

Embedding of copper sulfate and copper oxide on multipurpose paper

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Abstract. Copper sulfate salts were embedded on multipurpose paper using simple soaking technique while copper oxide particles were embedded using *in-situ* technique. In simple soaking technique, the papers were simply soaked in copper sulfate solution in order for the copper salts to be incorporated in the paper. In the *in-situ* technique, the copper sulfate-embedded papers were soaked in sodium hydroxide solution for reactions to occur that will lead to the formation of copper oxide. Copper sulfate-embedded papers have blue green color while copper oxide-embedded papers have brown color. The copper sulfate-embedded paper shows excellent antimicrobial property against *Staphylococcus aureus*. Meanwhile, the copper oxide-embedded paper shows small zone of inhibition against *Escherichia coli* and *Staphylococcus aureus*.

1. Introduction

Copper-based materials [1-12] such as copper metal, copper sulfate, copper oxide, have antimicrobial properties. Copper sulfate is a salt, which has high solubility in water. This salt has many applications; they are used as antifungal and antiseptic agent, they are also used for the prevention and cure of foot rot in animals, eliminate algae in pools and many more. Meanwhile, copper oxide is a p-type semiconductor with a band gap of 1.7 eV and has a brown color. It has promising application in the creation of solar cells, gas sensor, photodetectors, supercapacitor, photocatalysis, and antimicrobial products. Copper oxide can be synthesized by reacting copper salts (e.g., copper sulfate) with precipitating agent (e.g., sodium hydroxide) to form metastable copper hydroxide, which in turn could oxidize to copper oxide.

Paper is a flexible substrate, which can be easily tailor-fitted to many different forms to suit different applications. The use of multipurpose paper (copier/bond paper) would be very convenient and practical since it is very cheap and widely available almost anywhere. The incorporation of material on paper [13-18] would create product, which would hybridize the flexibility of the paper and functionality of the immobilized material. Copper oxide embedded on paper has many possible applications such as creation of antimicrobial sheets that can be used as patches, covering, lining and the likes. The use of copper-based materials is much cheaper than the use of silver-based particles [19-22], which are commonly used as inorganic antimicrobial agents.



The objective of this research is to embed copper sulfate on 90-gsm multipurpose paper using soaking technique and copper oxide using *in-situ* technique. In soaking technique, the paper is simply immersed in the copper sulfate solution in order for copper ions to penetrate in the pores. Meanwhile, in *in-situ* technique, the copper sulfate-embedded paper is immersed in sodium hydroxide solution for reactions to happen to form the embedded copper oxide particles. The use of copper sulfate and sodium hydroxide as reacting materials are commercially advantageous since they are cheap and widely available.

2. Methodology

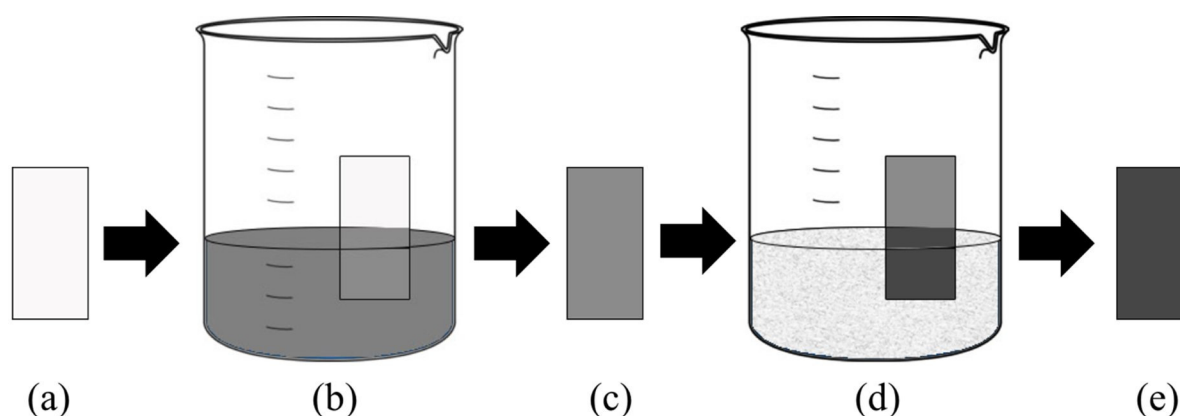


Figure 1. Embedding of copper sulfate salts (a-c) and copper oxide particles (c-e) on multipurpose paper.

Copper sulfate salts were embedded on the paper using soaking technique (Figure 1a-c) while copper oxide particles were embedded using *in-situ* technique (Figure 1c-e).

In the soaking technique, the paper was immersed in copper sulfate solution for 180 minutes. The copper sulfate solution was done by mixing copper sulfate pentahydrate ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$) in distilled water. After drying, the soaking would result to a blue-colored paper indicating the presence of copper sulfate salt.

In the *in-situ* technique, the copper sulfate-embedded paper was immersed in sodium hydroxide solution for 180 mins. After drying, the treated paper changed color to brown as indication of the formation of copper oxide particles.

The images of copper sulfate-embedded and copper oxide-embedded papers were taken using HP Deskjet 2510 Series. The color of the images was analyzed using ImageJ software, in such, the color of the samples was resolved into three components: red, green, and blue (RGB). The RGB values ranges from 0 to 255 wherein the colors become lighter when it approaches 255 while it became darker when it approaches 0.

Furthermore, agar diffusion test was used to determine the antimicrobial property of the 6-mm diameter samples. In this test, the zone of inhibition (ZOI) of the samples was taken.

3. Results

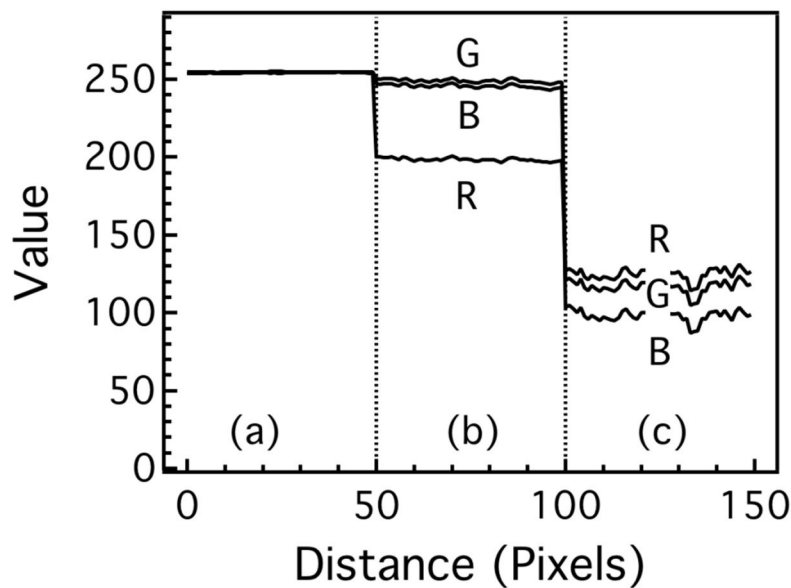


Figure 2. Red, Blue and Green (RGB) values of (a) multipurpose paper (0-49 pixel), (b) copper sulfate-embedded paper (50-99 pixel), and (c) copper oxide-embedded paper (100-149 pixel). The values are taken from different 50-pixel segments.

Table 1. Red, Blue and Green (RGB) values of the different samples and the unevenness of their distribution. The unevenness values are taken to be the standard deviation of RGB values on the 50-pixel segment present in Figure 1.

Sample	RGB values			Unevenness (standard deviation)		
	R	G	B	R	G	B
a. Multipurpose Paper	254	255	255	0.2	0.2	0.2
b. Copper sulfate-embedded in paper	198	249	246	1.0	1.0	1.1
c. Copper oxide-embedded in paper	125	117	98	4.0	4.0	4.0

Table 2. Antimicrobial property of 6-mm diameter copper sulfate-embedded paper against *Escherichia coli* and *Staphylococcus aureus*.

Samples	Zone of inhibition (mm)	
	<i>E. coli</i> (gram negative)	<i>S. aureus</i> (gram positive)
Copper sulfate-embedded in paper	0.0	20.3
Sterile distilled water (- control)	6.0	6.0
Streptomycin (+ control)	19.7	15.3

Table 3. Antimicrobial property of 6-mm diameter copper oxide-embedded paper against *Escherichia coli* and *Staphylococcus aureus*.

Samples	Zone of inhibition (mm)	
	<i>E. coli</i> (gram negative)	<i>S. aureus</i> (gram positive)
Copper oxide-embedded in paper	8.0	13.3
Sterile distilled water (- control)	6.0	6.0
Streptomycin (+ control)	16.0	16.3

4. Discussion

We have embedded copper sulfate salts and copper oxide particles on multipurpose paper. Figure 2 shows the Red, Blue and Green (RGB) values of (a) multipurpose paper (0-49 pixel), (b) copper sulfate-embedded paper (50-99 pixel), and (c) copper oxide-embedded paper (100-149 pixel). For presentation purpose, the RGB values of the three types of paper are condensed in one illustration, each occupying 50-pixel segment. Table 1 summarizes the average RGB values of the 50-pixel segment. Meanwhile, the standard deviations of RGB values at the 50-pixel segment are taken to be a measure of unevenness or non-uniformity.

4.1. Embedding of copper sulfate salts on multipurpose paper

Figure 2b shows the RGB values of the multipurpose paper, after it was soaked in copper sulfate solution. The RGB values changed from 254, 255 and 255 (Table 1a) to 198, 249, and 246 (Table 1b), this is from white color to blue color. The bluish color indicates the presence of embedded copper sulfate salt on the paper. In aqueous solution, copper sulfate salt dissolves into ions enabling them to penetrate to the pores and small spaces on the soaked paper. The ions recrystallized into salt when the paper was removed from the solution and then dried. The unevenness of the RGB color distribution are 1.0, 1.0 and 1.1 (Table 1b), respectively. These values are small, which indicates uniformity in color distribution.

Table 2 shows the antimicrobial properties of the copper sulfate-embedded paper against *Escherichia coli* and *Staphylococcus aureus*. The zone of inhibition of the 6-mm diameter paper sample against *S. aureus* (gram-positive) is 20.3 mm; this value is greater than that of the positive control (Streptomycin) which is 15.3 mm. However, no zone of inhibition was formed against *E. coli* (gram-negative). This was attributed to the stronger cell wall of this gram-negative bacteria.

4.2. Embedding of copper oxide on multipurpose paper

Figure 2c shows the RGB values of the copper sulfate-embedded paper, after it was soaked in sodium hydroxide solution. The RGB values changed from 198, 249, and 246 (Table 1b) to 125, 117, and 98 (Table 1c), this is from blue to brown color. This brownish color indicates the presence of embedded copper oxide particles on the paper. Sodium hydroxide reacts with the embedded copper sulfate salt to form metastable copper hydroxide, which oxidized to form copper oxide particles. However, the unevenness of the RGB color widens to 4.0, 4.0, 4.0 (Table 1c) indicating higher non-uniformity of color distribution.

Table 3 shows the antimicrobial properties of the copper oxide-embedded paper against *Escherichia coli* and *Staphylococcus aureus*. The zone of inhibition of the 6-mm diameter paper sample against *S. aureus* (gram positive) is 13.3 mm; this is lower than that of the positive control (Streptomycin) which is 16.3 mm. The higher zone of inhibition of copper sulfate-embedded paper compare with copper oxide-embedded paper against *S. aureus* is attributed with the higher ability of copper sulfate to be dispersed in aqueous solution due to its higher solubility. Meanwhile, the zone of inhibition against *E. coli* (gram negative) is 8.0 mm, which is a little more than the size of the paper (6 mm). This is far less than the positive control (Streptomycin) which is 16.0 mm.

5. Summary and Conclusion

Copper-based materials (copper sulfate and copper oxide) were embedded on multipurpose paper for antimicrobial applications. Copper sulfate salts were embedded by soaking the paper on copper sulfate solution, while copper oxide was embedded by soaking the copper sulfate-embedded paper in sodium hydroxide solution to undergo series of reactions. After the paper was soaked in copper sulfate solution, its color changes to blue color, indicating the presence of embedded copper sulfate salt. After the paper was embedded in sodium hydroxide solution, the color changes to brown indicating the presence of copper oxide particles. The copper sulfate-embedded paper shows zone of inhibition of 20.3 mm against *S. aureus*; this value is higher than that of the positive control. Meanwhile, the zone of inhibition of copper oxide-embedded paper against *E. coli* and *S. aureus* are 8.0 mm and 13.3 mm, both are lower than the positive control.

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