00	23.75	24.30
2B	19.54	20.61	21.18	21.83	22.98	23.43
3B	19.54	20.72	21.60	22.55	23.34	24.19
4B	19.54	21.28	22.29	23.28	24.00	24.85
5B	19.54	21.46	22.73	23.93	24.57	25.20
6B	19.54	21.85	23.12	24.27	24.95	25.61

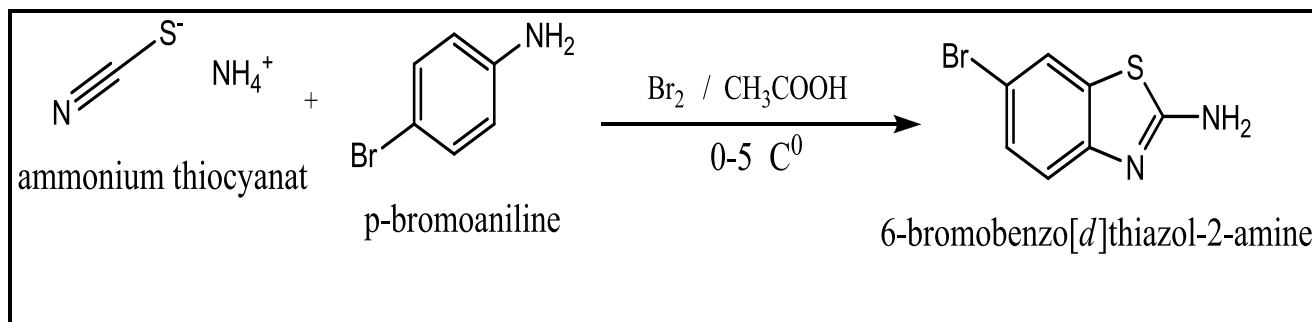
**Table 4: Rate of burning (R.B) for unsaturated polyester resin with group B of additives**

Tests	Additives %						Additives
	Non	0.1	0.15	0.2	0.25	0.3	
AEB (cm.)	10	10	10	10	9.1	7.6	B
	10	10	8.9	7.8	6.3	5.0	1B
	10	10	10	9.4	8.7	7.1	2B
	10	10	10	9.0	7.9	5.8	3B
	10	9.3	8.1	6.8	5.3	-	4B
	10	7.4	6.2	5.1	-	-	5B
	10	5.8	4.0	2.7	-	-	6B
ATB (Min.)	6.92	7.09	7.35	7.69	7.39	6.78	B
	6.92	10.20	10.22	10.68	9.69	14.70	1B
	6.92	7.25	7.63	7.58	7.37	6.69	2B
	6.92	8.40	8.77	8.33	7.82	7.07	3B
	6.92	10.22	9.87	10.00	10.19	-	4B
	6.92	8.91	8.61	9.81	-	-	5B
	6.92	9.06	8.33	7.50	-	-	6B
R.B. (cm./ Min.)	1.44	1.41	1.36	1.30	1.23	1.12	B
	1.44	0.98	0.87	0.73	0.65	0.34	1B
	1.44	1.38	1.31	1.24	1.18	1.06	2B
	1.44	1.19	1.14	1.08	1.01	0.82	3B
	1.44	0.91	0.82	0.68	0.52	-	4B
	1.44	0.83	0.72	0.52	-	-	5B
	1.44	0.64	0.48	0.36	-	-	6B
S.E	-	-	-	-	-	-	B
	-	-	-	yes	yes	yes	1B
	-	-	-	-	-	-	2B
	-	-	-	-	-	-	3B
	-	-	yes	yes	yes	yes	4B
	-	yes	yes	yes	yes	yes	5B
	-	yes	yes	yes	yes	yes	6B
N.B	-	-	-	-	-	-	B
	-	-	-	-	-	-	1B
	-	-	-	-	-	-	2B
	-	-	-	-	-	-	3B
	-	-	-	-	-	yes	4B
	-	-	-	-	yes	yes	5B
	-	-	-	-	yes	yes	6B

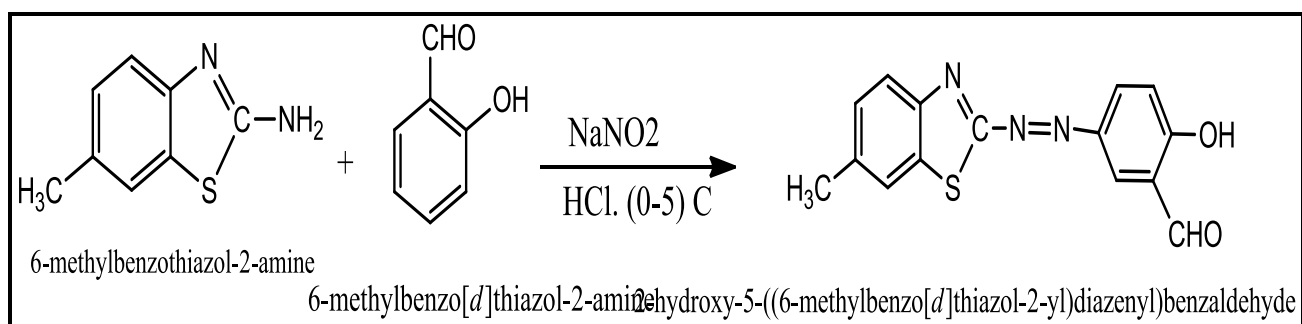
**Table 5: Rate of burning (R.B) for epoxy resin with group B of additives**

Tests	Additives %						Additives
	Non	0.1	0.15	0.2	0.25	0.3	
AEB (cm.)	10	10	10	10	9.4	8.6	B
	10	10	9.2	8.1	6.8	6.1	1B
	10	10	10	9.6	8.7	7.9	2B
	10	10	9.5	9.0	8.3	7.5	3B
	10	9.1	8.6	7.2	6.4	5.7	4B
	10	7.3	6.2	5.0	3.1	2.6	5B
	10	5.8	4.8	3.5	-	-	6B
ATB (Min.)	5.12	5.46	5.68	5.91	6.39	6.14	B
	5.12	7.19	7.13	6.98	6.47	6.10	1B
	5.12	6.21	6.53	6.95	6.90	6.52	2B
	5.12	6.84	6.88	7.20	7.41	7.01	3B
	5.12	8.05	7.88	6.99	6.52	7.13	4B
	5.12	6.75	6.96	6.95	5.43	5.77	5B
	5.12	6.82	7.05	6.86	-	-	6B
R.B. (cm./ Min.)	1.95	1.83	1.76	1.69	1.47	1.40	B
	1.95	1.39	1.29	1.16	1.05	1.00	1B
	1.95	1.61	1.53	1.38	1.26	1.21	2B
	1.95	1.46	1.38	1.25	1.12	1.07	3B
	1.95	1.13	1.09	1.03	0.85	0.80	4B
	1.95	1.08	0.89	0.72	0.57	0.45	5B
	1.95	0.85	0.68	0.51	-	-	6B
S.E	-	-	-	-	-	-	B
	-	-	-	-	-	-	1B
	-	-	-	-	-	-	2B
	-	-	-	-	-	-	3B
	-	-	-	-	yes	yes	4B
	-	-	-	yes	yes	yes	5B
	-	yes	yes	yes	yes	yes	6B
N.B	-	-	-	-	-	-	B
	-	-	-	-	-	-	1B
	-	-	-	-	-	-	2B

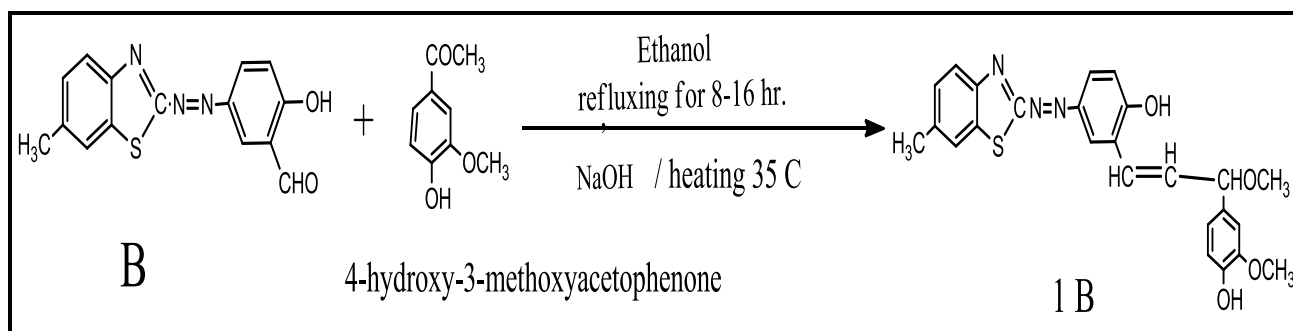
	-	-	-	-	-	-	3B
	-	-	-	-	-	-	4B
	-	-	-	-	-	-	5B
	-	-	-	-	yes	yes	6B



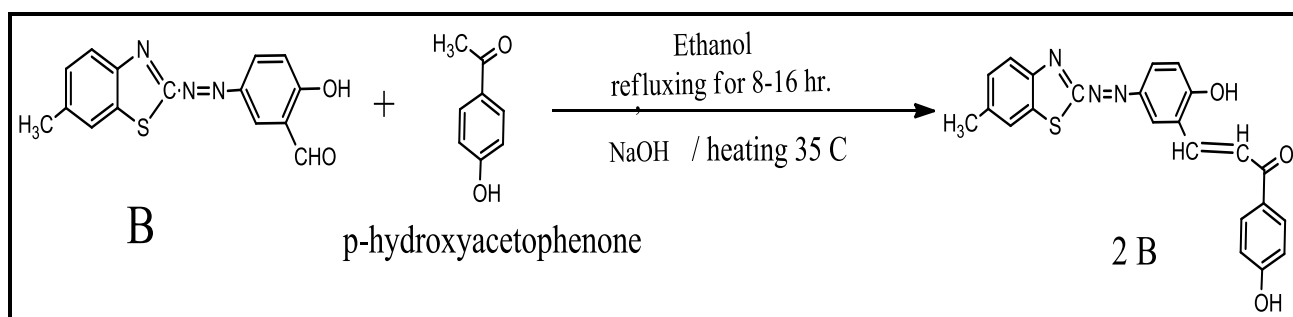
Equation 1: Synthesis of 6-bromobenzo[d]thiazol-2-amine



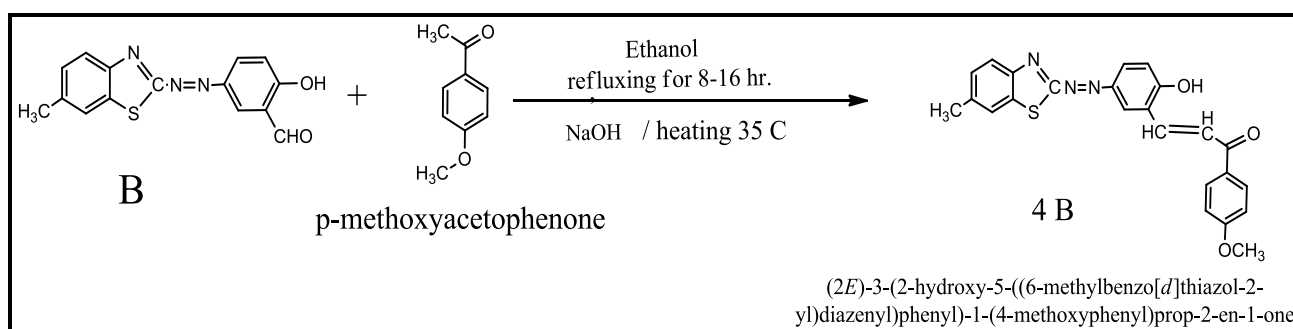
Equation 2: Synthesis of compound B



Equation 3: Synthesis of compound 1B (Chalcone)

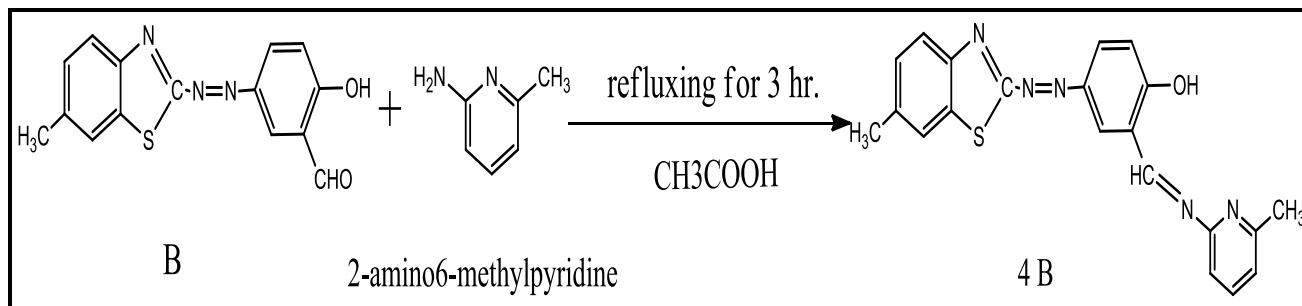


Equation 4: Synthesis of compound 2B (Chalcone)

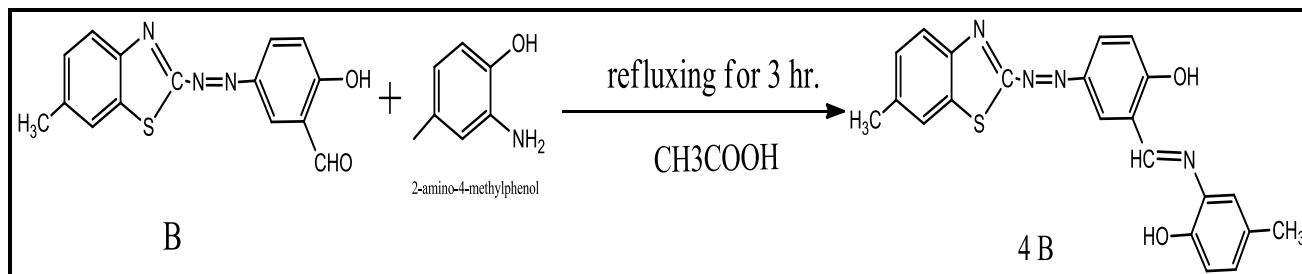


Equation 5: Synthesis of compound 3B (Chalcone)

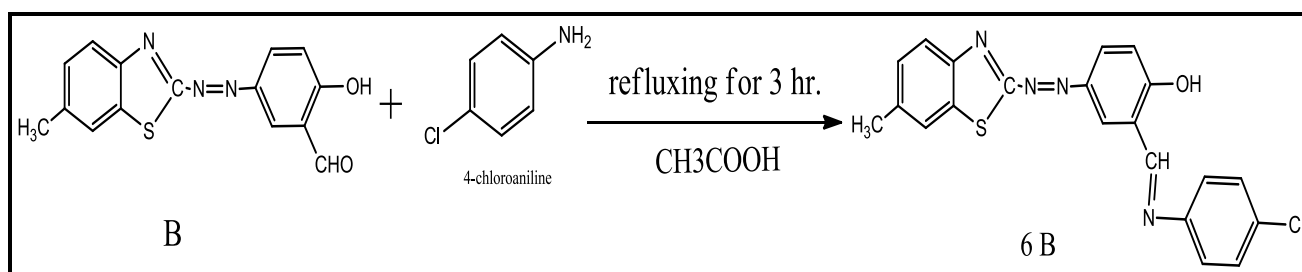




Equation 6: Synthesis of compound 4B (shiff base)



Equation 7: Synthesis of compound 5B (shiff base)



Equation 8: Synthesis of compound 6B (shiff base)

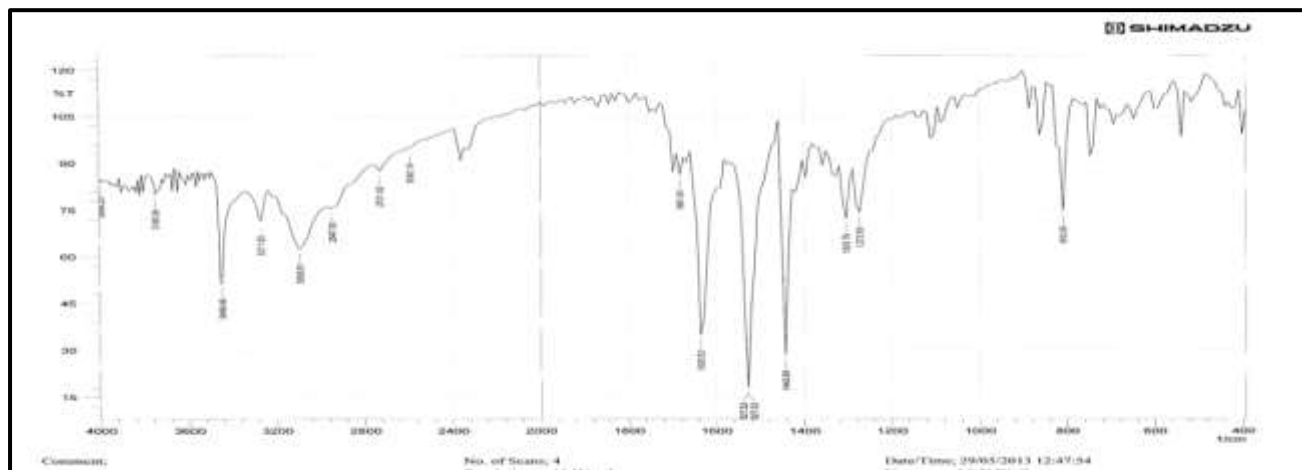


Figure 1: The FT-IR spectrum of compound 6-bromobenzothiazol-2-amine

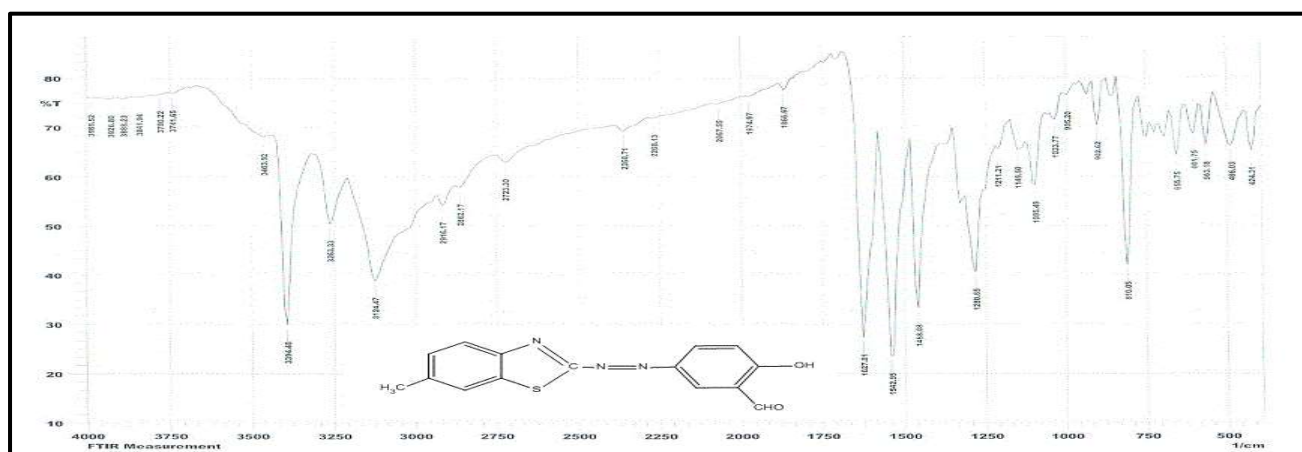


Figure 2: the FT-IR spectrum of compound B



Figure 3: The  $^1\text{H}$ -NMR spectrum of compound B

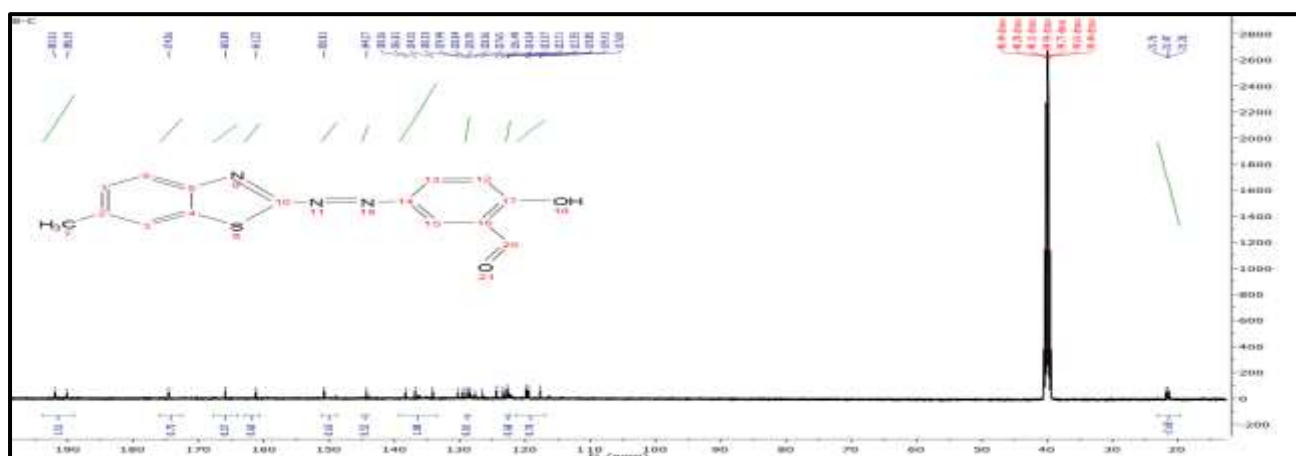


Figure 4: The  $^{13}\text{C}$ -NMR spectrum of compound B

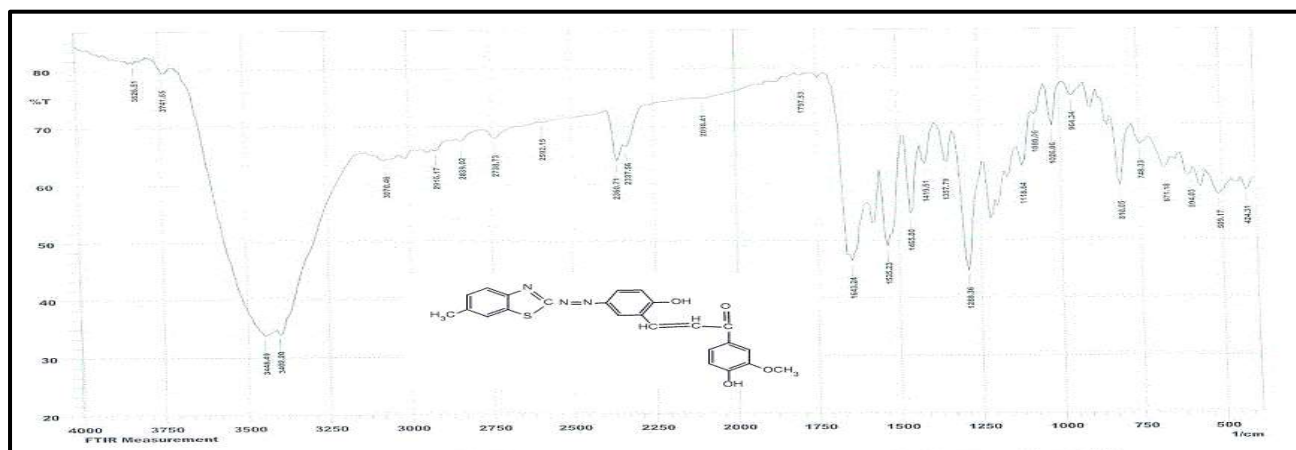


Figure 5: The FT-IR spectrum of compound 1B

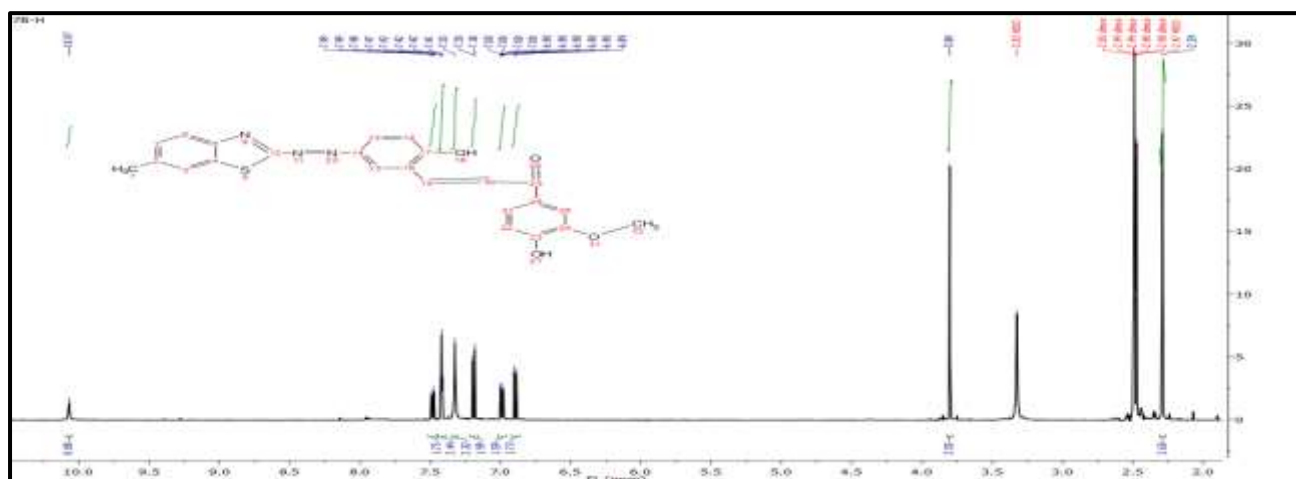


Figure 6: The  $^1\text{H}$ -NMR spectrum of compound 1B

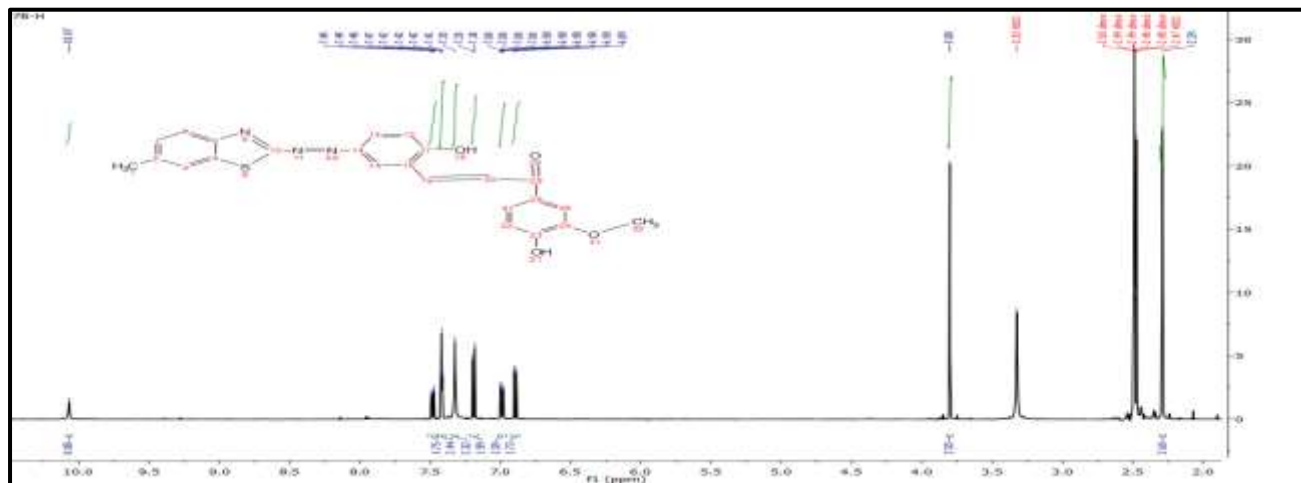
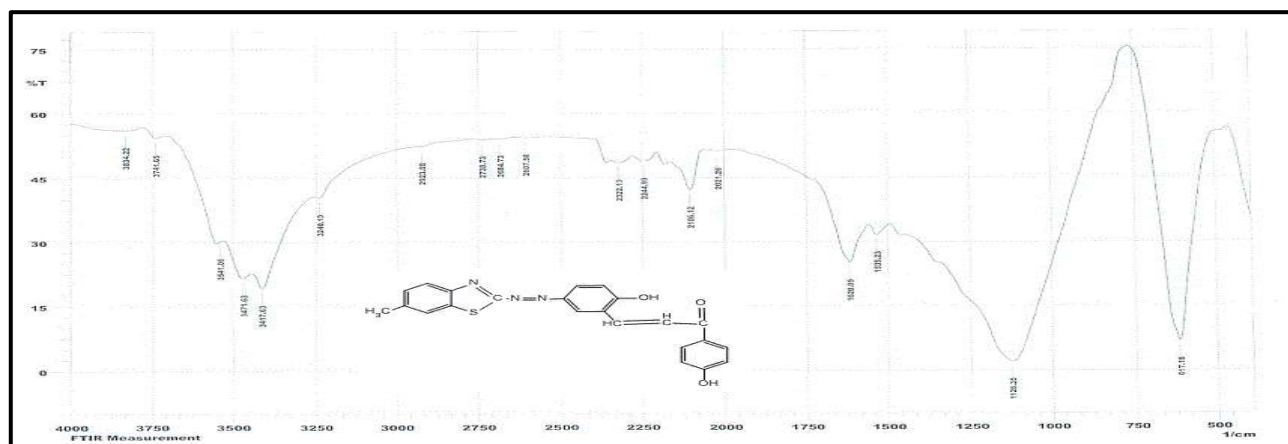


Figure 7: The  $^{13}\text{C}$ -NMR spectrum of compound 1B



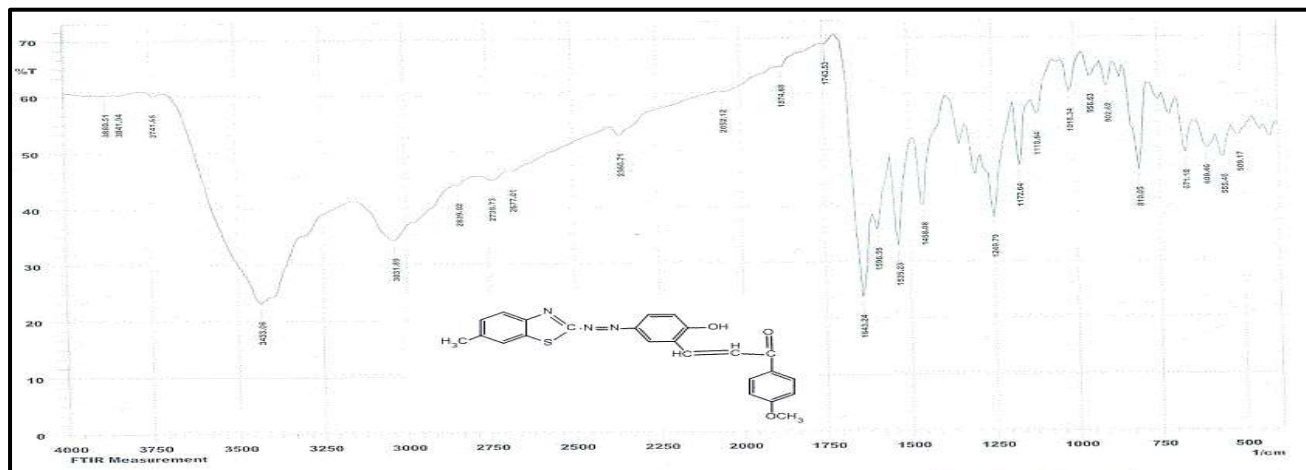


Figure 11: The FT-IR spectrum of compound 3B

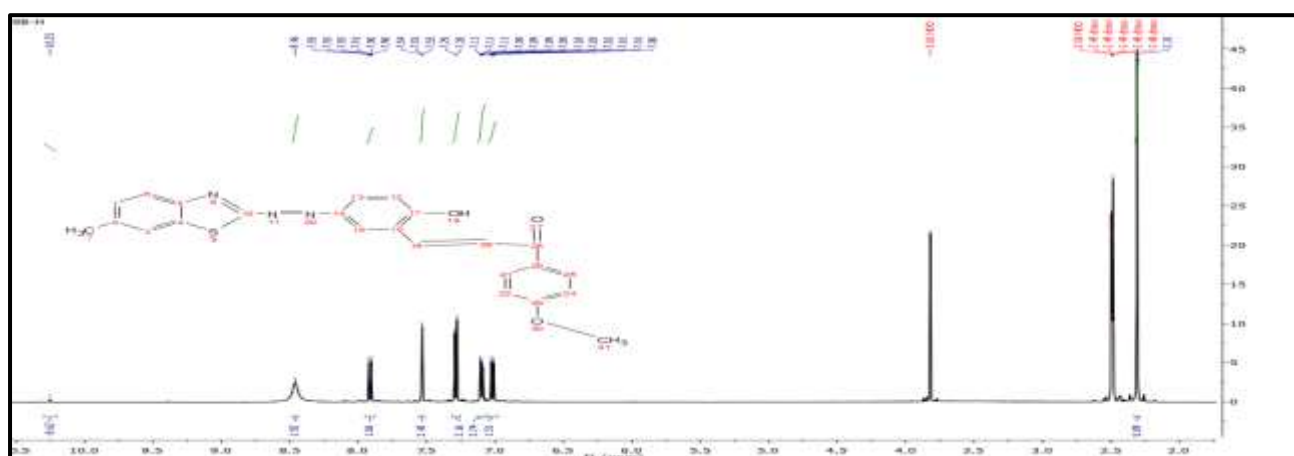


Figure 12: The <sup>1</sup>H-NMR spectrum of compound 3B

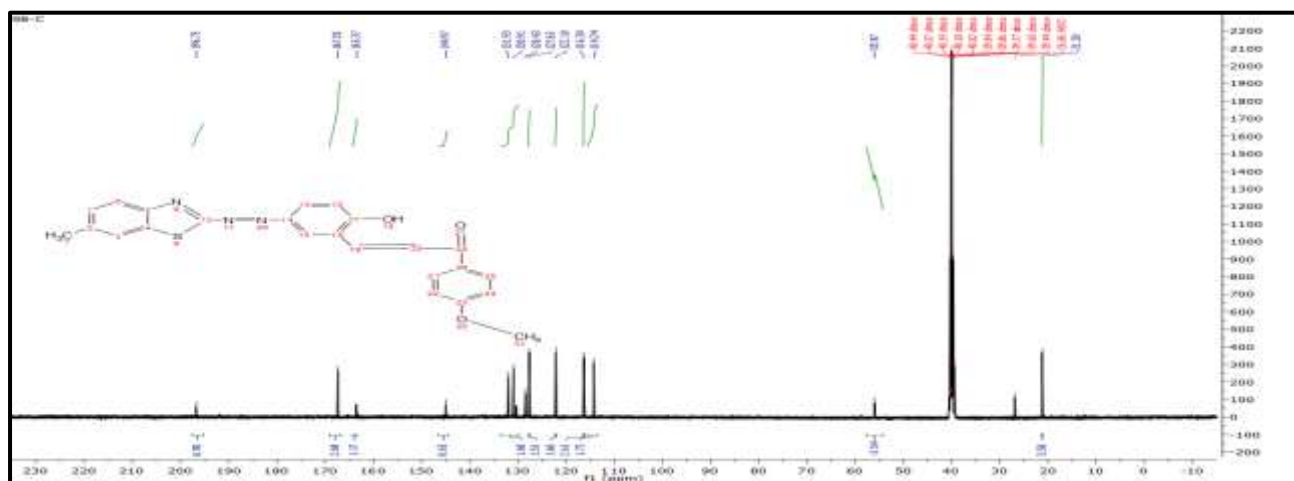


Figure 13: The <sup>13</sup>C-NMR spectrum of compound 3B

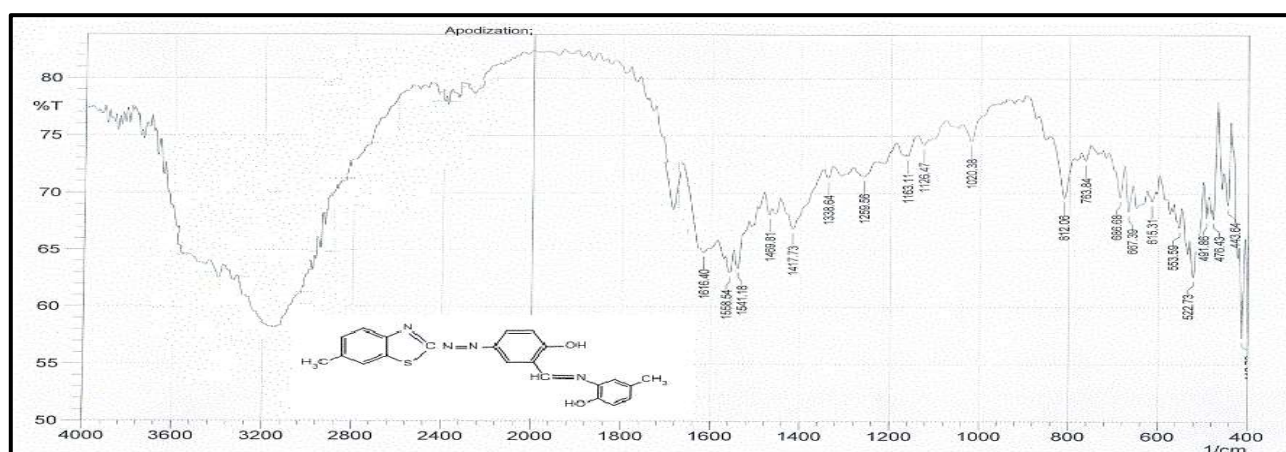


Figure 14: The FT-IR spectrum of compound 4B

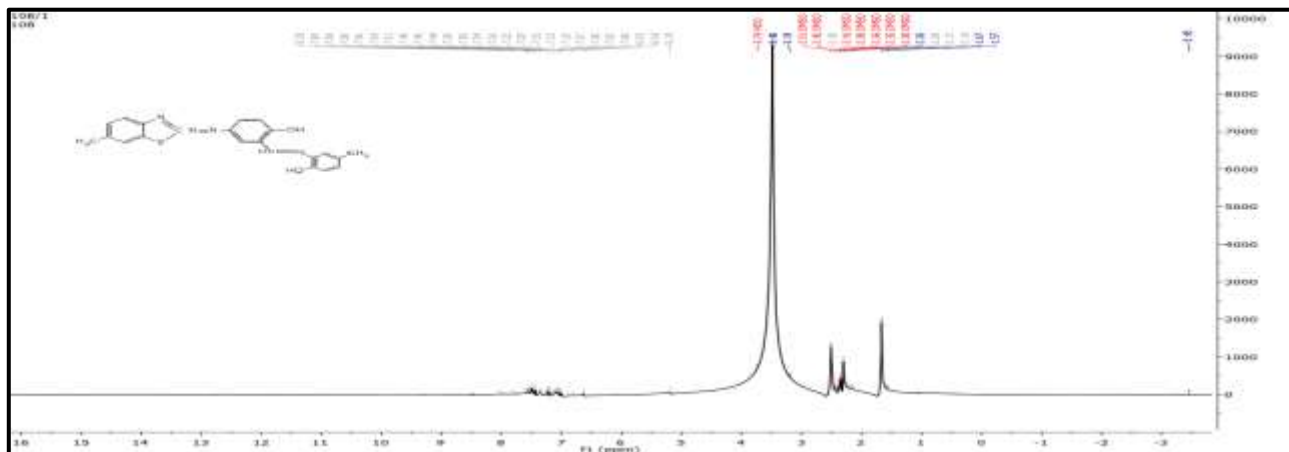


Figure 15: The  $^1\text{H}$ -NMR spectrum of compound 4B

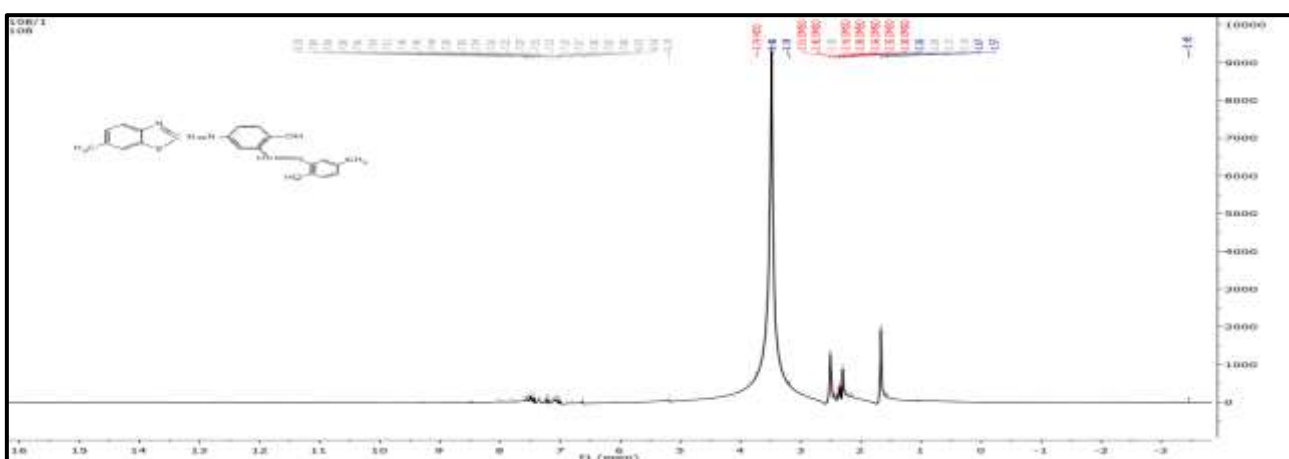


Figure 16: The  $^{13}\text{C}$ -NMR spectrum of compound 4B

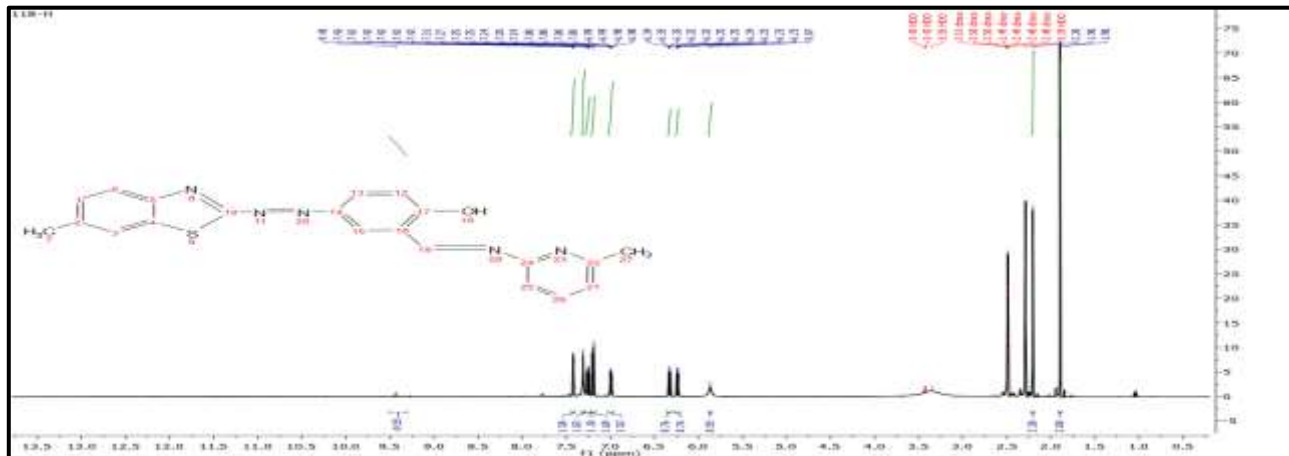


Figure 17: The  $^1\text{H}$ -NMR spectrum of compound 5B

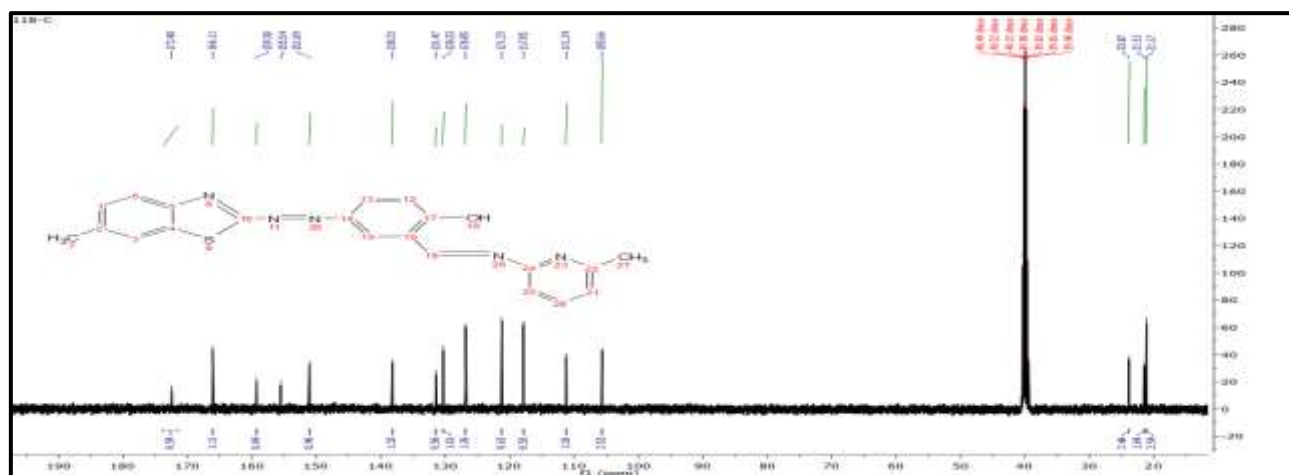


Figure 18: The  $^{13}\text{C}$ -NMR spectrum of compound 5B



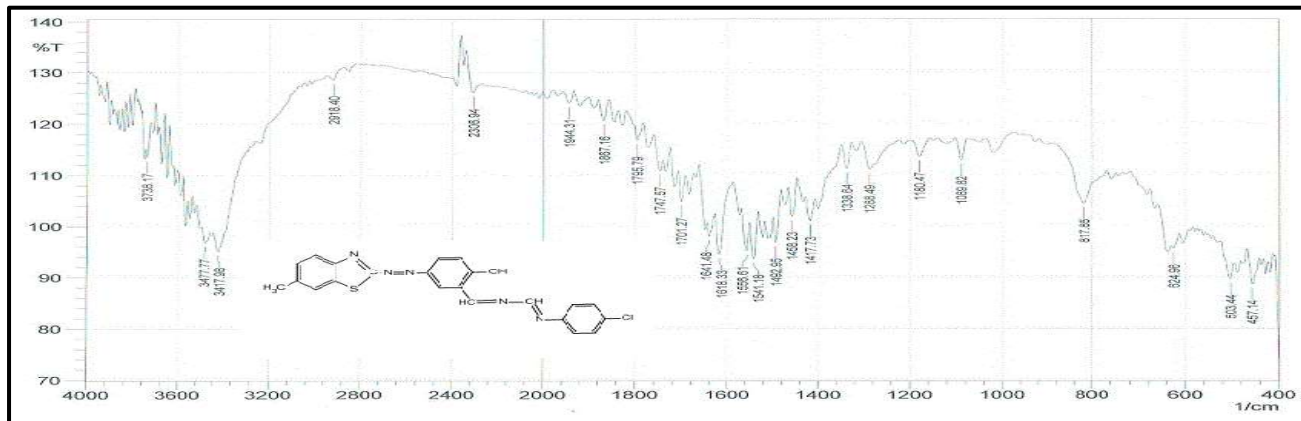


Figure 19: The FT-IR spectrum of compound 6B

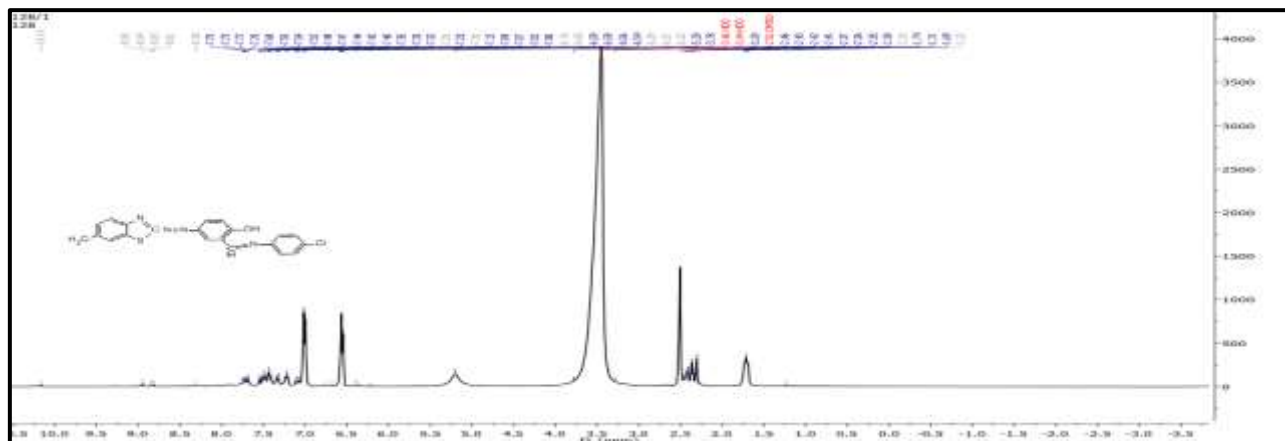


Figure 20: The <sup>1</sup>H-NMR spectrum of compound 6B

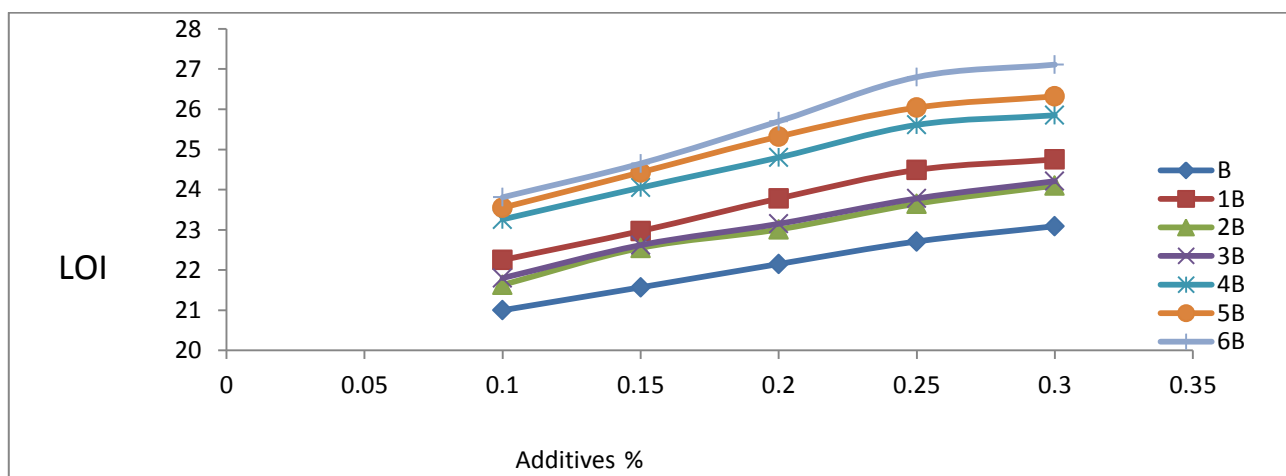


Figure 21: (LOI) for unsaturated polyester resin with group B of additives

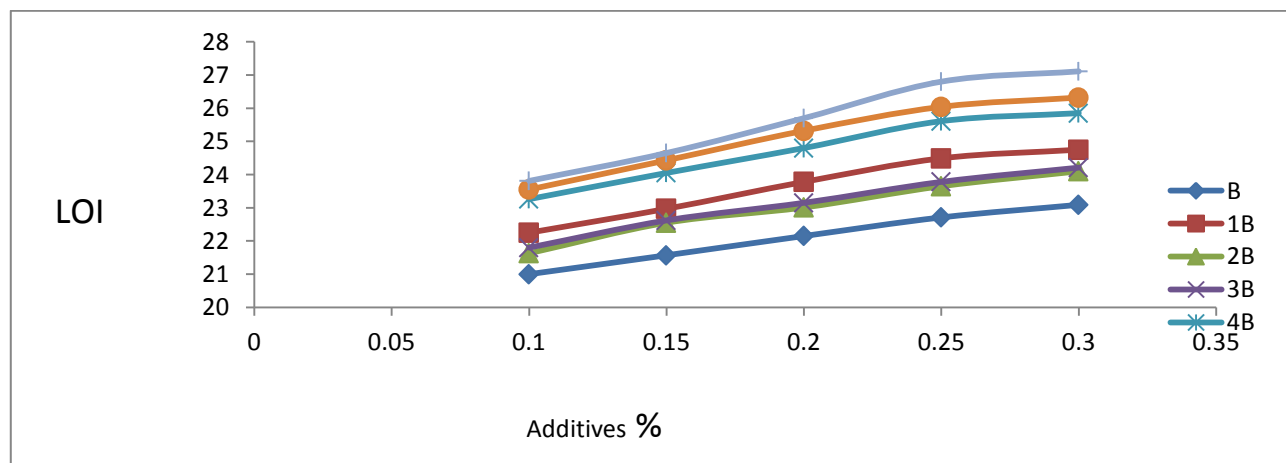


Figure 22: (LOI) for epoxy resin with group B of additives

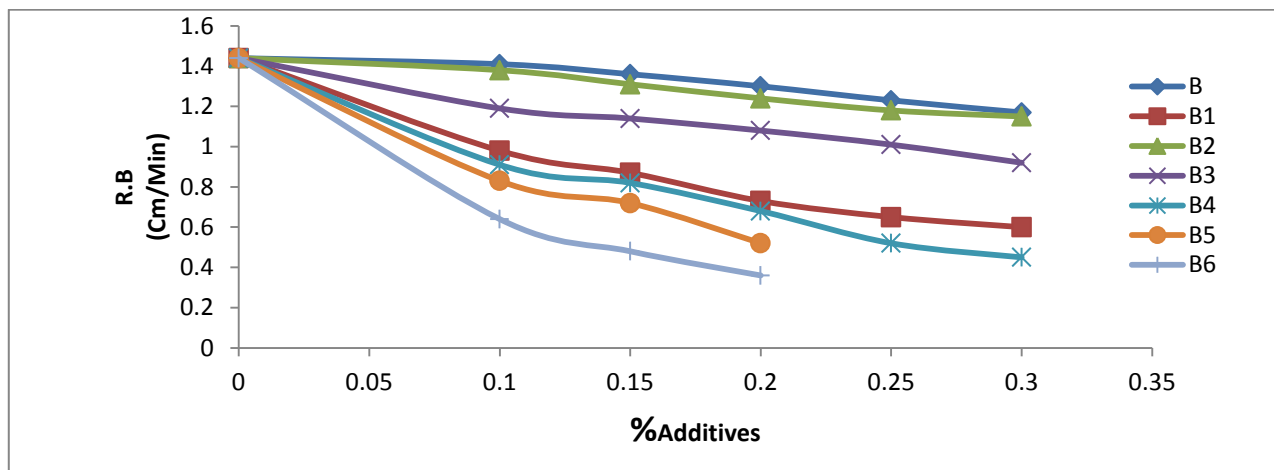


Figure 23: (R.B) for unsaturated polyester resin with group B of additives

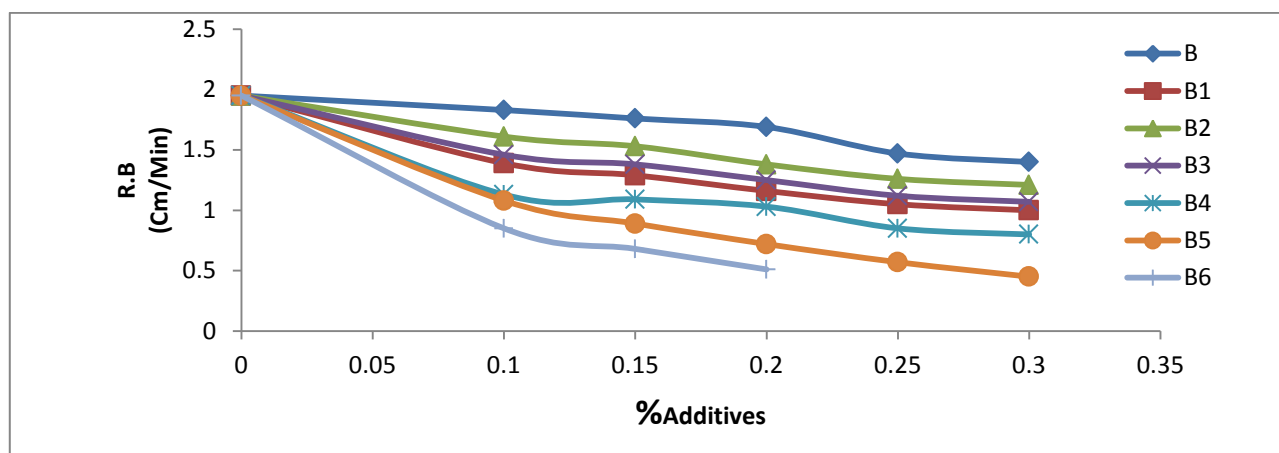


Figure 24: (R.B) for epoxy resin with group B of additives

## Conclusion

The results obtained can be summarized that, the limiting oxygen index (LOI) increased with increasing the weight percentages of the additives, as well as, the rate of burning (R.B), decreased with increasing the weight percentages of the additives. The flame retarding efficiency of the additives has the following order:

6B> 5B> 4B> 1B> 3B> 2B>B Apparently, the action of these additives due to by the formation of char as result of removing the hydrogen atoms from the polymer chain with formed the inert compounds. Finally, the combustion products like; free radicals, chare...etc., will form allayer to prevent burning and displacing oxygen that help continues burning of polymers.

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