Synthesis of CuO/Epoxy nanocomposites for the preparation of antifungal coating

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ABSTRACT
Objective(s): Antibacterial and antifungal nanocomposites are widely used in food packaging and pharmaceutical and medicine industries. Among the polymers of these nanocomposites, epoxy coatings are commonly used for health and industrial applications. The present study aimed to synthesize CuO nanoparticles using the chemical reduction method and characterized them by ultraviolet-visible (UV-Vis) spectroscopy and dynamic light scattering (DLS) analysis.

Materials and Methods: The nanoparticles were synthesized with the mean size of 45 nanometers. Following that, the CuO/epoxy nanocomposite were prepared in three concentrations of 1%, 3%, and 5% of the CuO nanoparticles. The results of X-ray diffractometry (XRD) and scanning electron microscopy (SEM) confirmed the presence of nanoparticles on the nanocomposite surface. In addition, the disc-diffusion method was used to assess the antifungal properties of the nanocomposites.

Results: The results of XRD and SEM confirmed the presence of CuO nanoparticles on the nanocomposite surface. The optimal nanocomposite concentration for the maximum antifungal activity was 3%.

Conclusion: It seems that the CuO nanoparticles could be used to provide antifungal nanocomposites, which are applicable in medicine and food industries.

Keywords: Antifungal, Coating, CuO/Epoxy Nanocomposites

INTRODUCTION
Today, the production of antibacterial and antifungal materials is of paramount importance. These materials are used in major industries, such as the food and pharmaceutical industries. Polymers are considered to be more important compared to other antibacterial and antifungal products owing to their numerous applications. Various polymers constitute approximately 70% of the materials found in the environment, and it is essential to modify the polymers used in the generation of antibacterial and antifungal effects. In addition, the production of antibacterial and antifungal nanocomposites should be taken into account due to their numerous applications in the pharmaceutical, food, packaging, and medicine industries. Epoxy coating is a popular protective coating, which is widely used for industrial applications. Therefore, researchers have attempted to overcome the problems associated with epoxy resin through the addition of various nanomaterials. Recently, antimicrobial and antifungal epoxy coatings have attracted the attention of researchers for surface protection. As such, it is essential to develop epoxy coatings with antimicrobial and antifungal properties. According to Sadeghnejad et al. [1], the antibacterial properties of silver nanoparticles increase with the reduced size of the particle. Moreover, Ghorbani et al. [2] have investigated the antibacterial effects of polypropylene/silver on E. coli and S. aureus. In the mentioned study, the polypropylene film surface was modified using the corona discharge method. Surface pre-treatment with corona discharge was reported to increase the adhesion of resin to the film surface for nanoparticle coating. In another research in this regard, polyethylene film was coated with copper nanoparticles, and its antibacterial properties were evaluated. In the mentioned research, the optimum copper concentration was determined in the coating
solution for the preparation of the nanocomposite film in order to enhance the antibacterial effects [3, 4, 5].

In another study, Kim et al. investigated the antibacterial effects of silver nanoparticles on E. coli, yeast, and S. aureus [6, 7]. Furthermore, Ojeda et al. evaluated the morphology and mechanical properties of polyamide/silver composite [8, 9]. In another study, Phu et al. assessed the antibacterial activity of silver nanoparticles using various stabilizers, and the findings indicated that silver-alginate nanoparticles had the highest antibacterial activity against E. coli compared to the other agents, such as silver nanoparticles (AgNPs)/polyvinyl pyrrolidone, AgNPs/polyvinyl alcohol, and AgNPs/sericin [10,11]. In 2015, the mechanical properties of epoxy nanocomposites were investigated, while Moosa et al. used multi-wall carbon nanotubes as filler for the polymer matrix [12, 13, 14].

In another study, the anti-corrosion properties were evaluated after the addition of Zn and Fe2O3 nanoparticles to epoxy [15,16,17]. In 2018, the antifungal activity of polyurethane/CuO film against penicillium was investigated, and the findings demonstrated that the optimum condition was observed with a 2% solution, power of 10,000 W, and using the corona discharge method for duration of five minutes [18, 19, 20].

The present study aimed to synthesize and characterize CuO nanoparticles using ultraviolet-visible (UV-Vis) spectroscopy and dynamic light scattering (DLS) analysis, respectively. Following that, the nanocomposite was prepared through the addition of CuO nanoparticles to the epoxy at various concentrations to assess its antifungal activity. Moreover, the optimal conditions for the antifungal activity were determined.

**MATERIALS AND METHODS**

**Synthesis of CuO nanoparticles**

At this stage, two grams of copper acetate were dissolved in 80 milliliters of pure ethanol and stirred for 30 minutes, and 0.8 gram of sodium hydroxide was added [21]. The solution was poured inside the reactor and placed in an oven for two hours at the temperature of 120°C. After cooling to room temperature, a black product was separated by centrifugation, which was washed repeatedly with deionized water and pure ethanol. Finally, the product was dried at the temperature of 60°C for six hours. UV-Vis spectroscopy, DLS analysis, and transmission electron microscopy (TEM) were used to confirm the presence of the nanoparticles and their size.

**Preparation of the CuO/Epoxy nanocomposite Coating**

The CuO/epoxy nanocomposite coating was prepared by dispersing various concentrations of CuO nanoparticles in the epoxy resin. In the first step, CuO nanoparticles were dispersed at the concentrations of 1, 3, and 5 wt% by ultrasonic examination in acetone solution for 30 minutes. In the second step, two solutions of nanoparticles and epoxy resin were combined using a mixer for 30 minutes.

Afterwards, the hardener was added to the prepared samples under continuous mixing and homogenized.

The product was stabilized for 10 minutes and sprayed directly into steel panels and a filter. Finally, the prepared thin film was dried to evaluate the antifungal properties. The thickness of the obtained film was approximately 75±5 micrometers.

**Antifungal effects of CuO/Epoxy nanocomposites**

The disc-diffusion method is a viable technique for the evaluation of antifungal and antibacterial properties [22]. In the present, a small section of the nanocomposites was removed with various concentrations of CuO nanoparticles. To assess the antifungal activity of the nanocomposites, the inhibition zone was measured in millimeters using a ruler. In addition, tetracycline and cephalaxin discs were used for comparison with the nanocomposites.

![UV-Vis Spectra of CuO Recorded Colloids](image)
RESULTS AND DISCUSSION

**Evaluation of CuO nanoparticle synthesis using UV-Vis spectroscopy**

The color of the reaction solution changed from blue to black, which indicated the formation of CuO nanoparticles [2]. UV-Vis spectroscopy was used to confirm the formation of CuO nanoparticles, and the absorption amount was observed within the wavelength range of 200-600 nanometers. The presence of a peak in the region within the range of 200-300 nanometers indicated the formation of CuO nanoparticles (Fig 1).

As is depicted in Fig 1, a strong surface plasmon resonance was centered at 272 nanometers, which indicated the presence of CuO nanoparticles. This method is an appropriate technique to confirm the presence of metal nanoparticles and metal oxides, such as copper, silver, and copper oxide.

**Evaluation of CuO nanoparticle size by DLS analysis**

DLS analysis was used to determine the distribution of nanoparticles size. Fig 2 shows the size distribution of CuO nanoparticles based on the DLS analysis. The mean size of the nanoparticles was estimated at 45 nanometers (Fig 2).

**Evaluation of the prepared nanocomposite by X-ray diffractometry (XRD)**

The crystalline structure of the prepared nanocomposite was analyzed using X-ray diffractometry (XRD).

The obtained results confirmed the presence of crystalline copper oxide in the prepared nanocomposite [10]. Comparison of the results of the XRD analysis with the standard findings [23, 24, 25] demonstrated that the CuO nanoparticles in the prepared nanocomposite were spherical and quasi-spherical as observed by the peaks at the 2θ values of 36.54°, 38.7°, 53.4°, 58.3°, 62°, 66.4°, and 68.1° (Fig 3).

**Table 1. Antifungal Activity of CuO/Epoxy Nanocomposite against Penicillium and Inhibition Zone (mm)**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Nanoparticle Concentration</th>
<th>Inhibition Zone (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control</td>
<td>-</td>
<td>0</td>
</tr>
<tr>
<td>CuO/Epoxy Nanocomposite</td>
<td>1%</td>
<td>3</td>
</tr>
<tr>
<td>CuO/Epoxy Nanocomposite</td>
<td>3%</td>
<td>6.5</td>
</tr>
<tr>
<td>CuO/Epoxy Nanocomposite</td>
<td>5%</td>
<td>7.5</td>
</tr>
<tr>
<td>Tetracycline</td>
<td>-</td>
<td>14</td>
</tr>
<tr>
<td>Cephalexin</td>
<td>-</td>
<td>0</td>
</tr>
</tbody>
</table>

**Evaluation of the prepared nanocomposite by scanning electron microscopy (SEM)**

CuO/epoxy nanocomposites were evaluated at three concentrations of 1%, 3%, and 5% from the CuO nanoparticles using scanning electron microscopy (SEM) (Fig 4).

SEM analysis showed that the nanoparticles size increased in the nanocomposite with at the highest concentration (5%). However, the nanoparticles size was the same at the concentrations of 1% and 3%. Therefore, it could be inferred that the optimal concentration for the preparation of the nanocomposites was at the concentration of 3%.

**Antifungal activity of the CuO/Epoxy nanocomposites**

The disc-diffusion method was used to assess the antifungal activity of the CuO/epoxy nanocomposite. The inhibition zone was measured using a ruler (Fig 5).

As is depicted in Fig 5-a, fungal growth was observed in the control sample (epoxy), and the CuO/epoxy nanocomposites caused the inhibition zone of the fungal growth. However, the antifungal effects of the prepared nanocomposite
were confirmed (Figs 5-b-5-d). In addition, the inhibition zone was observed to increase at higher nanoparticle concentrations. Table 1 shows the diameters of the inhibition zone. Considering the shape of the inhibition zone and its diameters, it could be concluded that the antifungal effects of the prepared nanocomposite was associated with the concentration of the nanoparticles.

According to the results of the present study, the inhibition zone of fungal growth increased at the higher concentration of the nanoparticles (up to 3%), while the inhibition zone remained the same at the concentrations of 3% and 5%. In fact, the optimal concentration for the maximum antifungal activity was 3%.

![Fig 4. SEM Image of CuO/Epoxy Nanocomposite; a) 1% Concentration; b) 3% Concentration; c) 5% Concentration](image)

![Fig 5. Inhibition Zone; a) Control Sample; b) 1% CuO/Epoxy Nanocomposite; c) 3% CuO/Epoxy Nanocomposite; d) 5% CuO/Epoxy Nanocomposite](image)

![Fig 6. Curve of Inhibition Zone of CuO/Epoxy Nocomposite](image)
Fig 6 shows the diameter curve of the inhibition zone versus the percentage of CuO nanoparticles in the nanocomposite film.

CONCLUSION

In the current research, CuO nanoparticles were synthesized via chemical synthesis and characterized via UV-Vis spectroscopy and DLS analysis. The mean size of CuO nanoparticles was estimated at 45 nanometers. In addition, CuO/epoxy nanocomposite was prepared by mixing the CuO nanoparticles at various concentrations (1%, 3%, and 5%) with epoxy resin. The results of XRD and SEM confirmed the presence of CuO nanoparticles on the nanocomposite surface. The disc-diffusion method was also applied to evaluate the antifungal activity of the prepared nanocomposite. According to the obtained results, the optimal nanocomposite concentration for the maximum antifungal activity was 3%. It seems that the CuO nanoparticles could be used to provide antifungal nanocomposites, which are applicable in medicine and food industries. However, further investigation is required for scale-up.

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