

New anticorrosion and flame retardant coating compositions based on 4-amino triazole derivatives , their Schiff's bases and epoxy varnish

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ABSTRACT

In this study, some recently amino triazole derivatives mainly 4-amino-5-phenyl-2,4-dihydro-3H-1,2,4-triazole-3-thione(I), 4-((furan-2-yl-methylene)amino)-5-phenyl-2,4-dihydro-3H-1,2,4-triazole-3-thione(II), 4-((4-nitrobenzyl- idene)amino)-5-phenyl-2,4-dihydro-3H-1,2,4-triazole-3-thione (III), 4-((4-(dimethylamino)benzylidene)amino)-5-phenyl-2,4-dihydro-3H-1,2,4-triazole-3-thione (IV), 5-phenyl-4-((3-phenyl-4-((3-phenylallylidene) amino)-2,4-dihydro-3H-1,2,4-triazole-3-thione(V), 4-(((3-phenyl-5-thioxo-1,5-dihydro-4H-1,2,4-triazol-4-yl)imino) methyl)benzal-dehyde(VI), and 3-(((3-phenyl-5-thioxo-1,5-dihydro-4H-1,2,4-triazol-4-yl) imino) methylene) indolin-2-one (VII) were synthesized and have been characterized on the bases of FT- IR, 1H-NMR, mass spectrometer and elemental analyses. The synthesized amino triazole derivatives (I-VII) were physically inserted into epoxy resin formula by the ratio of [0.5%, 1.5%, 3%] to design a new green modified epoxy surface coating varnishes. The modified epoxy coating varnishes were evaluated using international standard test methods (ASTM), the main results are shown that; modified varnishes were considered a good anti-corrosion of mild steel coated by employing salt spray test, and a good flame retardant by using a limited oxygen index (LOI) method, also study the effect of this modification on some physical, chemical and mechanical properties of epoxy resin.

INTRODUCTION

Epoxy resin, as one of the most important thermosetting materials, has been widely used in coating, adhesives, aerospace and electronics industry due to its outstanding adhesion, lower density, high strength, good durability, excellent chemical resistance (Jiang et al., 2019). Organic coatings are widely used to prevent corrosion of metallic structures. It is generally believed that most organic coatings adhere to metals via hydrogen bonds or secondary bonds, these bonds develop when an organic coating and a metallic surface are brought closely together and so the binders with polar groups display excellent adhesion characteristics. The bond strength of epoxy resins on steel is strongly dependent on the hydroxyl group content. It was found that the epoxy compounds were dissociated between the phenoxy oxygen and the aliphatic carbon. The surface interactions were believed to be via the phenoxy and hydroxyl oxygen atoms (Nakazawa, 1994; Abd El-Khalik N, 2016, as illustrated in Fig.1.

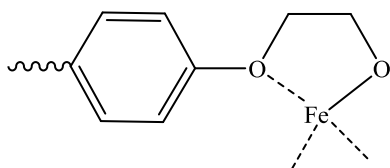


Figure 1: Adhesion of epoxy to steel by interfacial bonds

Mild steel is considered as one of the most commonly used in daily life. It is widely used in number of industries such as buildings construction; machines; pipelines; and bridges (Callister, 1997). Steel is suffering corrosion as results of these uses and hence it is destroyed easily. Thus, protection of steel from corrosion has attracted the attention of researchers. The application of inhibitors is one of the most common methods of corrosion protection and has been very effective, and low cost (Arshad et al., 2019; Obot et al., Xhanari et al., 2017; Prabakaran et al., 2017). The inhibitors interact with the metal surface and thereby adsorbed themselves on the metallic surface. The chemical structure of an inhibitor is closely related to its adsorption ability. Earlier studies have reported that organic compounds having hetero atoms, conjugated π -bonds and aromatic nucleus show appreciable efficiency to adsorption (Singh et al., 2018; Yıldız et al., 2019; Zhang et al., 2018; Guo et al., 2017). Thus, hetero-aromatic compounds have been widely studied as corrosion inhibitor because of having greater tendency to form metallic complex with metal (Singh et al., 2019).

Since 1,2,4-triazole and its derivatives possess a variety of biological activities including anti-bacterial (Shivananda and Prakash, 2011), antimicrobial (Al-Khuzai and Al-Majidi, 2016), antifungal (Zoumpoulakis et al., 2012), anticancer

and antitumor (Ma et al., 2014), anti-inflammatory (Murtaza et al., 2014), anticonvulsant (Plech et al., 2014), antiviral (Pandey et al., 2012), antitubercular (Singh et al., 2018), antioxidant (Barbuceanu et al., 2014) and as well as anticorrosive and flame retardance.

Triazoles ($C_2H_3N_3$) and their substituted derivatives are a group of heterocyclic compounds that attracted scientist's attention. Corrosion inhibitors derived from Schiff bases containing triazoles and its derivatives had taken a lot of concerns in the last years (Gopi et al., 2010; Swathi et al., 2017; Frolova. and Kuznetsov, 2009). These compounds can be considered as environment-friendly inhibitors due to their strong chemical activity and limited toxicity. Plus, they have a special tendency towards metal surface enabling them to replace water molecules localized on the metal surface. In addition, they have pairs of unshared electron and many π -electrons on the nitrogen atoms that can participate with d-orbitals of iron metal to provide such a protective film maintains the metal from corrosion (Bentiss et al., 1999). Encouraged by the previous reports and in continuation of our efforts in green chemistry, 1, 2, 4-triazole and a new series of novel Schiff base were designed, and the efficiency of the them were investigated as a green corrosion inhibitors for mild steel by applying salt spray technique.

Flame retardants are chemicals which are added to combustible materials to render them more resistant to ignition. Nitrogen-generating flame-retarding polymers have certain advantages over other flame-retarding polymers due to their low production of smoke and of toxic combustion compounds. Recently, triazole has been reported to release nitrogen gas during decomposition, and incorporating triazole into the bisphenol polymer can significantly enhance this polymer's thermal stability and flame retardancy (Poduval, 2014).

Nitrogen flame retardants are believed to act by several mechanisms: In the condensed phase, Nitrogen-containing compound is transformed into cross-linked structures which promote char formation. These are relatively stable at high temperatures and physically inhibit the decomposition of material in flammable gas. Ammonia is also released in these reactions. And a mechanism in the gas phase may be the release of molecular nitrogen which dilutes the volatile polymer decomposition products (Visakh and Yoshihiko, 2019).

In recent years, development of nonhalogenated flame retardants has been created as emerging area of research because of the blanket ban on halogenated flame retardants in view of the environmental and health concerns (Pereira and Martins, 2014; (Ravichandran et al., 2011)). So flame retardants based on nitrogen containing materials find scope as one of the alternatives to halogenated ones. Flame retardants can be either incorporated deposited onto polymer chain by physical means known as additive flame retardants or inserted into its backbone during the course of polymerization popularly known as reactive flame retardants.

In the present work, we aimed to prepare a novel 1,2,4-triazole and its Schiffs' bases functionalized flame retardants N-containing moieties for imparting a flame retardant properties to epoxy varnish, the objective behind the design of epoxy modified with triazole derivatives has been (a) promoting char, (b) nitrogen-rich 1,2,4-triazole moiety for the swelling of char because of the release of N_2 gas and (c) hydroxyl groups of terminal epoxy resin, All of the above factors constitute a hindrance that impedes heat to reach to combustible materials.

MATERIALS AND METHODS

Experimental materials

Epoxy resin was obtained from Pachin Paints Company-Egypt, all other chemicals used during the research work were sourced both from internationally or from local companies were of pure grade quality, and used without any additional purification. All laboratory chemicals were packed and distributed by El-Nasr Pharmaceutical Co. Egypt.

Synthesis of 1,2,4-triazole derivatives (I-VII)

Synthesis of 4-amino-3-phenyl-1,2,4-triazole-5-thione (I)

A mixture of benzoyl hydrazide derivative in 20ml of absolute ethanol, potassium hydroxide (5ml, 10% aqueous solution) and carbon disulphide (0.01mol) was cooled well then stirred (magnetic stirrer) for 4hrs. After that, it was added to the reaction mixture few drops of concentrated HCl. The product that separated was collected by filtration, then a mixture of this product (0.01 mol.) and hydrazine hydrate (0.02 mol.) in 3ml of water was refluxed for 4hrs. After complication of the reaction, it was diluted

with water and acidified with concentrated HCl to give 4-amino-3-phenyl-1,2,4-triazole-5-thione (I) as white ppt. The product was recrystallized from benzene in 50% yield; m.p. 175°C; IR(KBr): 3299-3190 cm^{-1} of NH_2 , NH; at 3113, 2941 cm^{-1} for CH; at 2756 cm^{-1} for SH; at 1633 cm^{-1} (s) for C=N and at 1231 cm^{-1} for C=S; mass spectrum of (I) showed molecular ion peak at m/e 192 (100%) which was already the base peak and this indicates the stability of 4-amino-3-phenyl-5-thio-1,2,4-triazole (I); Anal. Calcd. for $\text{C}_8\text{H}_8\text{SN}_4$ (192).

Element	C	H	N	S
Calculated %	50.00	4.16	29.16	16.66
Found %	51.10	4.25	30.06	17.56

Synthesis of 4-((furan-2-yl-methylene) amino)-5-phenyl-2,4-dihydro-3H-1,2,4-triazole-3-thione(II)

A mixture of equimolar quantities of (I) (0.01 mol.), (0.01mol.) of furfuraldehyde in 10ml of absolute ethanol was heated under reflux for 4hrs. After cooling, the product that separated was collected by filtration, washed well with water and dilute ethanol and recrystallized from benzene as a dark brown ppt. in 87% yield; m.p. 174°C; IR (KBr): 3101 cm^{-1} for NH, 3036, 2992, 2838 cm^{-1} for CH (aromatic and aliphatic), 2760 cm^{-1} for SH, 1617 cm^{-1} for C=N and characteristic absorption for C=S at the region 1189 cm^{-1} ; Anal. Calcd. for $\text{C}_{13}\text{H}_{10}\text{N}_4\text{OS}$ (270).

Element	C	H	N	S
Calculated %	57.77	3.703	20.74	11.85
Found %	58.67	3.79	21.64	12.75

Synthesis of 4-((4-nitrobenzyl- idene)amino)-5-phenyl-2,4-dihydro-3H-1,2,4-triazole-3-thione (III)

A mixture of equimolar quantities of (I) (0.01 mol.), (0.01mol.) of 4-nitro benzaldehyde in 10ml of absolute ethanol was heated under reflux for 4hrs. After cooling, the product that separated was collected by filtration, washed well with water and dilute ethanol and recrystallized from benzene as orange ppt. in 83% yield; m.p. 199-200°C; IR(KBr): 3114 cm^{-1} for NH, 2989, 2940, 2849 cm^{-1} for CH (aromatic and aliphatic), 2755 cm^{-1} for SH, 1628 cm^{-1} for C=N and characteristic absorption for C=S at the region 1270 cm^{-1} ; $^1\text{H-NMR}$ (DMSO- d_6) (δ ppm): 14.3 (s,1H-NH), 10.08 (s,1H-CH) and 7.54-8.38 (m,9H-Ar-H); Anal. Calcd. for $\text{C}_{15}\text{H}_{11}\text{N}_5\text{O}_2\text{S}$ (325).

Element	C	H	N	S
Calculated %	55.38	3.38	21.53	9.84

Found %	56.28	4.28	22.43	10.74
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Synthesis of 4-((4-(dimethylamino) benzylidene) amino)-5-phenyl-2,4-dihydro-3H-1,2,4-triazole-3-thione (IV)

A mixture of equimolar quantities of (I) (0.01 mol.), (0.01mol.) of 4-N,N-dimethylamino benzaldehyde in 10ml of absolute ethanol was heated under reflux for 4hrs. After cooling, the product that separated was collected by filtration, washed well with water and dilute ethanol and recrystallized from benzene as reddish violet ppt. in 98.9% yield; m.p. 182-184°C; IR (KBr): 3121 cm^{-1} for NH, 3036, 2993, 2956, 2836 cm^{-1} for CH (aromatic and aliphatic), 2764 cm^{-1} for SH, 1613 cm^{-1} for C=N and characteristic absorption for C=S at the region 1282 cm^{-1} ; Anal. Calcd. for $\text{C}_{17}\text{H}_{17}\text{N}_5\text{S}$ (323).

Element	C	H	N	S
Calculated %	63.15	5.26	21.67	9.907
Found %	64.04	6.16	22.57	9.18

Synthesis of 5-phenyl-4-((3-phenyl-4-((3-phenylallylidene)amino)-2,4-dihydro-3H-1,2,4-triazole-3-thione(V)

A mixture of equimolar quantities of (I) (0.01 mol.), (0.01mol.) of cinnamaldehyde in 10ml of absolute ethanol was heated under reflux for 4hrs. After cooling, the product that separated was collected by filtration, washed well with water and dilute ethanol and recrystallized from benzene as yellow crystal; in 54.6% yield; m.p. 170-172°C; IR (KBr): 3107 cm^{-1} for NH, 3076, 2929 cm^{-1} for CH (aromatic and aliphatic), 2770 cm^{-1} for SH, 1624 cm^{-1} for C=N and characteristic absorption for C=S at the region 1273 cm^{-1} ; Anal. Calcd. for $\text{C}_{17}\text{H}_{14}\text{N}_4\text{S}$ (306).

Element	C	H	N	S
Calculated %	66.66	4.57	18.30	10.45
Found %	67.56	5.47	19.20	11.35

Synthesis of 4-(((3-phenyl-5-thio-1,5-dihydro-4H-1,2,4-triazole-4-yl)imino)methyl)benzaldehyde (VI)

A mixture of equimolar quantities of (I) (0.01 mol.), (0.01mol.) of terphthaldehyde in 10ml of absolute ethanol was heated under reflux for 4hrs, cooling, filtration, washing well with water and dilute ethanol and recrystallisation from benzene as yellow ppt. in 99% yield; m.p. 168-170°C; IR(KBr): NH at 3141 cm^{-1} ; CH at 3096, 2951 cm^{-1} ; SH at 2759 cm^{-1} ; CHO at 1720 cm^{-1} (w); C=N at 1609 cm^{-1} ; C=S at

1276cm⁻¹; Mass spectrum of (VI) agreed well with the proposed structure. It showed molecular ion peak at m/e 308(39.6%) for C₁₆H₁₂N₄OS and a base peak at m/e 40.3(100%) for N₂C; Anal. Calcd. for C₁₆H₁₂N₄OS (308).

Element	C	H	N	S
Calculated %	62.33	3.89	18.18	10.38
Found %	62.32	3.98	18.27	10.47

Synthesis of 3-(((3-phenyl-5-thioxo-1,5-dihydro-4H-1,2,4-triazol-4-yl) imino)methylene)indolin-2-one(VII)

A mixture of equimolar quantities of (I) (0.01 mol.), (0.01mol.) of isatin in 10ml of absolute ethanol was heated under reflux for 4hrs. After cooling, the product that separated was collected by filtration, washed well with water and dilute ethanol and recrystallized from benzene orange ppt. in 99% yield; m.p. 154-156°C; IR(KBr): 3114 cm⁻¹ for NH, 3062, 2948, 2892, 2814 cm⁻¹ for CH (aromatic and aliphatic), 2757 cm⁻¹ for SH, 1731 cm⁻¹ for C=O in (isatin), 1616 cm⁻¹ for C=N and 1185 cm⁻¹ for C=S; ¹H-NMR (DMSO-d₆) (δ ppm): 13.9, 11.00 (s,1H-NH) and 6.88-8.04 (m,9H-Ar-H); Mass spectrum of (XIII) showed molecular ion peak at m/e 321 (2.86%) for M of C₁₆H₁₁N₅OS and the base peak at m/e 117 (100%) for C₇H₅N₂; Anal. Calcd. for C₁₆H₁₁N₅OS (321).

Element	C	H	N	S
Calculated %	59.81	3.42	21.806	9.96
Found %	60.71	4.32	22.71	10.86

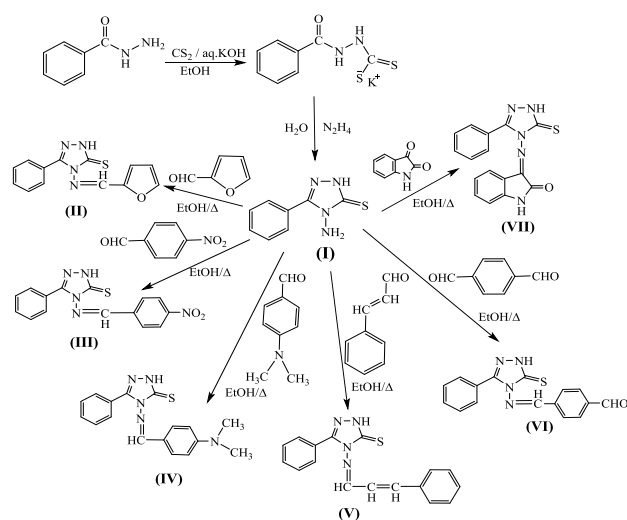


Figure 2: Preparation of epoxy paint films modified with 1,2,4-triazole derivatives (I-VII)
The coating compositions were prepared by physically incorporating triazole derivatives (I :

VII) on the ratio of (0.5, 1.5 and 3.0 %) by whight into epoxy varnish. The coating compositions were applied to steel, wood and glass panels by means of sweeping or brush. All efforts were made to maintain a uniform film thickness of 50 ±10μm for evaluating different properties.

Physical and mechanical testing of painting films

A range of physical and mechanical evaluations of the painted films were conducted according to appropriate American Society for Testing and Materials (ASTM) standard test methods.

a) Colour

The color of epoxy resin formulations was measured using the Gardner standard color scale (ASTM D1544-04).

b) preparation of glass test panel

Glass plates surface were free from grease by dipping into petroleum ether, cleaning by fine cloth, washing with distilled water, after that washed by ethyl alcohol and allowed to dry in air. The prepared steel panels measured by(ASTM D 3891).

c) measurement of film thickness

the film coating thickness measured by (ASTM D 1005-07). The instrument used for measuring the dry film thickness (DFT) was the "Rossmann Dial Thickness Gauge, Model 233".

d) Adhesion

The adhesion 'cross hatch' test measured by (ASTM D 3359-02). Adhesion is a very important property of a paint system, and some useful empirical information can obtain by the crosshatch test. If a coating is to fulfill its function of protecting or decorating a substrate, it must adhere to it for expected service life.

e) Flexibility

The flexibility "bend" test measured by (ASTM D 522-93a). The test performed to determine the adhesion power of the varnish to the substrate by bending apparatus. The varnish considered satisfactory if no marks for cracking or dislodging observed after bending.

f) Specular Gloss

Glass plates (100 mm- 150 mm), coated with the individual formulations, were utilized to measure the degree of gloss at an angle of 60° (ASTM D 523-08).

g) Hardness

The purpose of this test is to assess the ability of a surface coating to withstand scratching. The film hardness measured by means of the pencil test (ASTM D 3900).

Anti-corrosive performance evaluation of formulations through salt spray Accelerated Testing

1- a set of three coated steel panels to be tested were put in salt spray (Fog) cabinet at 30 °C and 100% humidity the test solution was 5% NaCl in water for 500 hours.

2- The films examined for any defects such as blistering, rusting, and scribe failure. (ASTM Method, D: 609).

a) Blistering

This test method employs photographic reference standard to evaluate the degree of blistering that may develop when paint systems are subject to condition, which will cause blistering. Degree of blistering of paints measured by (ASTM Method, D: 714-02).

b) Rust grade

The color photographic reference standards available for use with this method are representative of degree of rusting on painted steel (or iron) surface. The color photographic reference standards and the associated rust grade scale cover, only rusting not accompanied by blistering and evidenced by visible rust. The degree of rusting measured by (ASTM Method, D: 610-01).

c) Scribe failure

This test method covers the treatment of previously steel painted or coated specimens for accelerated and atmospheric exposure tests and their subsequent evaluation in respect to corrosion, blistering associated with corrosion, loss of adhesion at scribe mark, or other film failure. The scribe failure measured by (ASTM Method, D: 1654-92).

Flame-retardant testing method

The panels coated with epoxy varnish or modified varnish, were evaluated in a limited oxygen index (LOI) chamber. (LOI) values were determined using the standardized test methods (ASTM Method, D: 2863).

RESULTS AND DISCUSSION

Regrettably, epoxy resins tend to burn easily with high smoke and gas release. Therefore, this work is concerned with demonstrating enhancing the flame retardants of epoxy resins by adding some 1,2,4-

triazole derivatives, which act as normal flame retardants containing nitrogen and sulphur, as well as a green corrosive inhibitor.

Synthesis and characterization of 4-amino-5-phenyl-1,2,4-triazole-3-thione derivatives and corresponding Schiff's bases:-

In the present paper, a 4-amino-5-phenyl-1,2,4-triazole-3-thione (I) was prepared followed by condensation with different aldehydes or ketones to get their Schiff base derivatives (II – VII) aiming to explore their fire retardant and anticorrosion properties. The synthesis of triazole derivatives (I – VII) was performed according to indications mentioned in the experimental section and shown in figure 2. The compounds were obtained with yields (50-99%) with high purity. The formation of 1,2,4-triazole derivatives was proven by elemental analysis, FTIR and ¹H NMR, as well as by mass spectrometry for the compounds.

Chemical properties:-

Preliminary evaluation studies are carried out for all formulations toward water, acid, alkali, and solvent resistances. The evaluation tests are conducted according to standard methods. Rating of water, acid, alkali and solvent resistances were carried out according to the following statements:

Ex	means	Excellent	, Almost no change,
G	means	Good	, Very slight change,
F	means	Fair	, Partially attacked,
P	means	Poor	, Complete film failure.

The extensive evaluation studies for chemical resistances were given in Tables 1-4. The tests were carried out on actual coating films for 60 days immersion periods and the films are examined for any defects.

The data for the extensive evaluation studies given in Tables 1-4 indicate that: All dried films exhibited an excellent immovability towards solvent resistance. The presence of triazole derivatives as modifiers led to increased resistance of dry films toward water, acid and alkali. Increasing the percentage of modifier leads to improvement in the film resistances towards water, acid, and alkali. All films examined for water, alkali and acid were almost unaffected for 30 days excepted blank. The modifier (VII) had highest film resistance towards acid, and alkali.

Table 1: Water resistance data of various modified epoxy resins

Sample		Time per day													
		7	10	14	18	22	26	30	34	38	42	46	50	55	60
Blank		Ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	G
I	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G
II	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
III	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G
IV	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G
V	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G
VI	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	V.G
VII	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex

Table 2: Acid resistance data of various modified epoxy resins

Sample		Time per day													
		7	10	14	18	22	26	30	34	38	42	46	50	55	60
Blank		ex	Ex	ex	ex	ex	V.G	V.G	G	G	G	f	f	p	p
I	0.5%	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G
II	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	G	G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	V.G
III	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	V.G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
IV	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	G	G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
V	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.GV.	V.G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	G	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
VI	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G
VII	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	V.G	V.G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex

Table 3: Alkali resistance data of various modified epoxy resins

Sample		Time per day													
		7	10	14	18	22	26	30	34	38	42	46	50	55	60
	Blank	ex	Ex	ex	ex	ex	ex	ex	V.G	V.G	G	G	p	P	P
I	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	G	G	P
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	G	G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	V.G
II	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	V.G
III	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
IV	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	G	G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	G	G	G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G
V	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	G	P	P	P
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	G	G	P
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	G	G	G
VI	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	G	G	P
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	G	G	G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G
VII	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G	V.G	V.G	G
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	V.G	V.G
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex

Table 4: Solvent resistance data of various modified epoxy resins

Sample		Time per day													
		7	10	14	18	22	26	30	34	38	42	46	50	55	60
	Blank	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
I	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
II	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
III	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
IV	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
V	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
VI	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
VII	0.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	1.5 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex
	3 %	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex	ex

Physical and mechanical testing of painting films

The effects of adding triazole derivatives to the epoxy varnish, in respect of to the physical and mechanical properties, were evaluated as per the standard test methods. This was done to ascertain any negative aspects that might arise due to the presence of the additives. The physical evaluation

included drying time, colour, gloss, scratch hardness, cross hatch adhesion and flexibility were all measured.

Drying and colour characteristics

The drying characteristic of all formulations are carried out in air and colour was measured with

Gardnars tube, which consists of 18 colors numbered from (1 to 18). The method determines the color by comparing with standards of definite color compositions.

Table 5: Effect of modification samples (I-VII) on air drying time and colour of the epoxy varnish

Sample	Modif. %	Drying time		Colour
		Hours	Minutes	
Blank	---	25	15	6
Epoxy modified with I	0.5	13	-	6
	1.5	11	-	6 ⁺
	3	10	-	8
Epoxy modified with II	0.5	25	-	14
	1.5	24	-	15
	3	20	30	16
Epoxy modified with III	0.5	22	-	10
	1.5	18	-	11
	3	15	-	11
Epoxy modified with IV	0.5	17	-	10
	1.5	15	-	10
	3	14	-	10
Epoxy modified with V	0.5	24	15	12
	1.5	15	30	13
	3	11	-	14
Epoxy modified with VI	0.5	17	-	14
	1.5	14	-	14
	3	13	-	15
Epoxy modified with VII	0.5	10	-	<18
	1.5	9	15	<18
	3	8	30	<18

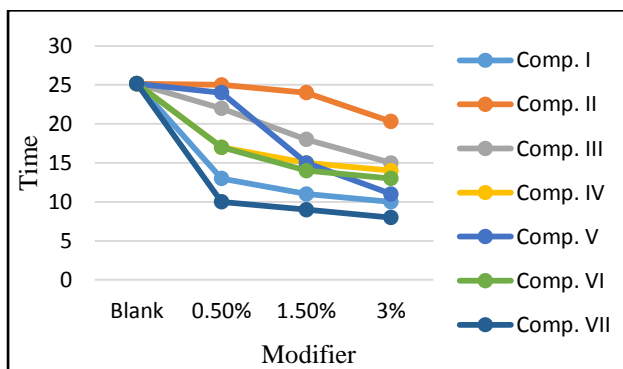


Figure 2: Effect of modification samples (I-VII) on air drying time

The most important finding from the characteristic data given in table 5 and figures 2 it is clear that :

- The air drying time of modified epoxy varnish were more shorter than that of unmodified epoxy one.

- As the percentage of modifiers increase the drying time decreased.
- The colour of epoxy resin is slightly darkens by adding modifiers.

Film performance

All films had a transparent and homogeneous appearance. All physical outcomes are listed in table 6.

Cross-hatch adhesion test

This test is in conformity according to (ASTM D 3359-02) where adhesion is assessed on a 0 to 5 scale. When study the hardness property of the neat epoxy and modified epoxy coating films, it was observed that all the coated film demonstrated a good cross-hatch adhesion. The triazole derivative compounds (I-VII) additive did not change the adhesion properties of the epoxy varnish formulation.

Flexibility (bend) test

This test method covers the determination of the resistance to cracking (flexibility) of organic coatings on substrates of sheet metal or rubber-type materials. The films for all the coating compositions passed the ¼ inch Mandrel bend test. The varnish was considered satisfactory if no cracking marks, or dislodging, is observed following the bending procedure. It is clear from the obtained results that the adding of triazole derivatives didn't affect on the flexibility of the coating film, it can be stated that all the films showed a good flexibility.

Specular gloss

This was measured using a Sheen UK gloss meter, The obtained results in table 6 clearly indicate that, triazole (III) does not change epoxy varnish gloss, while with adding triazoles (I, II, IV, and VI) there is a reasonable increase in gloss level, but presence of modifiers (VII, and V) lead to decrease the epoxy varnish gloss.

Scratch hardness test

This test method covers a procedure for rapid, inexpensive determination of the film hardness of an organic coating on a substrate in terms of drawing leads or pencil leads of known hardness, scratch hardness was determined by using a sheen UK hardness tester. The scratch was observed to vary

between 1.5: 2 Kg; it is clear from the results obtained that triazole derivative compounds (I-VII)

increase the scratch hardness of the epoxy coating, except triazole (V).

Table 6: Physical and mechanical characteristics of modified epoxy varnish with 1,2,4- triazole derivatives (compounds I- VII)

Sample	%	Adhesion	Flexibility	Gloss at 60°	Hardnes (Kg)
Blank	-	5B	Pass	83	1.5
Epoxy with comp. I	0.5%	5B	Pass	86	2
	1.5%	5B		98	2
	3%	5B		103	2
Epoxy with comp. II	0.5%	5B	Pass	90	2
	1.5%	5B		109	2
	3%	5B		109	2
Epoxy with comp. III	0.5%	5B	Pass	82	2
	1.5%	5B		83	2
	3%	5B		83	2
Epoxy with comp. IV	0.5%	5B	Pass	80	2
	1.5%	5B		85	2
	3%	5B		99	2
Epoxy with comp. V	0.5%	5B	Pass	83	1.5
	1.5%	5B		77	1.5
	3%	5B		67	1.5
Epoxy with comp. VI	0.5%	5B	Pass	82	2
	1.5%	5B		86	2
	3%	5B		92	2
Epoxy with comp. VII	0.5%	5B	Pass	74	1.5
	1.5%	5B		66	2
	3%	5B		45	2

Evaluation of triazole modifiers compound as corrosion inhibitors

After Physically incorporating of triazole compounds (I-VII) in epoxy resin varnish, it would expected improve the corrosion resistance .Since the amines usually used as an organic inhibitors that affected the surface of metal when present in sufficient concentration. These types of inhibitors work on decreasing the permeability of coating to water , oxygen and aggressive (such as chlorides and sulphates) via improving adhesion, adsorption onto the metal surface ,forming an insoluble complex salts with metal and promoting the growth and stability of a passivating layer.

The modified epoxy paint compositions were prepared by means of incorporating triazole derivatives (I -VII) by the ratio of (0.5, 1.5 and 3.0 %) by weight into epoxy paint and adjusted film thickness about 50 ± 5µm were evaluated as the anti-corrosive. After preparing the coated test panels

according to experimental section, they were exposed to salt spray tests (salt fog) according to ASTM for 500 h. The blistering size is graded from 10 to 0, where 10 represented no blistering and 0 represented the largest blister. Blistering frequency is denoted by F, M, MD and D (few, medium, medium dense and dense). Painted, or coated, specimens subjected to a corrosive environment are also evaluated by recording the average maximum and minimum creep age from the scribe mark. Scribe failure is also rated on a scale from 10 to 0, with 10 being (zero mm) from the scribe mark and 0 is (16 mm) from the scribe mark. Finally, the comparison of the surface appearance, to determine the percentage of the area that has been rusted, is also used. The rust grade is rated on a scale from 10 to 0, where10 is non-rusting and 0 is severing rusting. The corrosion resistance of the painted films of modified sample is given in Table 7 and Figures (2:); explains the photo of the painted films after salt test spray (5% of NaCl).

Table 7: Anticorrosive activities of modified epoxy varnish with new triazole derivatives (compounds I- VII)

Sample	modifier %	Blistering Size	Blistering frequency	Scribe failure(mm)	Rust grade
Blank	-	2	M	14	1
Epoxy with comp. I	0.5%	4	D	6	2
	1.5%	4	D	2	5
	3%	6	MD	2	7
Epoxy with comp. II	0.5%	7	M	4	5
	1.5%	7	M	4	5
	3%	8	M	4	5
Epoxy with comp. III	0.5%	3	D	10	3
	1.5%	3	MD	7	6
	3%	6	F	6	7
Epoxy with comp. IV	0.5%	3	D	9	3
	1.5%	5	D	5	3
	3%	6	F	5	2
Epoxy with comp. V	0.5%	4	D	11	3
	1.5%	5	M	9	3
	3%	7	F	5	3
Epoxy with comp. VI	0.5%	3	M	9	3
	1.5%	3	MD	5	5
	3%	5	M	3	7
Epoxy with comp. VII	0.5%	5	F	7	4
	1.5%	7	F	7	4
	3%	8	F	3	7

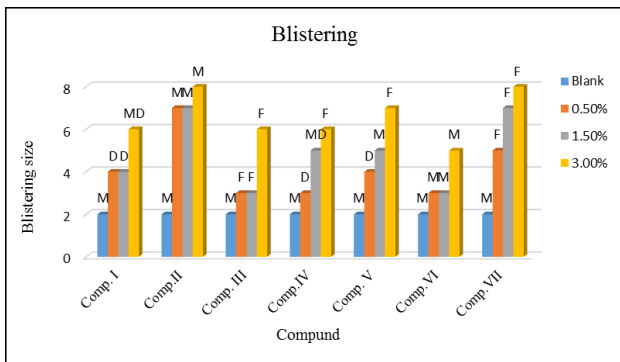


Figure 3: Effect of modification samples (I-VII) on blistering of corrosion

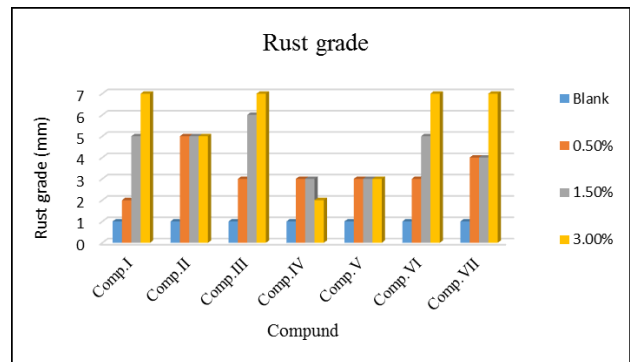


Figure 5: Effect of modification samples (I-VII) on rust grade of corrosion

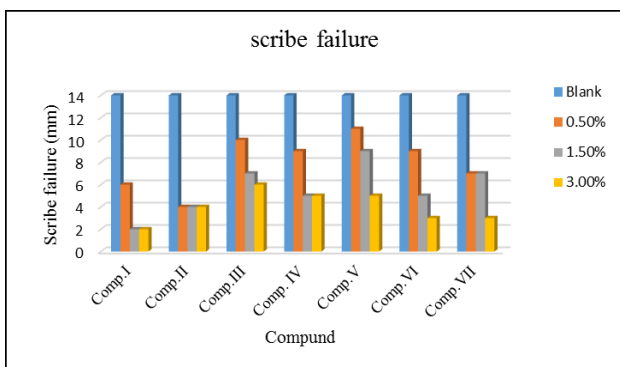


Figure 4: Effect of modification samples (I-VII) on scribe failure of corrosion

The data given in table 7 and figure.3 shown that all samble improve the blisterng and as the concentration of modifier increase blistering resistance increase particularly composite. It is clear that the adding of modifiers improve the scribe vauve from 14 mm within epoxy varnish alone to 2 mm with in epoxy varnish modived 3% with compound I (Figure 4)

It is also clear that the adding of modifiers improve the rust grade from 7 mm within epoxy varnish modified 3% in VII, VI and III , 0.5% in I and alon to 1 mm within epoxy varnish.

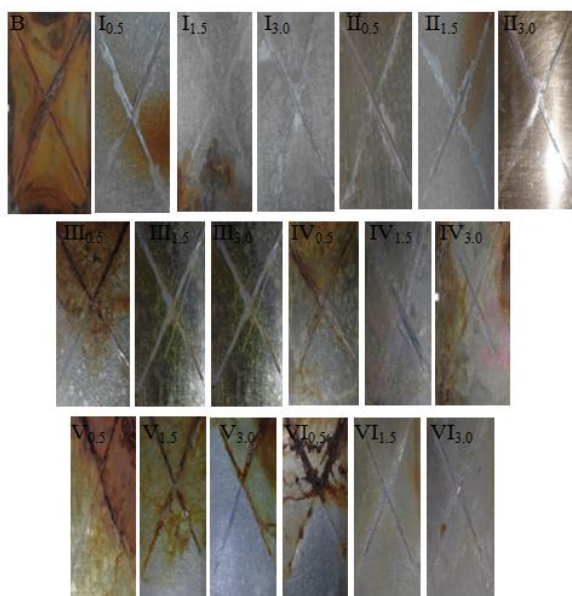


Figure 6: explains the photo of the painted films after salt spray test

Evaluation of triazole modifiers compound as flame retardant

Flame retardancy are materials that are conducted to provide fire protection for flammable consumer goods, as well as to mitigate fire growth in a wide range of fires. This paper is in the field of applied

science that is concerned with the study of flame retardants.

The LOI is defined as the minimum concentration oxygen, expressed as a percentage that will support combustion of a polymer. It is measured by passing a mixture of oxygen and nitrogen over a burning specimen, and reducing the oxygen level until a critical level is reached. The LOI is expressed as:

$$LOI = 100 \frac{[O_2]}{[O_2] - [N_2]}$$

It is clear that the incorporation of triazole derivative compounds (I-VII) which physically added into epoxy varnish in the ratios mentioned in the experimental section, the results which tabled in table 8 and fig.7 showed that triazole modifiers have an excellent flame retardancy when compared with epoxy blank sample, and as the modifiers percentage increase the value of flame retardancy increase.

The high value of flame retardation is due to the large molecular weight of triazole derivatives as well as its containment on nitrogen and sulfur atoms. The flame retardant efficiency of triazole derivatives compound (I-VII) was in the following order [VII > III > II > IV > V > I > VI].

Table 8: The effect of LOI on modified epoxy varnish with new triazole derivatives (compounds I- VII)

Sample	Modifier%	Limiting Oxygen Index (LOI)	Sample	Modifier%	Limiting Oxygen Index (LOI)
Blank	---	20	Epoxy modified with IV	0.5	28
				1.5	36
				3	42
Epoxy modified with I	0.5	27	Epoxy modified with V	0.5	26
	1.5	32		1.5	33
	3	40		3	41
Epoxy modified with II	0.5	40	Epoxy modified with VI	0.5	23
	1.5	46		1.5	26
	3	50		3	33
Epoxy modified with III	0.5	42	Epoxy modified with VII	0.5	43
	1.5	47		1.5	50
	3	52		3	56

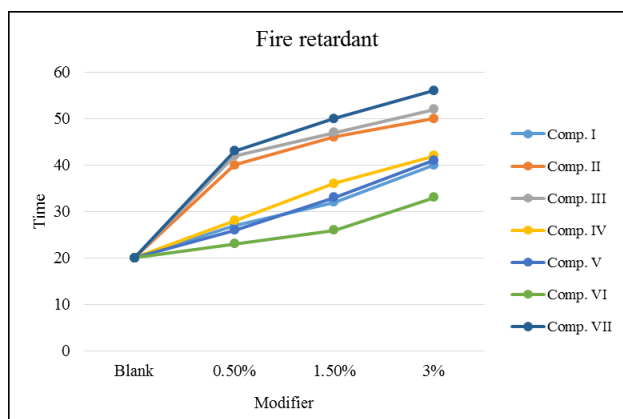


Figure 7: Effect of modification samples (I-VII) on LOI

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