New Flame Retardant and Antimicrobial Paints Based on Epoxy Paint Incorporated by Hexachlorocyclodiphosphazane Derivatives for Protective Coating

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Abstract: In this particular research, hexachlorocyclodiphosphazane derivatives types (I-II) were synthesized for use as a flame retardant and antimicrobial additives with epoxy varnish. Experimental coatings for wood and steel panels were carried out on a laboratory scale. The fire retardant efficiency of both coating types was considered by using the limiting oxygen index (LOI) test. The flame retardants mechanical properties were also studied. LOI results showed that coating with a compound containing chlorine, nitrogen and phosphorus demonstrate a significant retardant effect when combined with epoxy varnish comparing with the blank sample, which not contain on the hexachlorocyclodiphosphazane derivative as an additive. The hexachlorocyclodiphosphazane derivative also exhibits mild results as a preservative against microbiological attack. The mechanical properties of the painted dry films were investigated according to ASTM.

1. INTRODUCTION

Epoxy coatings are widely used in the flooring markets and protective coatings due to their advanced mechanical properties chemical resistance and corrosion protection. Recently, epoxy coatings have developed from highly VOC structure to more environmentally benign structure such as solvent-free, high solids and waterborne coatings [1]. Polymeric materials are widely used in our daily lives. Instead of numerous advantages related to polymeric materials, there are serious shortcomings; for instance, poor fire resistance. Fire causes death due for 10 to 20 people per million in developed countries, and the number of injured persons is ten times higher [2]. Eventually, different approaches have been considered to improve the fire resistance of these materials. These include the use of naturally flame retardant polymers [3], an adaptation of the polymer backbone [4, 5], or merging of flame retardants into the polymers. As usually, flame retardant polymers can produce elevated production costs; the modification of existing systems is still respected by the industry. The polymer backbone modification by the inclusion of Si, P, N or B elements frequently offers good fire retardant properties to the newly modified polymer [6]. Epoxy resins usually tend to burn quickly and release high amounts of gas and smoke, and one of the conventional solutions is the inclusion of halogen-containing flame retardants [7]. Therefore, the epoxy resins enhancement towards fire retardant has been developed. It was previously reported that flame retardant additives containing nitrogen, such as cyclodiphosph(V) ozone and Schiff’s base monomer derivatives could be used [8-10]. Susceptible to microbial attack for epoxy resins when they are exposed to the atmosphere is another defect. Microorganisms cause blistering of coatings and disbonding under different service conditions [11-13]. Paint formulations containing biocide species are used to protect the coating surfaces from aquatic microorganism [14]. Epoxy resin integrated by hydrazone ligand derivative, o-hydroxycetophenone benzoylhydrazone (HBH) and its metal complexes, to establish enhancing in the antifouling and antimicrobial activity when physically incorporated. The physical and mechanical resistance was also studied to evaluate any associated drawbacks with the chemical addition [15]. FRs could be divided into five groups according to their chemical composition (a) halogen compounds; (b) metallic oxides and sulphides; (c) phosphorous compounds; (d) nitrogen compounds; and (e) inorganic hydrates, hydroxy compounds, boric compounds, silicates, carbonates [16-18].

Flame retardants could be included in polymeric materials as additives or as reactive materials. Widely used flame
retardants additive could be added into polymers, physically. This considered as the most economical and quick way for promoting flame retardant properties of commercial polymers. Flame retardant addition to polymer increased the material resistance to ignition, and to diminish the flame spread. So, rather than the flame rising itself extinguishes. The resulting products are still combustible, and the use of the flame retardant is minimized [19].

The literature survey confirmed that there had been significant interest in the phosphazene-based materials because they have a wide range of chemical and thermal stabilities, and can also improve flame-retardant and thermal properties to polymers and polymers composites [20-22]. Cyclotriphosphazene consists of alternating nitrogen and phosphorus atoms with substituent attached to the phosphorus atoms in a ring compound. It exhibits abnormal thermal properties such as self-extinguish ability and flame retardancy. The flame retardants reported in several papers have focused on these particular two hetero-atoms [23-27].

Hexachlorocyclodiphosphazane has several advantages as a flame retardant functional material. Firstly, the synthetic method can be developed for the preparation of hexachlorocyclodiphosphazane-based copolymers with various substituent groups. Secondly, non-flammable and thermal properties of the cyclotriphosphazene moieties could be awarded to the resultant polymers [28-31]. The phosphazene-based polymers are much efficient flame retardant, making them a new spotlight for study [32-33]. Potential purpose of some coumarin derivatives included thiazole as eco-friendly flame retardant, corrosion inhibitor, and antimicrobial additives for polyurethane coating are carried out. Little amount incorporation of the prepared coumarin derivatives within polyurethane coating improves the flame retardancy, corrosion resistance, antimicrobial activity, scratch hardness and gloss of the polyurethane-coated films [34]. Antibacterial coatings are rapidly emerging field of research. The three major approaches for designing antibacterial coatings are antibacterial agent release, anti-adhesion / bacteria-repelling and contact-killing [35]. Chitosan is now used in antimicrobial coatings as a rapidly growing research district [36]. In this work, a new additive flame retardant based on hexachlorocyclodiphosphazane of types (I-II) were physically added to the epoxy varnish as a biocide additive and flame retardant. LOI (limiting oxygen index) test was used to measure the flame retardant properties of the additives. Also, the paint tested according to gram-negative, gram-positive and fungal as antimicrobial additives. Physical and mechanical properties were also investigated to estimate any drawbacks related to the additives.

2. EXPERIMENTAL

2.1. Materials

All the chemicals used during the project were sourced either locally or from international companies.

2.2. Methods and Techniques

2.2.1. Synthesis of Hexachlorocyclodiphosphazane of Types (I-II)

The hexachlorocyclodiphosphazane was prepared by Chapman et al. [37], Kirsanov and Zhmurova, methods. And their complexes were prepared and characterized at the beginning of this century by Sharaby [38, 39]. A solution of aromatic amine (0.1 mol) such as para toluidine or P-Anisidine in dry benzene (100 ml) was added dropwise using dropping funnel to phosphorus pentachloride (20.9g, 0.1mol) in dry benzene (200 ml) at 15°C. The reaction mixture was heated under reflux for 3 h under anhydrous conditions with continuous stirring using magnetic stirrer and hot plate (the experiment was done in a well-ventilated area because benzene is cancer suspect agent). The reaction mixture was cooled to room temperature, and the formed solids were filtered on the pump, and then washed several times with dry benzene, and dry diethyl ether, hexachlorocyclodiphosphazane of types (I-II) were obtained as solid crystals. The reaction yields were determined as well as physicochemical characteristics such as colour, melting point and elemental analysis.

Scheme 1: Chemical structure of hexachlorocyclodiphosphazane derivatives type I-II.

Table 1: Epoxy Resin Composition Based on Hexachlorocyclodiphosphazane of Types (I-II)

| Component                                      | Wt. % |
|------------------------------------------------|-------|
| Epikote™ resin 1001-X-75 included aliphatic amine (base: hardener, 87 : 13 %) | 18.4  |
| Nonylphenol                                    | 2.9   |
| Aromatic hydrocarbon solvent                   | 3.9   |
| Mixtures of fine 65-75 micron                  |       |
| Talc                                           | 15.0  |
| Iron oxide                                     | 5.0   |
| CaCO₃                                          | 20.0  |
| BaSO₄                                          | 15.0  |
| Modified polyamide this agent                  | 2.0   |
| Methoxy propanol                               | 5.5   |
| Aromatic hydrocarbon solvent                   | 11.3  |
| Hexachlorodiphosphazane hexachlorocyclodiphosphazane additives | 0.5, 1.0 |

Viscosity: M-N (Gardner), colour: >18 (Gardner), solid content: 65±2%.
2.3. Coating Composition and Film Preparation

The coating compositions were prepared by means of incorporating hexachlorocyclodiphosph(V)azane of types (I-II), in the ratio of 0.5 and 1%, into the epoxy varnish. The composition of the varnish is tabulated in Table 1. The samples of different molar ratio were then applied to both steel and wood panels by using a brush. All efforts were made to maintain a uniform film thickness of 100 +/- 5µm for evaluating the physical and mechanical properties.

**Contact Angle Measurement**

Equipment for research performance hydrophobic contact angle includes: conductivity meter, pipette, lights, cameras, and computers as shown in Figure 1.

**Figure 1: Equipment for surface contact angle measurements.**

2.4. Flame Retardant Testing Method

The performance of epoxy varnish, with incorporated additives, was evaluated in a limited oxygen index (LOI) chamber. LOI values were determined by standardized tests such as ISO 4589-1: 1996 and ASTM D: 2863-97. Test panels were prepared using a combustible material (wood specimen). It was important that the panels were free of any surface contamination, or imperfections, before the coating application. Hand tool cleaning (sandpaper) was carefully used to treat the faces and edges of the panels. Final dry film thickness (DFT) was 100 +/- 5µm. In all cases, the film application was applied through brushing. Following 10 days of air-drying, the panels under study were heated at 50-60°C for two hours to eliminate any remaining solvent.

2.5. Physical and Mechanical Testing of Films

A variety of physical and mechanical evaluations of the painted films were carried out according to appropriate ASTM standard test methods. These included the preparation of the steel panels (ASTM Method D: 609-95), measurement of film thickness (ASTM Method D: 1005-95), and the degree of gloss for individual resin coatings (ASTM 523-08), film hardness by pencil test (ASTM Method D: 3363-92a), adhesion 'cross-hatch test' (ASTM Method D: 3359-95a), flexibility 'bend' test (ASTM Method D: 522-93a).

**Microbiology Assay**

Test method of antimicrobial activity carried out according to ASTM, D5589-97 (Re-approved 2002).

a. **Assay Medium for Antibacterial Activity (g/l)**

The nutrient agar medium was used for the antibacterial assay. The medium has the following composition:

Peptone: 5.0, beef extract 3.0, NaCl 5.0 and agar 20. The PH of the medium was adjusted at 6.8-7.0 before sterilization.

b. **Assay Medium for Antifungal Activity (g/l)**

The Dox medium used for antifungal activity. This medium composed of:

Sucrose 20.0, NaNO₃ 2.0, K₂HPO₄ 1.0, MgSO₄ 7H₂O 0.5, KCl 0.5, FeSO₄. 5H₂O 0.001 and Agar 20. The PH was adjusted at 6-6.4.

2.6. Organism Used

Bacteria employed in the microbiological assay were Micrococcus lutes NCTC 9341, Staphylococcus aureus NCTC 7447, E.coli Bppol, Salmonella. While fungi were Candida albicans IMRU 366g, Aspergillus flower, Penicillium citrinum and Suserium.

2.7. Preparation of Tested Sample

The tested varnish was brushed on each side of a Whatman filter paper (No. 30) and allowed to dry for 24 h. Squares of the coated filter paper of (1.25 in) were made. The square was sterilized by dipping in alcohol and placed centrally on the agar surface in the Petri dishes using a sterile for sepsis.

2.8. Antimicrobial Activity

(a) **Method Used for Bacteria Under Investigation**

Twenty-four hours old culture of each of the test bacteria and forty-eight hours old culture of each of the test yeasts were used. 5 ml of sterile distilled water was added to the culture tube and mixed well by a vortex mixer. Five drops of the suspension were used to inoculate 100ml. Nutrient agar medium (for test bacteria) or 100ml yeast extract-malt extract medium (for test yeasts) at 45°C. This was dispensed among Petri dishes in 20ml portions. A coated filter paper discs (13mm) were aseptically put on the surface of the seeded plates with the different test organisms. The plates were left for 2 hrs in a refrigerator for diffusion after which the plates were incubated at 300°C for 18 hrs (for bacteria) or 48 hrs (for yeasts). The detection of a clear zone around the paper disc is an indication of the antagonistic properties of the coated filter paper disc and liquid varnishes under study. For each test organism, at least four discs were used for different concentration of hexachlorocyclodiphosphazane of types (I-II), 0, 0.5, and 1.00 % of the varnish under investigation.
(b) Method used for Fungi under Investigation

The spores and mycelia of each of the test fungi (48 hrs old culture) were streaked on the surface of the plates of Dox medium after pouring and solidification. The method was preceded as previously described under the item. The antimicrobial activity of the coated filter paper disc against a variety of microorganisms, including Gram-Positive, Gram-Negative, Yeasts and fungi was investigated.

3. RESULTS AND DISCUSSION

The uses of hexachlorocyclodiphosphazane of types of (I-II) as flame retardant additives in epoxy varnish were studied. Phosphazene is known to impart flame retardant characteristics and thermal stability to the polymer structure. The strength of the P-N bonds provides the majority of phosphorus-nitrogen compounds remarkable thermal stability [40].

3.1. Synthesis of Hexachlorocyclodiphosphazane of Types (I-II)

In this study, hexachlorocyclodiphosphazane of types (I-II) were prepared to demonstrate its superior flame retardant properties. Scheme 1, 2 shows the chemical structures of the prepared hexachlorocyclodiphosphazane types (I-II).

Elemental analysis, physical properties such as melting point and product colour and reaction yield, were measured and scheduled in Table 2. The high contrast between the theoretical and experimental values of the C, H, P, S and N levels demonstrates the successful methods of synthesis and purification of the products.

3.2. Evaluation of Film Properties

3.2.1. Evaluation of Hexachlorocyclodiphosphazane of Types (I-II) as Flame Retardant Additives Integrated into Epoxy Paint

Flame retardant properties of the Epoxy paint mixed with hexachlorocyclodiphosphazane of types (I-II) were estimated through the limiting oxygen index (LOI) test. The LOI is defined as the minimum concentration of oxygen, expressed as a percentage that will support combustion of a polymer. It is measured by passing a mixture of nitrogen and oxygen over a burning sample and reducing the level of oxygen until reached to a critical level [41]. A high concentration of oxygen requirement indicates improved flame retardancy of the sample. This technique is suitable as a semi-qualitative indicator of the efficiency of the flame retardant during the development phase of the work and the research. This is as a result of the fact that the size of the test sample required is...
**Table 2: Physico – Chemical Properties of Hexachlorocyclodiposphazane of Types (I-II)**

| Characteristics | Compound I     | Compound II    |
|-----------------|---------------|---------------|
| Melting point (°C) | 194-200       | 199-201       |
| Yield (%)       | 73            | 80.5          |
| Colour          | Gray          | Gray          |

**Elemental analysis**

|   | Calc. | found |
|---|-------|-------|
| C % | 34.88 | 32.53 |
| H % | 2.91  | 2.73  |
| N % | 5.98  | 5.42  |
| Cl % | 43.86 | 41.15 |
| P % | 12.77 | 11.98 |

small and the equipment is relatively inexpensive. Hexachlorocyclodiposphazane, besides its polymeric products are of best-known and the oldest class of nitrogen-phosphor compounds. Recently, as has previously been revealed, there has been renewed interest in the family of phosphazene-based of materials not only because of chemical stabilities and their wide range of thermal but also due to the potential for the improvement of the flame retardant properties of polymers and their composites. The incorporation of hexachlorocyclodiposphazane of types (I-II), physically added into epoxy paint in the ratios revealed in the experimental section, results in excellent flame retardancy when compared alongside a blank epoxy specimen. The results found from the LOI test are shown in Table 3 and Figures 3-5. It observed that the value of LOI of epoxy varnish without an additive is 25, but the maximum LOI with additive is 92. The air that we breathe in (Normal atmospheric air) is about 21% oxygen so that material with an LOI less than 21% would burn without difficulty in air. In comparison, a material with an LOI value is higher than 21%, but less than 28% would be considered as ‘slow-burning’. Nevertheless, a self-extinguishing material is one that would stopover burning after the removal of the ignition or fire source. The increased quantity of hexachlorocyclodiposphazane of types (I-II) additives increases the LOI value of the epoxy paint formulation. This could be as a result of the incorporation of the hexachlorocyclodiposphazane into the network of the thermoset epoxy paint, which increases the polymer flame retardancy. This could be due to the flame retardant synergy of the nitrogen and phosphorus content. This also is because of the fact that they are high molecular weight compounds containing phosphorus, nitrogen and chlorine and therefore provide superior properties of flame retardant compared to flame retardants, which have a low molecular weight. The additional exciting point is that the LOI of compound II is the highest more than that coated films based on the compound I, all the obtained results from wood and steel panels were represented as in Figures 3-5, and the Limiting oxygen index (LOI) chamber also represented in Figure 6.

**Mechanism of the Attractive Properties (Flame Retardant) of the Proposed Paints**

The coating system consists of a base coating layer, top coating layer and foam layer. When the material exposed to fire, it starts to foam after the atmospheric temperature reaches 250 to 300°C. The foam swells to a volume 25 to 50 times higher to get superior heat insulation. The Mechanism of the attractive properties (flame Retardant) of the proposed paints was represented in Figure 2.

**Antimicrobial Activity of Epoxy Paint based on Hexachlorocyclodiposphazane Derivatives**

Biocide additives are generally used to extend the life of surface coatings. They avoid or slow down, the growth of organisms on the surface of the coating. Antibacterial agents can be either constitute an integral part of the coating or trapped among layers, hexachlorocyclodiposphazane have a good potential application as an antimicrobial [42, 43]. So, based on the obtained results, which tabulated in Table 6, the antimicrobial activities of the prepared hexachlorocyclodiposphazane derivatives were evaluated against One Gram-negative strain (Pseudomonas aeruginosa), One Gram-positive bacteria strain (Bacillus subtilis) and Two fungi strains (Aspergillus baraseliensis, and Candida alibicans). A disc-diffusion method was used to confirm the activity. The results from the anti-microbial activity are shown in Table 4 and Figure 7. The prepared hexachlorocyclodiposphazane derivatives type I, II exhibited a marginally better effect on Two fungi strains (Aspergillus baraseliensis, and Candida alibicans) and mild effect against Gram-positive (Bacillus...
Table 3: Flame Retardant Characteristics of Epoxy Paint Incorporated Hexachlorocyclodiphosphazaneas Flame Retardant Additives

| Formulation          | FR % | Limiting Oxygen Index (LOI) |
|----------------------|------|-----------------------------|
| Blank Epoxy          | -    | 25                          |
| Epoxy and Comp. I    | 0.5  | 75                          |
|                      | 1.0  | 84                          |
| Epoxy & Comp. II     | 0.5  | 81                          |
|                      | 1.0  | 92                          |

The design approaches to control the release of antibacterial agents over space and time could be grouped under three major categories. (A) Active approaches. Outside stimuli could be used to trigger the local release of embedded compounds. (B) Bacterial trigger approaches. Bacteria-responsive coatings release antibacterial agents locally when challenged with bacteria. (C) Passive approaches. By tuning the properties of the coating, it is probable to impose specific preloaded release kinetics, giving the possibility to produce a variety of release profiles, including linear release (right) or rapid bursts (left) from antibacterial (AB) coatings [44].

3.2.2. Evaluation of the Physical and Mechanical Properties of Epoxy Paint Combined Hexachlorocyclodiphosphazane Derivatives

The effects of adding flame retardant to the epoxy paint, relating to the mechanical and physical properties, were

Figure 2: Represented the Mechanism of the attractive properties (flame Retardant) of the proposed paints.

Figure 3: Some of the coated wood specimens before ignition.
New Flame Retardant and Antimicrobial Paints Based on Epoxy Paint Incorporated

Figure 4: Some of the coated wood specimens after ignition.

Figure 5: Some of the coated steel specimens after ignition.

Figure 6: Limiting oxygen index (LOI) chamber.

Table 4: Antimicrobial Activity of an Epoxy Paint Incorporated Hexachlorocyclodiphosphazane Type (I-II) as a Biocide Additives

| Microorganisms         | Blank Sample | Hexachlorocyclodiphosphazane type I, 0.5-1.0 % | Hexachlorocyclodiphosphazane type II, 0.5-1.0 % |
|------------------------|--------------|-----------------------------------------------|-----------------------------------------------|
|                        |              | 0.5   | 1.0   | 0.5   | 1.0   |
| Bacillus subtilis      | -ve          | +ve   | +ve   | +ve   | ++ve  |
| Pseudomonas aeruginosa | -ve          | -ve   | +ve   | +ve   | ++ve  |
| Aspergillus barasellensis | -ve      | +ve   | +ve   | +ve   | +++ve |
| Candida albicans       | -ve          | +ve   | +ve   | +ve   | +++ve |

Where, Inactive = -Ve,
Mildly active: Inhibition values = 0.1–0.6 cm beyond control = +,
Moderately active: Inhibition values = 0.65–1.0 cm beyond control = ++.
Highly active: Inhibition values = 1.1–1.5 cm beyond control = +++.
Very highly active: Inhibition values = 1.6–2.00 cm beyond control = ++++.
estimated as per the standard test methods. It was done to ascertain any negative aspects, which might arise owing to the presence of the additives. The scratch hardness, gloss, flexibility and adhesion were all measured. The resulting data is shown in Table 5.

Interface between Paints and Wood

The interface among wood and adhesives are much more complicated than the interface between painted or lacquered metals or polymers due to the porosity of wood which gives a broad, unavoidable roughness to the surface. It is clear that a lacquer, glue or paint could have sufficiently low viscosity to penetrate even small cracks and fill up the surface roughness. The epoxy and wood must be developed and compatible with some kind of bonding forces to reach a good bond. Also, the viscosity of the adherent must be sufficiently low to allow the polymer at a microscopic scale chains to fill the wood’s surface features. So based on the obtained results of the mechanical properties for the dry coated films we can say that because of the high computability between the epoxy paint and the high penetration of the paint in the wood proses the adhesion strength, hardness and gloss were good results. This also investigated by using SEM micrograph of wood painted with epoxy paint containing the hexachlorocyclodiphosphazane derivatives as in Figure 9, 10. It can be observed that the wood, steel sample and epoxy paint containing the hexachlorocyclodiphosphazane derivatives are seen to be compatible. The electron micrographs also indicate that the epoxy paint is containing the hexachlorocyclodiphosphazane derivatives penetrated into the surface of the wood and steel panels and also indicating that the mixing process of paint formulation was successful and high crosslinking.

(a) Gloss

It was measured using a Sheen UK gloss meter. Using a 60° angle on observing the films, it could be seen that the hexachlorocyclodiphosphazane additive essentially increased the gloss levels. This is a positive result that could be attributed to the introduction of aromatic rings, present in the additive structure this is exhibited on the steel panels and represented in Figure 11.

(b) Scratch Hardness Test

It was measured by using a Sheen UK hardness tester. The scratch hardness is observed to vary among 1500 – 2000g and from the data it is clear that as we increase the hexachlorocyclodiphosphazane additive concentration from 0.5 to 1.00%, the scratch hardness of the film increases, this test on steel panels was represented in Figure 12.

(c) Cross-Hatch Adhesion Test

It was determined by using a Sheen UK crosscut adhesion tester. For this test method, a lattice with six cuts in each direction was made in the film (the cuts were spaced at 2
mm). The pressure-sensitive tape was then applied over the lattice and then removed. All the coating film demonstrated good cross-hatch adhesion. The hexachlorocyclodiphosphazane additives did not change the adhesion properties of the epoxy paint due to the properties of the very good mechanical properties of the epoxy paints, so this test on coated steel panels was represented in Figure 13.

(d) Flexibility (Bend) Test

Flexibility was measured by using a ¼ inch Mandrel bend tester from Sheen UK, in such a way that the surface of the panel was directed outside. For all the coating compositions the films passed the ¾ inch Mandrel bend test. The varnish was considered acceptable if no cracking marks, or dislodging, are detected following the bending procedure. It can be stated that all the films exhibited reasonably very good flexibility, based on this qualitative measurement.

Properties (hydro and oil-philic or phobic) of epoxy paints with hexachlorocyclodiphosphazane derivatives additives

Contact angle measurements aim to determine the surface properties of materials, absorb water (hydrophilic) or water-
repellent (hydrophobic). Hydrophobicity of the surface of the test material is influenced by the nature of the material and the composition of the filler component. By measuring the contact angle among distilled water dripped on the test materials on the material surface, the hydrophobic properties can be determined. Surface contact angle value of the droplets of liquid material obtained through the digital camera scene by direct observation, which is then stored on the computer [45]. The photographs processed by software to obtain the contact angle on the left and the right side of the test specimen is measured. The contact angle of epoxy resin material increased after the addition of pigment, filler material and the hexachlorocylodiphosphazane derivatives type I-II. Surface hydrophobicity properties of the epoxy resin material increases hexachlorocylodiphosphazane derivatives type I-II per cent. This happens because there is on the hydrophobic epoxy more a methyl group (CH$_3$). Methyl group is derived from the hexachlorocylodiphosphazane derivatives type I-II. The contact angle results in measurements before the aging, demonstration that all measurements of the contact angle of the tested sample have reached the contact angle 94°, and meaning that the tested sample is normally hydrophobic.

**CONCLUSION**

The coated films by epoxy varnish without hexachlorocyclo-diphosphazane derivatives type I-II (blanket sample) showed some fire retardant effect but not any antimicrobial effect; however, they did not show inhibitory zones towards the microorganisms. However antimicrobial and antifungal activity properties recorded for coated films by epoxy incorporated with 0.5 and 1.00% hexachlorocyclophosphazane derivatives type I-II may be due to Cl, P, and a lone pair of electrons on N atoms in the structure of the dimer. The best condition that offered the most potent inhibitory activity of coated film was the coated film with 1.0% of hexachlorocyclophosphazane derivatives type I-II against the Gram-positive, Gram-negative bacteria and fungi. The p-anisidine dimer (compound II) gave the most robust inhibitory activity at 250-300mg/ml against all the tested microorganisms more than p-toluidine dimer (compound I). From the obtained results, it can be observed that the maximum LOI with additive compounds is 92 and the LOI value of Epoxy varnish without an additive compound is 18. The air that we breathe in (Normal atmospheric air) is almost 21% oxygen, so a material with an LOI of less than 21% can be burn easily in air.

In comparison, a material with an LOI value is higher than 21%, but less than 28% would be considered as ‘slow-burning’. Nevertheless, a self-extinguishing material is one that would stopover burning after the removal of the ignition or fire source. The increased quantity of hexachlorocyclophosphazane of types (I-II) additives increases the LOI value of the epoxy paint formulation. This could be as a result of the incorporation of the hexachlorocyclophosphazane into the network of the thermoset epoxy paint, which increases the polymer flame retardancy. This could be due to the flame retardant synergy of the nitrogen and phosphorus content. This also is because they are high molecular weight compounds containing phosphorus, nitrogen and chlorine and therefore provide...
superior properties of flame retardant compared to flame retardants, which have a low molecular weight. The additional interesting point is that the LOI of compound II is the highest more than that coated films based on compound I.

Also, the antimicrobial activity of the epoxy paint based on hexachlorodiphenylphosphazene derivatives against the target micro-organisms increases with the increase in the hexachlorodiphenylphosphazene derivatives addition level — also overall, the obtained results of suggesting that the multifunctional epoxy coating shows very good mechanical stability.

So based on the obtained results in the work and the literature survey the hexachlorodiphenylphosphazene of types additives are very good multifunctional additives for industrial applications like paints additives and used as the advanced packing and packaging materials.

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