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Antimicrobial copper nanoparticles synthesized from waste printed circuit boards using advanced chemical technology

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ABSTRACT

Waste Printed Circuit Boards (WPCBs) were classified as one of the most important resources for urban mining containing high purity Copper (Cu) and other valuable materials. Recently, a dissolution recycling approach enhanced by ultrasonic treatment succeeded in the liberation of Cu foils from WPCBs as received. This research aims to synthesize Copper Nanoparticles (Cu-NPs) from the recovered Cu by using an advanced chemistry approach to obtain nano-product with high added value taking into consideration environmental risks. The experiments were carried out on the Cu foils recovered from the three types of WPCBs with different purity of Cu (Motherboard, Video Card, and Random Access Memory (RAM)). The synthesis process was performed in two stages: (a) preparation of Copper (II) Sulfate aqueous solutions from the recovered Cu and (b) chemical reduction of solutions for synthesis of Cu-NPs by using Native Cyclodextrins (NCDs), particularly ß-NCD as stabilizers. The efficiency of the developed approach for raw material of different purity was assessed and the final yield and the estimated recovery cost of synthesized Cu-NPs were calculated with high accuracy as well as the properties of the synthesized Cu-NPs. The obtained Cu-NPs were examined using SEM-EDS, TEM, XRD, Raman Spectroscopy, and TGA. To maximize the potential biomedical application benefits, the antibacterial activity of Cu-NPs was investigated by the standard microdilution method for *E. coli*, *P. aeruginosa*, and *S. aureus* bacterial cultures. The results showed that the produced Cu-NPs had an average size of 7 nm and yield 90%, while the preparation costs were 6 times lower in comparison to the commercial counterparts. In addition, the results indicated that the synthesized Cu-NPs from RAM sample had a good antimicrobial action.

1. Introduction

Electrical and electronic equipment (as well as electronic waste) contains many precious and valuable metals to make it more functional and provide specialized properties (Cayumil et al., 2016; Zhang et al., 2017a, b, c, d). Copper (Cu) is typically the most representative metal in electronics, especially in the case of Printed Circuit Boards (PCBs) due to its good electrical characteristics (Hait, 2018); Cu can be found in the electronic components within complex structures, what makes it difficult to recover Cu with high yield at the end of life of waste PCBs (WPCBs) (Sun et al., 2016). According to the studies conducted in the field of E-waste stream analysis and management, 50 million tons of E-waste are produced each year with an annual growth rate around 5% as a result of rapid increase in lifestyle demands of human beings and shortening lifespan of the products (Ardi et al., 2016; Ning et al., 2017). Waste Printed Circuit Boards represent 10 wt% (5 million tons annually) of E-waste and the estimated amount of Cu in WPCBs represents about 22 wt% (1.1 million tons annually, what is about 6.1% of total smelter copper production) (Brininstool, 2016), therefore WPCBs are considered a source for urban mining of Cu that can be used to face the shortages in EU and other countries (Ciacci et al., 2017; Salhofer et al., 2016).

However, Brominated Epoxy Resin (BER), which adheres all layers of WPCB, can be a source of toxic and carcinogenic materials that may lead to environmental and health problems (Verma et al., 2017; Ikhlayel et al., 2017). Similarly, soldermask is which

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used to isolate specific layers in PCBs can be another source of toxins and some studies were developed to eliminate or reduce toxic emissions occurring during the treatment of electronic waste (Raele et al., 2017; Saini et al., 2017). In the last ten years, many WPCB recycling methods have been created, these methods include advanced techniques for Cu liberation developed due to the environmental safety considerations in order to reduce the rate of contamination resulting from decomposition of BER and economic benefit considerations in order to recover the valuable metals (Xing and Zhang, 2013; Ghosh et al., 2015). These processes often begin by removing the mounted electronic components from the WPCBs and manual separation using mechanical tools (such as plyers, etc.) is the most common route for that purpose that is unfortunately suitable for laboratory scale only (Verma et al., 2017), therefore several approaches were developed to approach disassembly of these components on larger scale while taking environmental issues in account (Bovea et al., 2016; Wang et al., 2015) and start recycling of the bare boards.

Zhang et al. (2016) classified these recycling methods into mechanical, thermal and chemical (Zhang and Xu, 2016). Also, there is another classification for recycling of E-waste (including WPCBs) based on the final product, in particular metallic and non-metallic fraction. Since Cu is the main element in WPCBs, most of the metal recycling practices were focused on Cu recovery using many approaches including leaching, bioleaching, ball milling process, etc. (Zhang et al., 2018; Işıldar et al., 2016; Zhang et al., 2017a, b, c, d). In addition, some studies were targeting other metals like tin, lead, iron, etc., since some of these elements are classified as a heavy metals and may cause major harm to human health (Yang and Xu, 2017; Torres et al., 2018). Regarding the recycling of non-metals fraction, many approaches have been developed to decrease the emissions of non-metallic components and decrease the amount of waste directed to landfills, for instance physical and chemical recycling processes (Gao et al., 2018; Kumar et al., 2018; Zhang et al., 2017a, b, c, d; Kaya et al., 2016).

Most of these techniques had certain limitations related to capacity, power consumption, and recovery rate. Among these methods, the chemical method, supported by the ultrasound treatment, succeeded in the separation of all layers of WPCB with recycling rate 100% and recovery of copper layers as received through dissolution of BER in organic solvent and separation of metal from non-metal layers (Tatariants et al., 2017b). The recycled non-metallic components, in particular fiberglass can be reprocessed into filler materials for composite applications or used as received (Zheng et al., 2009; Yousef et al., 2017; Hadi et al., 2013). Currently, there are no industrially feasible and environmentally friendly approaches for recycling of metal components from WPCBs, especially Cu-containing ones, therefore such process is rarely conducted. Also, the possible repeated usage of the recycled material in PCB manufacturing seems to be problematic at least for the time being because of two reasons: small pieces' size and contamination of the recovered metal (Verma et al., 2017). Therefore, the current challenge for the researchers lies in the development of Cu purification methods and finding the appropriate use for the recovered Cu that should be in the area of advanced applications, which need high purity of Cu and good electrical conductivity of materials.

Nanomaterials usually have specifically outlined field of application due to their unique properties, for instance Cu Nanoparticles (Cu-NPs) can be used for high thermal conductivity and high strength alloys as well as antibacterial and antiviral materials (Din and Rehan, 2017). Furthermore, Cu-NPs are widely used in various fields such as photochemical catalysis, electronics, optics, biosensing, gas sensors, electrochemical sensing and solar/photovoltaic energy conversion (Khodashenas and Ghorbani, 2014; Gawande et al., 2016). Recently, there were some attempts to produce Cu-containing nanoparticles from WPCBs, for example, Xiu et al. (2009), Shokri et al. (2017), and Xiu et al. (2017) used electrokinetic, thermal micronizing, and supercritical methanol processes for this purpose respectively (Xiu and Zhang, 2009; Shokri et al., 2017; Xiu et al., 2017). Although results were promising and nanoparticles with average size <500 nm were produced, different restrictions related to energy consumption, process efficiency, recovery yield or recycling rate, economic profitability, pollution rate, and focus on metals recovery only emerged during the nanoparticles preparation processes. In addition, the prepared nanomaterials were not only pure Cu-NPs but Cu-Tin alloy and nano-Cu_2O what indicates the difficulty of producing pure Cu-NPs from WPCBs since Cu can oxidize or dissolve other metals inside itself during the treatment process if WPCBs are treated as received without separation into fractions. In addition, the reported techniques did not recover most of non-metallic components of WPCBs; therefore, such methods can encounter a difficulty of the residue disposal. Accordingly, liberation of Cu foils from WPCBs at the beginning and preparation of Cu-NPs from the recovered foils can be a promising way to increase the yield and purity of the synthesized Cu-NPs.

Besides Cu-NPs, there were some attempts to produce other types of nanomaterials from WPCBs such as lead nanoparticles and carbon nanotubes (Zhan et al., 2016; Quan et al., 2010). There is a number of techniques to synthesize Cu-NPs such as chemical reduction, sonochemical reduction, thermal reduction, ɣ-radiation, laser ablation, reverse micelles, electron beam irradiation, microemulsion techniques, wire explosion, etc. (Rafique et al., 2017). Also, green chemistry was recently used as an advanced eco-friendly synthesis method to prepare Cu-NPs employing environmentally benign non-toxic solvents (Nasrollahzadeh et al., 2017). Native Cyclodextrins (NCDs) proved to be highly effective in the field of green chemistry; they are used in many applications related to pharmaceutical, food, and cosmetic industry (Davis and Brewster, 2004; Szejtli, 2005). NCDs are circular-shaped molecules with relatively simple structure main feature of which is an ability to form inclusion complexes by hosting small molecules within their hydrophobic cavity at basic pH. Native Cyclodextrins are soluble in water due to the presence of primary and secondary hydroxyl groups. In addition, NCDs are non-toxic by their nature therefore their usage can be highly recommended above other more hazardous chemicals (Loftsson and Duchêne, 2007).

Therefore, (Suárez-Cerda et al., 2016) used NCDs (α-, ß-, and γ-NCD) as a stabilizing agent to synthesize Cu-NPs through the chemical reduction of aqueous solutions of Copper (II) Sulfate ($CuSO_4$) with ascorbic acid (Suárez-Cerda et al., 2016). The results showed that the nanoparticles synthesized by the developed method were stable for several months without physical or chemical changes with the size range 2–23 nm. The optimum stabilizing agent was ß-NCD, which facilitated the production of nanoparticles with minimum size distribution due to its rigid structure and low water solubility what makes nanoparticle complex precipitate easily.

Summarizing all the information above, it can be outlined that the current research aims to use ß-NCD as a stabilizing agent to synthesize Cu-NPs from the Cu foils recovered from WPCBs using BER dissolution recycling method. Finally, the antibacterial activity of the synthesized Cu-NPs is examined in order to indicate their applicability in the biomedical field.

2. Experimental

2.1. Materials and recovered copper

Dimethylacetamide (DMA), pure ethanol, and β-cyclodextrin ($C_{42}H_{70}O_{35}$, 1134.98 g/mol) ⩾ 97% were supplied by Sigma–Aldrich. Other reagents used in this work were laboratory grade.