POLYPROPYLENE/nano-copper composites have been prepared with 0.5, 1, 1.5 and 2.5 wt.% nano-copper particles by mixing the nano-copper physically with the polypropylene granules then extruded by a twin screw extruder. A comprehensive study on these composites was conducted by measuring the material relative permittivity, electrical conductivity, thermal properties and tensile strength. The results showed a significant enhancement of the composite behavior up to 1.5 wt.% nano-particles addition. The increase of the nanoparticles up to 1.5 wt.% Cu decreases the relative permittivity and slightly increases the electrical conductivity. Finally, the fabricated composites with different nano-copper contents have large findings within different applications especially in electronics, packaging, and environmental issues.

**Keywords:** Nano-copper/ polypropylene composite; Thermal properties; Electrical conductivity; Tensile strength

**Introduction**

During recent years, an incredible increase in plastics production was observed due to its wide application in many areas of life such as packaging, construction, the automotive industry or in electronic equipment. Polyolefin, polyethylene and polypropylene (PP) is about 50% of all produced plastics [1]. Polypropylene (PP) which is a non-synthetic thermoplastic polymer [2, 3] has many advantages as a matrix resin such as its relatively low density [4], low cost [5], and high chemical resistance coupled with ease of fabrication such as extrusion, injection and compression molding. [6]. PP has an excellent piezoelectricity [7], thus the growth of the usage of PP as a matrix or reinforced materials increased. PP added as a fiber reinforced in the concrete to improve its properties and indicate higher electrical resistivity than glass and steel fiber [3, 8]. A matrix of PP in almost cases such as PP/wood composite mainly increases its stiffness [9].

Polymeric/nanocomposites consisting of inorganic nanoparticles and organic polymers represent a new class of materials that exhibits an improved performance compared to their microparticle counterparts. It is therefore expected that they will advance the field of engineering
applications [10]. Some studies have shown that metal nanoparticles act as nucleation centers in the orientation of polymer chains which would in turn increase the crystallinity of the polymer [11]. The polymer film capacitors for the long chain branched PP which increases the energy density by increasing breakdown field (E) was studied [12]. Applying copper coating on a textile fabric such as polypropylene can improve its properties for using in fog collectors [13]. Thermal degradation of polymers has been investigated for many years to assist probing degradation mechanism and predicting the thermal stability of polymers [14].

Copper reinforced polymer matrix composites are widely studied for the applications in electrostatic dissipation (ESD) and electromagnetic interference (EMI) shielding and they display good impact energy absorption characteristics [2]. Copper and silver have been widely used as antimicrobial agents in agriculture, healthcare, and industry in general. Copper metal is cheaper than silver but it has a corrosion process in standard conditions. PP products are extensively used in medical devices, packaging products, textiles, delivery systems for solid and liquid pharmaceuticals and automotive industry due to its good processability, good physical and thermal properties [11, 15]. Copper nanoparticle corrosion in distilled water has a better behavior than microparticles [15]. The nanoparticle with ≤ 100 nm in size allowing close interaction with microbial membranes, further enhancing its effect [16]. The main chemical techniques to synthesize copper nanoparticles are chemical reduction, micro-emulsion, sonochemical reduction, electrochemical, microwave-assisted, and hypothermal synthetics [11]. This work aims to show the effect of the addition of nano-copper particles as a reinforcement to the studying of the polypropylene matrix to improve its electrical, thermal, and mechanical properties.

**Experimental**

**Materials**

The matrix is a polypropylene (PP) with density 0.9 g/cm$^3$ and melting index of 5.5 (g/10 min). PP is from petro rabigh Co. was used as a matrix. Nano-copper with density 8.9 g/cm$^3$ and 50 nm particle size and purity 99.99% was used as a reinforcement material. Nano-copper particles were prepared by an electroless deposition technique using formaldehyde as a reducing agent for nano-copper sulphate solution in an alkaline tartarate path at a pH ~ 12. For each liter of distilled water, 200 ml of formaldehyde, 35 g of CuSO$_4$, and 170 g of potassium sodium tartarate and ~ 50 g of sodium hydroxide were added. The copper powder was filtered and cleaned with distilled water. After drying at 120 °C for 2hr, the powder was subjected to hydrogen gas at 450 °C to convert oxidized copper into metallic copper according to the following reaction:

$$\text{CuO(s)} + \text{H}_2(\text{g}) \rightarrow \text{Cu(s)} + \text{H}_2\text{O}$$

**Nano-Copper Powder Characterization**

The Brukur advanced X-ray diffractometer model D8 Kristalloflex and Field emission scanning electron microscope (FE-SEM; QUANTAFEG250, Holland) were used to characterize the nano-copper powder.

**Preparation of Composite**

Polypropylene/nano-copper composite have been obtained by mixing PP granules with nano-copper and in the next stage by extrusion. The temperatures of cylinder heating area were: I- 160 °C, II- 190 °C, III- 220, IV- 230 °C, nozzle-220 °C.

**Microstructural Characterization**

The scanning electron microscopy (SEM) utilizing “FEI Company, Quanta 250 FEG, made in Netherlands” and accomplished with X-ray energy dispersive analysis (EDAX) were employed for pure PP and PP/ Nano-copper composites.

**Dielectric Permittivity and Electrical Conductivity**

The dielectric permittivity and electric conductivity of PP/nano-copper composite materials were measured by AC-Impedance Analyzer Model Hp-4192A- LF.

**Differential Scanning Calorimetry (DSC)**

Thermal analysis of PP and its composite with nano-copper has been performed using differential scanning calorimetry Model TGAQ500 with sample 10-15 mg in weight. Each sample was heated from room temperature to 600 °C with a heating rate of 10 °C/min under argon atmosphere. The melting and decomposition temperature of the composite was obtained as endothermic peaks from the heating scans.

**Tensile Strength**

The tensile strength of the PP/nano-copper composite materials was examined at room temperature using LMF-L20KN universal testing machine.
Results and Discussions

Nano-Copper Powder Characterization
The scanning electron micrograph of copper powder is presented in Fig. 1. This figure showed that the copper powder has a fine spherical particle shape with an average size of ~50nm. Figure 2 shows the X-ray pattern of the copper powder.

Microstructure of the Prepared PP/Nano-copper Composite
Scanning electron micrograph and X-ray energy dispersive analysis (EDAX) for pure PP are shown in Fig. 3. While the EDAX mapping for 0.5, 1, 1.5, 2 and 2.5 wt.% Cu PP/nano-copper composite are illustrated in Fig. 4 that shows the good distribution of nano-copper in the PP till 2 wt.%Cu.

Dielectric Properties
Figures 5 and 6 show the dielectric permittivity and electrical conductivity of PP and PP/nano-copper composite materials. True conduction has no effect on the real permittivity, which only measures energy storage [17]. The permittivity decreases with increasing the nano-copper percent up to 1.5 wt.%Cu then it increases again for 2.5 wt.%Cu. From Fig. 5, the permittivity (k) of the polypropylene is about 3.4 this may be higher than normal due to the polarization mechanism which is responsible for the reduction or increase in the overall permittivity of nanocomposite [18]. The electrical conductivity of 1.5 wt.%Cu...
Fig. 3. The SEM and EDAX of pure PP.

Fig. 4. The EDAX mapping of the PP/Nano-Copper composite for (a) 0.5 wt%Cu, (b) 1 wt%Cu, (c) 1.5 wt%Cu, (d) 2 wt%Cu and (e) 2.5 wt%Cu.
has the highest value as shown in Fig. 6. This can be attributed to the high electrical conductivity of copper more than PP but the decreasing of it for 2.5 wt.% Cu may be due to the non-homogeneous distribution of copper in the PP matrix due to its higher percent and the nature of nano-particles that tends to agglomerates. This affects the motions of the electrons in the PP matrix which decreases the conductivity.

**Differential Scanning Calorimetry (DSC)**

The heat flow of the five samples which have 0, 0.5, 1, 1.5 and 2.5 wt.% PP/ nano-copper composite are presented in Fig. 7 with heating rate 10°C/min. Figure 7 shows thermal analysis of PP and PP/nano-copper composite materials. The figure illustrates two peaks corresponding to endothermic reactions in the pattern of the heat flow as function of the temperature. The first peak corresponds to the melting point of polypropylene, which begins at 171.02 °C and decreases by the addition of nano-copper particles for 1.5 wt.%Cu sample to 167.46 °C then increases again to 170.19 °C for 2.5 wt.%Cu sample.

The slight increasing of melting point in the composites may be attributed to the great stability of the PP crystals likely that formed for stress-induced crystallization in contact with the nano-copper (oriented nucleation) after the mixing.
process [19]. The second peak corresponds to the decomposition reaction of polypropylene. The temperature of the decomposition of polypropylene is 455 °C and it decreases with increasing the nano-copper addition until 445.35 °C for 1 wt.%Cu sample, then increases again to 451.6 °C for 2.5 wt.%Cu sample. The decomposition rate is related to the thermal conductivity of the composite materials [20].

**Tensile Strength**

Figure 8 shows the tensile strength and elongation percentage for the prepared composite materials. It is clear that, the tensile strength decreases with increasing the nano-copper addition until 1 wt.%Cu suggesting a rise of rigidity due to the addition of nano-copper then it decreases again. The same behavior for the elongation, it decreases first until 1.5 wt.%Cu and then increases also.

**Conclusion**

Polypropylene/nano-copper composite materials were fabricated with 0.5, 1, 1.5 and 2.5 wt.%Cu reinforced nanoparticles. The increase of the nano particles up to 1.5 wt.% Cu decrease the relative permittivity and slightly increases the electrical conductivity. This associated with decreasing the melting and the decomposition temperatures due to changing the polarization mechanism of the polypropylene. These effects
accomplished with the decrease in tensile strength and the elongation of the composite materials. The properties of 2.5 wt.%Cu addition were reversed than 1.5 wt.%Cu. Finally, the fabricated composites with different nano-copper contents have large findings within different applications especially in the electronics, packaging, and environmental issues.

References


2.5 and 1.5, 1, 0.5. Prepared bismuth / nanoscale copper by mixing the nanoscale copper with bismuth particles, then thrown by a dual device. A comprehensive study was conducted on these compounds by measuring the relative electrical conductivity, thermal properties and tensile strength. The results showed a significant improvement in the behavior of the compound containing 1.5% nanoscale copper. Increases in the nanoscale particles increased the relative electrical conductivity slightly. Finally, the compounds containing different contents of nanoscale copper showed promising results for various applications, especially in electronics packaging and environmental issues.


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