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RESEARCH PAPER

Modeling the Kinetics of Degradation of Epoxy Nanocomposites in the Presence of Modified Nanodiamonds with Carboxyl

Sabah Auda AbdulAmeer [1]*, Russul Thabit [2], Mohammed S. Hadi [3], Israa Taha Ibrahim [4], Shakir Mahmood Saeed [5], Athmar Ali Kadhim [6], Ahmed Abbas Sahib [7]

[1] Department of Medical Laboratory Technics, Ahl Al Bayt University, Kerbala, Iraq
[2] Medical technical college, Al-Farahidi University, Iraq
[3] Medical Physics Department, Al-Mustaqbal University College, Babylon, Iraq
[4] Department of Medical Laboratory Technics, AL-Nisour University College, Baghdad, Iraq
[5] Department of Pharmacy, Al-Noor University College, Nineveh, Iraq
[6] Medical Laboratories Techniques, Hilla University College Babylon, Iraq
[7] Mazaya University College, Iraq

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ABSTRACT

Due to their suitable thermal, electrical, and chemical properties, epoxy composites are constantly employed in the multi-structure, construction, electronics, adhesive, and coating industries. The distribution and dispersion of nanoparticles in the polymer matrix are essential and influential factors in epoxy nanocomposites' mechanical properties. Utilizing baking kinetic equations to evaluate and optimize the manufacturing process. The widespread adoption of destruction kinetics modeling by engineers has decreased product costs and improved product quality. The curing reaction mechanism is unaffected by adding nanodiamonds modified with carboxyl groups to pure epoxy resin. Nanodiamond surface modification increases the curing reaction speed of epoxy nanomultistructure. The results of this study demonstrated that the presence of modified nanodiamond could increase the thermal stability of an epoxy nanocomposite hybrid sample. According to the findings, the activation energy of nano-multi structures modified with 0.2, 0.4, and 0.6% nanodiamond by weight decreased by 11.58, 18.95, and 22.96%, respectively. In nano-multi structures modified with 0.2%, 0.4%, and 0.6% nanodiamond by weight, the wear rate decreased by 83.14, 93.10, and 94.14%, respectively; the friction coefficient decreased by 46.03, 52.08, and 54.34%, respectively. The current study examined the influence of nanodiamond addition on morphology, mechanical and rheological properties, and modeling of baking kinetics, degradation kinetics, and tribological properties.

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* *Corresponding Author Email: sabahauda79@gmail.com*

INTRODUCTION

Today, simultaneous integration of features is required in many engineering and everyday applications, and it is impossible to use a single type of material to satisfy all the desired features [1]. For example, lightweight materials with high strength are required in many applications [2]. Consequently, the combination of various materials into composite products is inevitable. Consequently, a composite combines two or more components with distinct physical and chemical properties [3-5].

Polymer nanocomposites are particularly interested in modern research for high-performance applications due to their excellent mechanical properties, including high bending strength and stiffness, resistance to erosion and corrosion, perforation resistance, and their simple construction [4, 5]. Some common and specialized applications for these composites are parts of aerospace equipment, automobiles, vehicles, bulletproof vests, and armor [6]. Polymer nanocomposites have inherent disadvantages such as layering, cracking, and low fatigue. They lack specific performance characteristics required by spacecraft and missiles, including high electrical and thermal resistance for electrostatic destruction and lightning strike protection [7]. The application of nanomaterials and fibers dispersed in polymer matrices has garnered considerable attention. Utilizing different nanoparticles in polymer nanocomposite with an epoxy base has been shown in various studies to increase stiffness and tensile strength [8-12].

Epoxy resins are thermosetting polymers. Because of their unique mechanical, chemical, and physical properties, they are used in various applications, such as composites, adhesives, paints, and flooring [13]. Curing agents or epoxy nanocomposite hardeners play a significant role in how it is used and what properties it has. This means that the materials mentioned affecting their different properties [14]. These properties include the speed of the curing reaction and how much heat it makes, the viscosity, the time it takes for the nanocomposite system to gel, the amount of heat needed for curing, the way the nanocomposite is applied and how long it will last, the type of chemical bonding, the degree of crosslinking, and the final properties of the cured system, which include chemical and thermal resistance, physical, mechanical, and electrical properties [15-17]. In general, curing agents of epoxy resins are categorized according to their chemical structure into three primary groups [18]:

1. Compounds containing active hydrogens, such as amines of the first and second types, amides, hydroxyls, acids, and anhydrides
2. Catalysts (anionic and cationic initiators) such as Lewis acids, tertiary amines, and inorganic halides
3. Reactive crosslinkers, including melamine-formaldehyde, phenol-formaldehyde, and urea-formaldehyde.

Depending on the curing agent, epoxy resin systems have different service lives or durations of use. The curing reaction begins at normal temperatures when a conventional curing agent is combined with an epoxy resin, even though the mixture must be heated to cure fully [19]. Consequently, the mixture’s viscosity increases with time at room temperature, resulting in a substantial amount of industrial waste. Two systems consisting of two-component and one-component materials are used to repair this flaw [20]. Most epoxy resins are sold in the form of two components, resin and curing agent, and the resin and curing agent are mixed in a specific proportion before use [21]. However, two-component materials have drawbacks that hinder the performance of the resin. The formation of bubbles during mixing, the need to strictly observe the mixing ratio, the reduction of physical and mechanical properties, and incomplete mixing due to the adhesiveness of the resin are some of the disadvantages. The resin is mixed with a delay curing agent and sold as a single component in single-component materials [22].

One-component epoxy materials contain both epoxy resin and curing agents. However, at normal temperatures, the curing reaction takes a long time to initiate or does not occur at all. Resin curing commences when these curing agents are activated by external stimuli such as heat, light, pressure, magnetic field, ultrasound waves, electron radiation, or X-ray [23]. Heat and light radiation are the most essential and practical external stimuli due to their low cost and a high degree of safety [24]. Delay-thermal and delay-optical curing agents have been developed extensively. Light is the most convenient external stimulus, but heat induces a more uniform curing response [25]. It is complicated to achieve a homogeneous curing reaction with light irradiation because the homogeneity of the reaction depends on the radiation source and the reaction vessel.

On the other hand, the absorption and scattering of light in the environment prevent effective irradiation and make it difficult to achieve uniform irradiation [26].

In recent years, considerable research has been conducted to improve the mechanical properties of epoxy resin, particularly its tensile modulus. Nanoparticles were explicitly utilized to achieve this result [27]. Compared to microparticles, nanoparticles in polymer matrices provide superior matrix impregnation due to their small size; surface tension concentration is minimal. In addition, nanoparticle size significantly impacts the composite's final properties [28].

Researchers in various polymer fields are interested in using nanomaterials to prepare and construct polymer nanostructures to improve the properties of polymers in various applications [29]. In recent years, there has been much interest in nanomaterials that improve the thermal and wear mechanical properties of various resins, such as nanodiamonds. However, compared to other nanomaterials such as nanoclay, carbon nanotubes, nano-silica, and nanographene, there are relatively few works and studies in its field [30]. There have been significant developments in the production of synthetic diamonds in recent years, resulting in nanometer-sized diamonds. Nanodiamond is utilized in numerous industries due to its exceptional mechanical, thermal, and optical properties. Nanodiamond has been used in the rubber industry, cutting tools, and semiconductors [31]. The effect of carboxyl-modified nanodiamonds on the properties of epoxy resin was investigated in the present study.

Due to the numerous benefits and applications of epoxy resin and the possibility of enhancing its properties through nanomaterials, it is necessary to conduct research in this area. Due to the unique characteristics of nanodiamonds and the limited number of studies on them, it has become crucial in recent years to investigate the impact of nanodiamonds on the properties of epoxy resin. The utilization of carboxyl-modified nanodiamonds is the novel aspect of the present study, which aims to examine the thermal behavior and thermal stability of epoxy nanostructures.

MATERIALS AND METHODS

Materials

The current research utilized modified nanodiamonds with an average diameter of 4-6 nm and a purity of 97% and epoxy with an average molecular weight of 182 to 192 grams. To create epoxy nanocomposites, the required amount of epoxy resin was added to an acetone solution that had been pre-sonicated for 15 minutes. While the system was mixing, the solution was stirred for 30 minutes at ambient temperature, followed by 60 minutes at 100°C at a speed of 2,000 rpm to evaporate the acetone completely. The combination was then placed under a vacuum to guarantee that all traces of acetone were removed. To prepare the samples to evaluate their dynamic-mechanical properties, they were first degassed for 30 minutes in a vacuum at 45°C, followed by 24 hours of curing at ambient temperature. After baking, samples were maintained at 100°C for one hour.

Epoxy nanocomposites production method

To make epoxy nanocomposites, 60 g of ethanol was mixed with 100 g of epoxy colloid. The mixture was then stirred for 10 minutes at 80 °C to make the epoxy less thick. The needed amount of filler was then mixed with ethanol for 30 minutes, after which it was mixed for 1 hour at 80°C with the epoxy-ethanol solution that had already been made. After cooling, a two-phase mixture of liquids and liquids was made in the freezer. The top layer was a low ethanol-epoxy solution that could be thrown away or evaporated. The bottom layer was the filler epoxy solution. The mixture was then milled with balls to break up the clumps and spread the filler even more. Keep in mind that the speed of the ball mill should not be more than 250 rpm so that sheets and parts do not come apart because of high shear stress. The mixture was purged of air under a vacuum at 80 °C for 24 hours to get rid of the solvent. Under mechanical mixing (280 rpm for 15 minutes) of the mixture, the hardener was added at a weight of 25% by weight. The mixture was then degassed at 65°C for 30 minutes. The epoxy nanocomposites were finally made after being cured for 1 hour at 110°C, 2 hours at 140°C, and 1 hour at 170°C.

Characterization

Thermogravimetric analysis was used to investigate thermal stability at a heating rate of 10 °C in N_2 atmosphere. At room temperature, X-ray diffraction was performed with a copper target at 40 kV, and 30 mA. Using Fourier transform infrared spectroscopy, infrared spectra were gathered. X-ray photoelectron spectroscopy was conducted using Mg-Ka X-ray irradiation as the excitation source for X-rays to confirm the surface chemistry of the epoxy nanocomposites.

RESULTS AND DISCUSSION

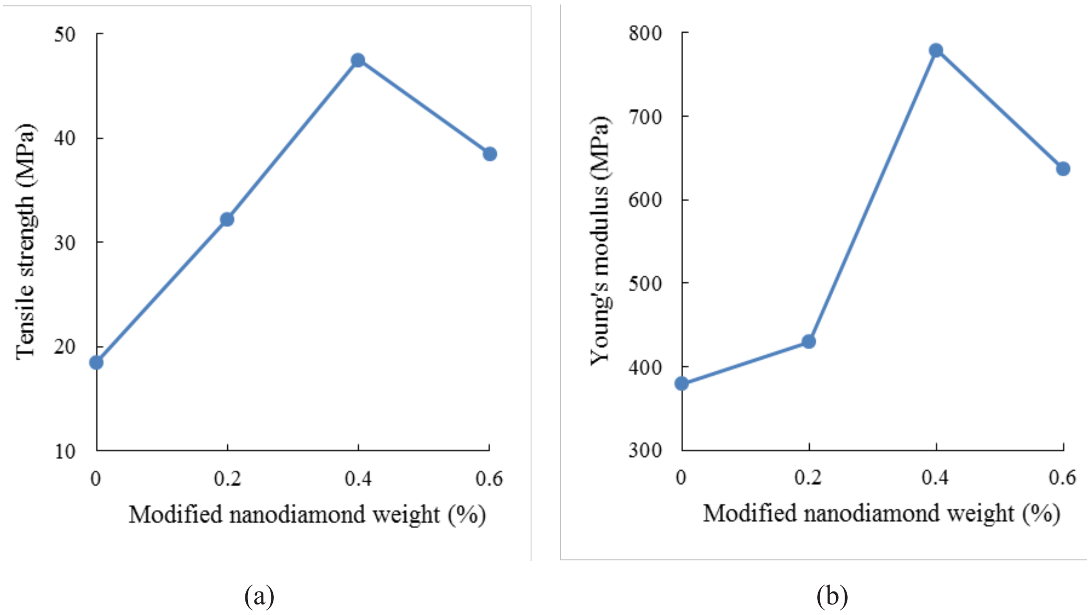
Mechanical properties

The investigation of the effect of adding various percentages by weight of modified nanodiamonds on the bending strength of epoxy resin revealed that by adding these amounts, the mechanical properties increase. In contrast, by adding 0.6% by weight, it decreases. Examining the impact strength of epoxy resin and epoxy nanocomposites in the presence of 0.2, 0.4, and 0.6% by weight of diamond nanoparticles revealed that adding 0.4% by weight increases the impact strength; however, adding 0.6% by weight decreases the impact strength of the epoxy nanostructure due to an increase in lumpiness.

The effect of 0.2 to 0.6% nanodiamond by weight on the mechanical properties of epoxy resin was investigated, and the results are presented in Fig. 1. Due to the clumping effect of nanoplates in the matrix of epoxy resin, the tensile strength increases by adding 0.2 and 0.4% by weight of the nanodiamond to the epoxy resin and decreases by adding 0.6% by weight of the nanodiamond. Also, the value of Young's modulus of pure epoxy resin is 371 MPa, and by adding 0.4% by weight of nanodiamond to it, the value increases to 768 MPa, indicating that the epoxy nano-multi structure in the presence of 0.4% by weight of nanodiamond is stronger than the sample epoxy nano-multi structure. The dimensional stability of pure epoxy resin has improved, and Young's modulus has increased by more than twofold.

Curing kinetics

Examining the heat flow of epoxy resin and epoxy nano-multi structure in the presence of modified nanodiamond revealed that the modified sample has a higher maximum thermal temperature than pure epoxy resin. This is due to the activity of the carboxyl group and the improved dispersion of the modified nanodiamond in the epoxy resin. Due to the modification of the nanodiamond surface with a carboxyl group and amine group on the heat flow of epoxy resin, researchers have stated in multiple studies that adding this type of modified nanomaterial to pure epoxy resin does not alter the curing reaction mechanism [32, 33]. In addition, the heat flow has a peak, and modifying the nanodiamond's surface accelerates the curing reaction of nanomultistructure epoxy [34-35]. Fig. 2 illustrates variations in the activation energy of pure epoxy resin and epoxy nanocomposite in the presence of 0.2, 0.4, and 0.6% by weight of modified nanodiamond. The results demonstrated that adding modified



(a) (b)

Fig. 1. Mechanical properties of epoxy resin and epoxy nanocomposites: a) Tensile strength, b) Young's modulus

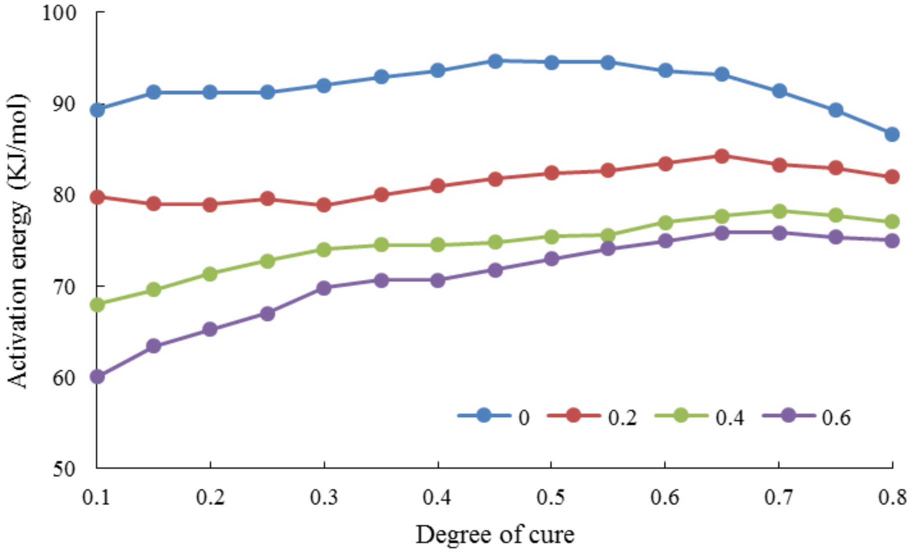


Fig. 2. The activation energy of epoxy resin and epoxy nano-multi structure

nanodiamonds to pure epoxy resin increases the activation energy of nanomultistructure epoxy relative to the pure epoxy resin while modifying the surface of modified nanodiamonds with a carboxyl group decreases the activation energy of nanomultistructure epoxy.

Modeling the curing kinetics of epoxy resins in the presence of modified nanodiamonds modified with a carboxyl group revealed that the laboratory data agree with the Sestak-Berggren model and that the order of the reaction is close to 2. The degree of curing of epoxy nano-multi structures in the presence of modified nanodiamonds revealed that all samples exhibit autocatalytic behavior, and the graph of the degree of curing is a sigmoid function. In addition, the results demonstrated that the amount of activation energy decreases with the addition of modified nanodiamonds and that these nanomaterials play a catalytic role in the curing reaction of epoxy resin.

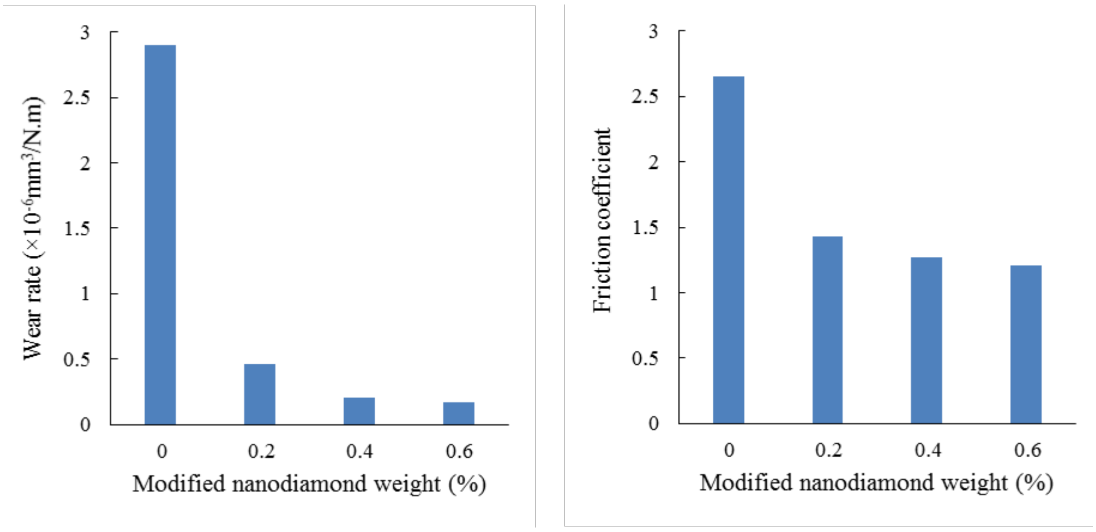
Tribological properties

The effect of nanodiamonds modified with a carboxyl group at weight percentages of 0.2, 0.4, and 0.6 on the tribological properties of epoxy resin was also studied. The results demonstrated that the tribological characteristics (friction coefficient and wear rate) increase due to the increase in hardness caused by the presence of modified nanodiamonds in the epoxy nano-composites and at the wear area. The mentioned results are shown in Fig. 3.

Thermal stability and degradation kinetics

Thermogravimetric analysis was used to investigate the degradation kinetics of pure epoxy resin and nano multi-structure epoxy resin samples in a nitrogen environment at ambient temperatures up to 800°C. The results demonstrated that the presence of modified nanodiamonds improved thermal stability. The addition of nanodiamond to epoxy resin acts as a retardant, an insulator, and a thermal degrading reaction rate reducer. The percentage of weight reduction of pure epoxy resin and nano-multi structure epoxy resin in the presence of 0.2, 0.4, and 0.6 weight percent of nanodiamond modified by annealing heat treatment method is examined in Table 1.

Due to the heat treatment method targeting impurities, adding modified nanodiamonds to the epoxy resin matrix improved the thermal stability compared to pure epoxy resin. In addition, the addition of 0.6% by weight of the modified nanodiamond does not significantly alter the weight loss percentage due to the nanodiamond's improper distribution in the epoxy resin matrix. Compared to other samples, the epoxy nanostructure containing 0.4% by weight of nanodiamond has the highest thermal stability. Comparing the samples revealed that the degradation of epoxy nanostructures



(a) (b)

Fig. 3. Tribological properties of epoxy resin and epoxy nano-composites: a) Wear rate, b) Friction coefficient

Table 1. Weight loss percentage of epoxy resin and nano-multi structure epoxy

Sample	Weight loss percentage at different temperatures (˚C)		
	25	50	75
Epoxy resin	368	392	416
0.2 weight percent of modified nanodiamond	375	398	422
0.4 weight percent of modified nanodiamond	383	404	436
0.6 weight percent of modified nanodiamond	378	401	428

requires a great deal of energy, that the rate of the degradation reaction decreases, and that the thermal stability increases.

According to the few studies conducted in the field of diamond nanoparticles, diamond nanoparticles improve the mechanical, thermal, electrical, and abrasion resistance and, in some instances, the antibacterial effect of resins, thereby enhancing these properties [7, 32]. Due to this enhancement of properties, diamond nanoparticles in resins are rising. In the coming years, extensive research should be planned and conducted in the field of nanodiamond superstructures, their properties, and their characteristics. Due to their unique properties, nanodiamond has the potential to be utilized in a variety of applications. Also, with the expansion of new synthesis methods and the decrease in production costs, their use in various industries, including the preparation of Bespar nanostructures and Bespars, will increase gradually.

CONCLUSION

The study of the tensile strength of epoxy resin and epoxy nano-multi structure hybrid in the presence of modified nanodiamonds revealed that the presence of modified nanodiamonds improves the distribution and interaction between the surfaces of nanomaterials in comparison to pure epoxy resin. Examining the degree of curing of epoxy nanostructures in the presence of nanodiamonds modified with a carboxyl group revealed that all samples exhibit autocatalytic behavior and that these nanomaterials play a catalytic role in the curing of epoxy resin. The investigation of the degradation kinetics of epoxy nanocomposites revealed that the addition of nanodiamonds to epoxy resin acts as a retardant and slows the thermal degradation reaction. Due to the issue of thermal stability and the lack of articles in the field of modeling the kinetics of thermal degradation and identifying the mechanisms of thermal degradation, this research requires additional research and study to determine the most effective model for modeling the kinetics of thermal degradation. Also presented is the examination of laboratory data using the model.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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