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Deposition and Characterization of Silver Oxide from Silver Solution Recovered from Industrial Wastes

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Authors' contributions

This work was carried out in collaboration of all authors: Authors UEE and VIA designed the study, interpreted the results. Authors OKO, AIA and CIN collected and performed all extraction and characterization, wrote the first draft of the work. Author ASO managed the literature searches. Author UEE managed the analyses of the work, corrected, and performed the final manuscript. All authors read and approved the final manuscript.

Research Article

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ABSTRACT

Aims: To recover silver from industrial waste and use its solution in silver oxide deposition.
Study Design: Extraction, electrodeposition on different substrates, XRD and topographical characterizations.
Place and Duration of Study: Sample: x-ray films were collected from Anambra State University Teaching Hospital, Awka, Nigeria. Electrodeposition at Physics and Industrial Physics Department, Nnamdi Azikiwe University, Awka. XRD characterization was done at Energy centre, Obafemi- Awolowo University, Ile-Ife, Nigeria in September.
Methodology: Silver was recovered from x-ray films by dissolving the silver compounds with concentrated nitric acid. The solution thus formed was used as electrolyte in the electrodeposition of silveroxide. Two metallic substrates (zinc and lead) were used for the electrodeposition both serving as cathode while a copper electrode served as the anode.

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Structural and topographical characterizations were done using XRD and micrograph techniques.

Results: From the result, it was observed zinc substrate gave a good deposition of silver oxide without any impurities whereas the lead substrate gave deposition of silver oxides with lots of impurities

Conclusion: Silver was successfully recovered from wastes and the solution used in silver-oxide deposition.

Keywords: Silver; x-ray films; electrodeposition; XRD; characterization; wastes.

DEFINITIONS, ACRONYMS, ABBREVIATIONS

XRD – X-ray Diffraction.

1. INTRODUCTION

Silver has long been valued as a precious metal and it is used as an investment to make ornaments, jewelry, high valued table wares, utensils, and currency coins. Today, silver metal is also used in electrical contacts and conductors in mirrors and in catalysis of chemical reactions. Among metals, pure silver has the highest thermal conductivity (the non-metal carbon in the form of diamond and super fluid helium II are higher) and one of the highest optical reflectivity [7,1]. In fact silver is the reflective coating of choice for solar reflectors [8]. Different methods have been utilized in the recovery of silver from x-ray films. The methods include; adsorption of silver from synthetic photographic and spent fix solutions on granulated activated carbon in a batch process [1,2]; industrial enzymatic process for the recovery of silver and poly(ethylene terephthalate) (PET) from used lithe film for printing [2,3]; spent fixing bath [3,4]; using a constructed pilot reactor with a protease from an alkalophile [4,5]; and recovery of silver from X-ray film processing effluents by precipitation. Hydrogen peroxide was used as the precipitating agent [5,6], etc. Results from recovery of silver from activated carbon showed that the recovery of silver was improved when these carbons were pre-treated with 0.5 mol/dm³ sulphuric and nitric acids at 25°C. In another research where silver was recovered from X-ray films by dissolving the silver compounds with concentrated nitric acid, it showed that a very high appreciable quantity of silver was recovered [6,7]. Silver is also recovered from photographic processing solution by replacing the silver with another metal such as zinc by electrolysis or by chemical precipitation with sulphide. Silver is then recovered from silver residue. Stripping the gelatin-silver layer by chemical methods using ammonium thiosulphate, sodium thiosulphate, nitric acid or reagents such as sodium cyanide, NaOH, nitric acid or organic compounds cause environmental hazards [8]. They are also either time consuming or very expensive. The use of NaOH at high temperatures also poses a serious industrial safety problem [9]. In this case the resultant effluents should be neutralized. Frequently, industrial waste waters may be acidic or basic and may require neutralization prior to any other treatment or prior to release to a municipal sanitary sewage system. Acid wastes waters may be neutralized with slake lime (Ca(OH)₂, Caustic soda (NaOH) or soda ash (Na₂CO₃). Alkaline waste waters may be neutralized with a strong mineral acid, such as H₂SO₄ or HCl or with CO₂. It has also been reported that increase in temperature up to 98°C increases the quantity of silver that leaches out into solution [10]. Around 18-20% of the world's silver needs are supplied by recycling photographic waste. It has been reported that 25% of the world's silver needs are supplied by recycling out of which 75% is obtained [11]. X-ray therefore can serve as a secondary

source for recycling of silver. Nigeria has the second highest road traffic accident fatalities among the 193 countries in the world, Health Minister, Prof. Onyebuchi Chukwu has said [12]. Many of the victims are either dead or fracture. As a result, the use of X-ray films increased tremendously in the past five years hence the need for recycling.

Silver oxides are commercially available because it is easily prepared by combining aqueous solution of silver nitrate and an alkali hydroxide [9,13]. Silver oxide is used commercially as battery. In organic chemistry, silver oxide is used as mild oxidizing agent. Such reactions often work best when the silver oxide is prepared in situ from silver nitrate and alkali hydroxide. In this research, silver was recovered from used x-ray films using nitric acid and the resultant solution was used for electrodeposition of silver oxide on zinc and lead substrates at room temperature because of their positions in electrochemical series.

2. MATERIALS AND METHODS

2.1 Recovery of Silver

Used x-ray films were collected from Anambra State University Teaching Hospital, Awka, Anambra State, Nigeria. The waste x-ray films were cut into small pieces and weighed. The weighed waste x-ray films were added to 10ml of 1M nitric acid in a 1000ml beaker and shaken to allow uniform dissolution of the silver compounds. The mixture was stirred for 10min after which the polyethylene plates were washed with 800ml of distilled water to ensure no lost of any silver solution [6].

2.2 Electrodeposition

Preparation of the silver oxide film on the metallic substrates was carried out using the electrodeposition technique. 10ml each of the resultant solution was used for electrodeposition. Zinc metallic substrate was used, serving as the cathode while a copper electrode was used as the anode. The electrodeposition was done under 4V and 20 secs at room temperature (26°C). The pH value of the solution was 5.2. Deposition was repeated using lead metallic substrate in place of zinc substrate. (Deposition was repeated at various voltages (1V, 2V, and 3V) with time kept constant but no visible or appreciable result. At the end of electrodeposition, the coated substrates were washed well with distilled water and air dried at room temperature. The thickness of the film was determined gravimetrically. The dried substrates were taken for topographical and structural characterization using photomicrograph (x 60 magnification) and XRD (x-ray minidiffractometer (10) with CuK α radiation and excitation energy of 25KV) manufactured by Radicon Ltd respectively.

3. RESULTS AND DISCUSSION

3.1 Structural Characterization

Fig. 1 shows the x-ray diffraction spectra of silver oxide thin films prepared on zinc metallic substrate at room temperature (26°C). The figure reveals the existence of only one (031) plane of reflection of monoclinic structure of Ag₃O₄ thin film. The lattice constant as was given in the XRD analysis is $a = 3.5787\text{\AA}$. The result showed only one prominent peak with intensity of 100 at (031) plane, and inter planar distance (d) of 2.6762 \AA . The crystalline grain size and thickness were calculated to be 2.9098 \AA and 0.2 μm , respectively. The implication of the results is that the deposition of silver oxide is pure without any impurities

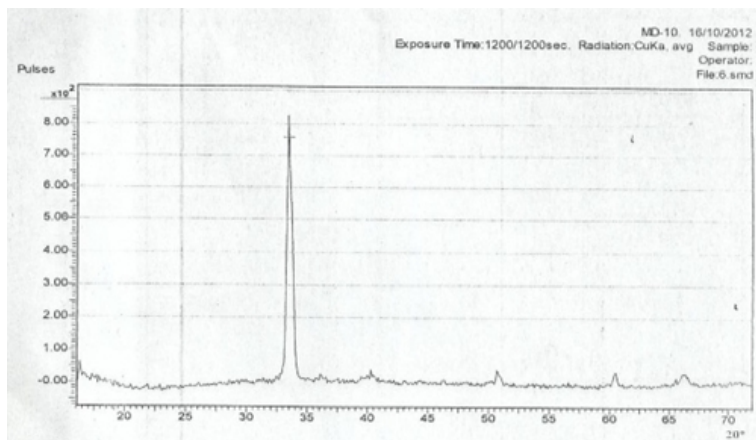


Fig. 1. X-ray diffraction spectra of silver oxide thin films prepared on zinc metallic substrate

Fig. 2 showed the x-ray diffraction spectra of silver oxide thin films prepared on lead metallic substrate at room temperature (26C). The figure reveals the existence of seven (7) prominent peaks of orientation (110), (111), (115), (020), (003), (211), (210) planes of reflections. (111) reflection of highest intensity gave an orthorhombic structure of Ag_2O_3 with intergrowth of (003) and (210) reflections of hexagonal structure of Ag_2O . Impurities were also recorded with (110), (211) reflections of tetragonal structure of PbO_2 , (020) reflection of orthorhombic structure of PbO . The crystals were sharp at inter planar distance of 3.5019\AA and collapsed at 1.55592\AA . The average crystalline grain size calculated is 6.02\AA and $0.5\mu\text{m}$ thickness.

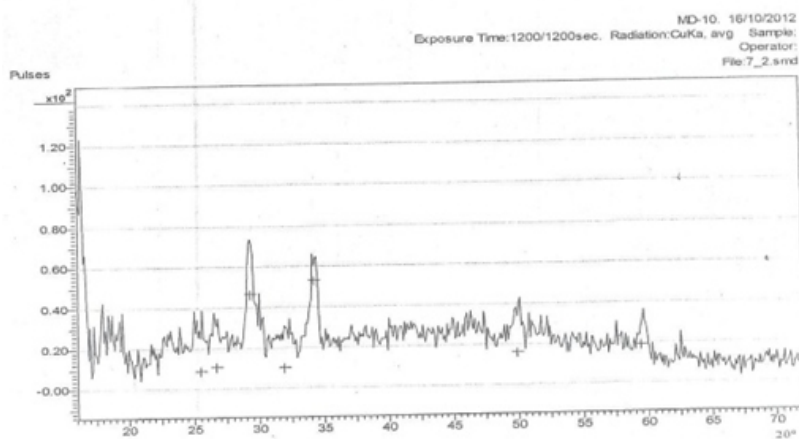


Fig. 2. X-ray diffraction spectra of silver oxide thin films prepared on lead metallic substrate

From the results it is shown that with lead metallic substrate, there were much impurities introduced in the thin film with intergrowth of Ag_2O and Ag_2O_3 , larger crystal sizes and larger thickness whereas with zinc metallic substrate there was no impurity recorded, smaller

crystal size and thickness. The structures (hexagonal, orthorhombic and monoclinic) obtained from these result is different from that (square planar) in report [10,14].

3.2 Topographical Characterization

The topographical characterization of the films deposited was studied in order to determine the small changes in the surface contour of films (lattice surface defects) like dislocations, stacking faults, etc as well evaluating the whole image field. In Figs. 3 and 4 are shown the micrograph of the films deposited on zinc and lead substrates respectively. From Fig. 3, it is deduced that the crystals are compact and have a good agglomeration than those on lead substrate. This could be because zinc is higher up than lead in the electrochemical series hence more effective in displacing silver from the solution.

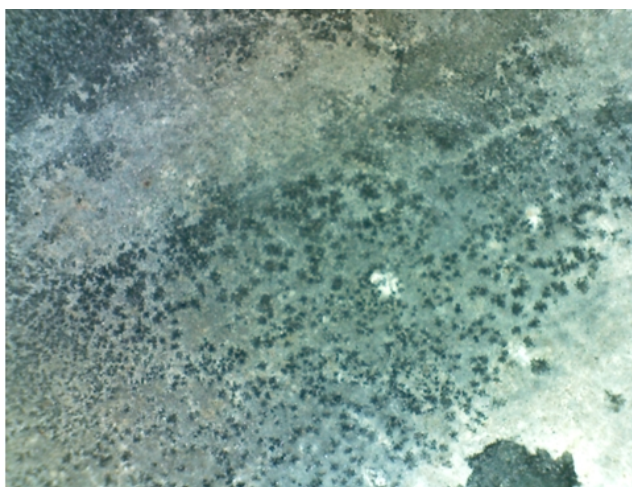


Fig. 3. Micrograph of the film deposited on zinc substrate

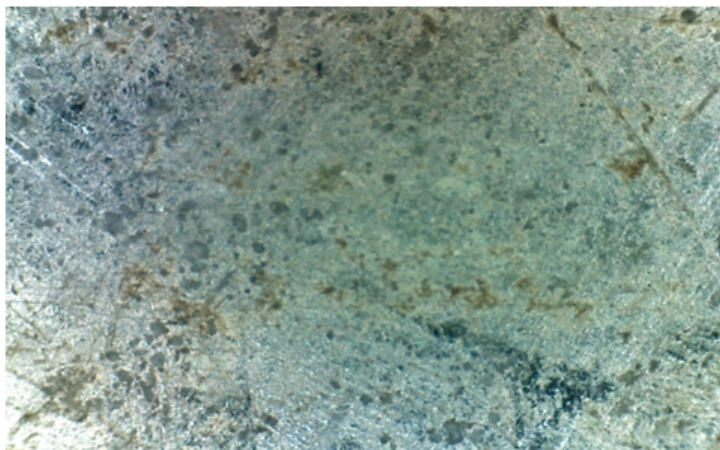


Fig. 4. Micrograph of the film deposited on lead substrate

4. CONCLUSION

Silver was successfully recovered from the industrial wastes and its solution used in thin film deposition. The films deposited on zinc metallic substrate gave a pure silver oxide thin film while the deposition on lead metallic substrate gave intergrowth of silver oxide thin films with impurities. The silver oxide deposited can find usage in silver oxide and rechargeable batteries.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

REFERENCES

1. Edwards HW, Peterson RP. Reflectivity of evaporated silver films. *Physical Review*. 1936;9(9):871.
2. Adani KG, barley RW. Pascoe. Silver recovery from synthetic photographic medical x-ray process effluent using activated carbon. *Minerals Engineering*. 2005;18(13-14):1269-1276.
3. Masui A, Yasuda M, Fujiwara N, Ishikawa H. Enzymatic hydrolysis of gelatin layers on used lith film using thermostable alkaline protease for recovery of silver and PET film. *Biotechnology Progress*. 2004;20(4):1267-1269
4. Han Z, Wei J, Zhao M, Hu J. A method to recover silver from waste x-ray films with spent fixing bath” *Hydrometallurgy*. 2008;92(3-4):148-151.
5. Fujiwara N, Yamamoto K, Masui A. Utilization of a thermostable alkaline protease from an alkalophilic thermophile for the recovery of silver from used x-ray film. *Journal of Fermentation & Bioengineering*. 1991;72(4):306-308.
6. Bas AD, Yazici EY, Deveci H. Recovery silver from x-ray film processing effluents by hydrogen peroxide treatment. *Hydrometallurgy*. 2012;121(4):22-27.
7. Ajiwe VIE, Anyadiegwu IE. Recovery of silver from industrial wastes, cassava solution effect. *Separation and Purification Technology*. 2000;18(2):89-92.
8. Drake PL, Hazelwood KJ. Exposure-related health effects of silver and silver compounds: A review” *Ann Occup Hyg*. 2005;49(7):575-585
9. Nakiboglu N, Toscali D, Nisli G. A novel silver recovery method from waste photographic films with NaOH stripping. *Turk Journal of Chemistry*. 2003;27:127-133.
10. Marinković J, Korać M, Kamberović Ž, Matić I. Recycling Of Silver From Exposed X-Ray. *Filmsacta Metallurgica Slovaca*. 2006;12:262-268.
11. Shankar S, More SV, Laxman RS. Recovery of silver from waste x-ray film by alkaline Protease from *conidiobolus coronatus*. *Kathmandu university journal of science, engineering and technology*. 2010;6(1):60-69.
12. Jaworske DA. Reflectivity of silver and silver coated substrates from 25°C to 800°C for solar collections. *Proceedings of the 32nd Intersociety Energy Conversion Engineering Conference, IECEC-97; 1997*.

13. Janssen DE, Wilson CV. 4-iodoveratoleⁿ org. synth, Coll.1963. 4: 547. Accessed 27th August 2012. Available: <http://www.orgsyn.org/orgsyn/preconten.asp?prep=cv4po547>.
14. Jansen M. Ag₂O₃, a Novel Binary Oxide. Angewandte Chemie International Edition in English. 1985;24(2):118-119.

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