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Silver Recovery Method from Radiographic and X- Ray Film Waste- A Review

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Abstract: The current study describes a simple and efficient method for recovering silver from waste x-ray film by employing sodium hydroxide and sodium sulfide. The principal sources of silver for silver recovery in this article are X-ray film effluent and radiographic waste. Environmental concerns are driving increased attempts to recover, regenerate, and recycle radioactive waste. Though not as hazardous as other soluble salts, soluble salt and emissions from the recovery process are severely damaging to the environment. High-purity products can be generated with low waste thanks to hydrometallurgical procedures such as chemical precipitation and adsorption. Silver recovery using oxalic acid chemical precipitation has been offered as a study topic.

Keywords: Hydrometallurgy, X-Ray film, Radiographic waste, method of Silver recovery, cementation.

Introduction

German physicist Wilhelm Roentgen made the unintentional discovery of the first X-ray machine in 1895. Radiation includes X-rays as well. Despite having significantly shorter wavelengths, they exhibit nearly identical behavior to light rays. The process of creating X-rays involves extracting electron energy and turning it into photons with the necessary energies [1]. A kilo of silver costs around $500, making it a precious and noble metal [2]. Due to its photosensitive properties, it is used extensively in the photographic industry.Recycling is said to provide 25% of the world's silver needs, with photographic waste accounting for the remaining 75% [3]. Silver recovery from photographic waste is critical to the environment and economy. Even after the development and fixing processes, Silver content in the emulsion on the polyester film base ranges from 0.7 to 2.0 percent by weight waste X-ray photographic films, compared to other films, which enhances sensitivity to transmitted X-rays [3].As a source of silver, these waste films are useful in the production of items that are light-sensitive. There have been numerous research over a long length of time to recover valuable silver from these wastes, which have been patented [4],[5],[6],[7],[8]. The films are either directly burned, the metallic silver is oxidized before electrolysis, or a gelatine-silver layer is removed by using various solutions to separate it from the silver. More commonly than the first strategy, two additional techniques have been used [3].In general, the methods reported for the recovery of silver from films involve two steps; As a general rule, silver extraction from films requires two steps: first, the silver must be removed from the film, and then it can be recovered by smelting or electrolysis, depending on the technique of extraction. Leaching is a frequent first step, and it can be It could be microbiological or chemical [8],[9],[10],[11] .Sodium cyanide, for example, is a hazardous chemical that can harm the environment. According to reports, however, the process of using microorganisms is a sluggish one [12],[13]. More and more research is being done on how to extract valuable metals from every type of trash and ore, which has led to an increase in demand. Organic and inorganic waste are the two main types of trash that are generated by industries. Before being disposed of in the environment. It is possible to create less dangerous compounds from biodegradable organic waste. Inorganic trash is composed of both metallic and nonmetallic components[14].

These metals include silver, which is a precious metal of importance to this study, as well as other heavy me tals, such as lead. Precious metal silver (Ag), a byproduct of metallurgical and industrial operations, has bee n used in various industries, including electronics, paper pulping, jewelry, and radiography [15],[16/] When

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it comes to the medical field, its antibacterial and anti-inflammatory characteristics have proven to be effective in the treatment of burn injuries [15]. Silver is utilized in radiography because of its high photosensitivity [17]. The demand for silver is expanding at a rate of 2-2.5 percent each year, according to a research from the World Silver Council. Increased silver manufacturing costs are due to the dwindling supply of silver and the high costs associated with recycling it [17],[18],[19] .According to Syed, et al. [14], the majority of a lack of cost-effective ways for extracting silver results in both economic and environmental difficulties. This and other industrial waste must now be recovered, regenerated, and recycled due to ecological difficulties created by improper disposal of silver-enriched effluents [14]. 5mg/l Ag is the ecological acceptability concentration [19]. Silver can be recovered from a variety of industrial wastes, including radiographs, photographs, and motion picture film. With roughly 2 billion radiographs taken worldwide every year [20], it is estimated that 94 percent to 98 percent of X-rays obtained in the medical area result in the production of photographic chemicals and scrap films as a waste product. Polyester sheets coated on both sides with radiation-sensitive chemicals are used in medical imaging to produce radiographic images [20]. Abdel-Aal and Farghaly estimated that a kilogram of X-ray film produced contains between 14 and 17 kilograms of silver based on their findings [21].When it comes to the synthesis of photographic chemicals, silver salt is used in the vast majority of cases, including the manufacture of X-ray film. The effluent from X-ray film manufacturing companies can have levels of silver ranging from 1 to 12 g/l [18].Generally speaking, there are two types of silver recovery processes: hydrometallurgical operations and pyrometallurgical procedures [21]. There are many different types of hydrometallurgical processes [15],[16],[18],[19],[20]. Water can be purified using a variety of techniques, including electrolysis, metallic replacement, chemical precipitation, adsorption, and liquid membranes, to mention a few. Additionally, biosorption is a promising method for recovering silver since it involves ion exchange, precipitation, absorption, adsorption, and other processes [22], [23], [24], [25], [26], all of which are supported by physico-chemical and metabolic Pyro-metallurgical processes. Combining steps like burning, smelting, drossing, sintering, and melting at high temperatures [27], among others, is the method that is most frequently used to recover silver from X-ray images. [20] When using this method, the manufacturer's website claims that X-ray film polymers are allegedly damaged at temperatures greater than 950°C..The purity of the silver produced is lowered due to the incorporation of carbon during the manufacturing process [20],[21].Silver emissions of 3–25 percent can pollute the air if the process is not well managed, in addition to resulting in low purity [17]. In hydrometallurgical processes, it is feasible to recover more than 99 percent of the heat that is consumed, and the heat demand Depending on the technology used, temperatures can be as low as 100°C. It is the primary goal of silver recovery to minimize the quantity of pollution emitted during the extraction process while also increasing the financial rewards. It is necessary to take into account the health and environmental implications of silver exposure, which are discussed below. Figure 1 shows a schematic representation of silver recovery. [28] Figure 1 depicts the process of recovering silver.

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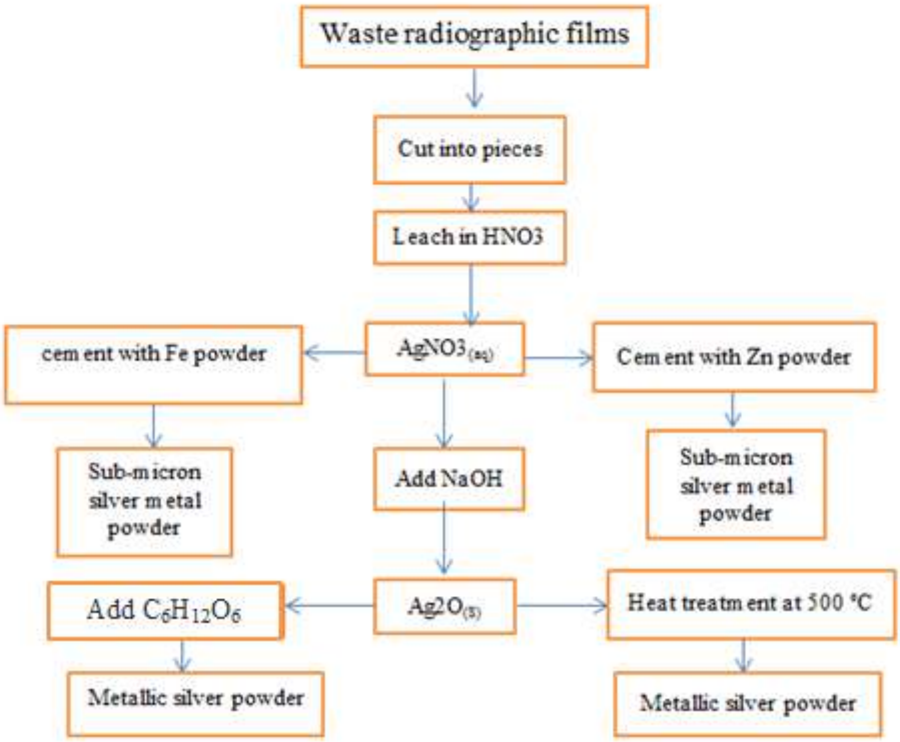

Fig.1. depicts the stages of the silver extraction process.

Fig. 2. X-ray silver recovery[29]

For recovering the silver from used X-ray films, the hydrometallurgical process was developed. It is a novel, easy, quick, inexpensive, and pollution-free technique. Because no burning, oxidizing, electrolysis, or purifying processes are necessary, this approach has a number of advantages. In contrast to other approaches, the conditions for silver recovery were tuned, and 99.8% of the silver was recovered in all experiments, which were distributed across the same flask [30]. Most of the recoverable silver found in x-ray films is present in the fix and bleach-fix solutions. The majority of photography and X-ray wastes contain silver thiosulfate, which contains 4 parts per million of silver [31]. There are numerous methods for

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recovering silver from x-ray waste, including chemical precipitation, metal replacement, burning the film, electrolysis, metal replacement, and metal replacement. The opposing ways to recover the silver are costly and time-consuming [32,33], with the exception of chemical procedures.

Removal of Silver From Radiographic Films That Have Been Wasted

HNO3 dissolves silver nitrate solution rapidly at ambient temperature because it is a noble metal, but HCl does not dissolve it under mild conditions [34]. As a result of this (Equation 1), the following reaction occurs:

$$3Ag_{(s)} + 4\ HNO3_{(a.q)} \rightarrow 3AgNO3_{(a.q)} + 2H2O_{(l)} + NO_{(g)} \qquad (1)$$

Silver's Health And Environmental Effects

Silver is relatively rare within the Earth crust 67th so as of natural abundance of elements [35]. Because of its virtually total biological inertness, pure solid silver has only a tiny negative impact on human health. It would therefore flow through the human body without being absorbed into the tissues even at low doses. Solvent-soluble silver salts, on the other hand, have been shown to cause organ failure and skin pigmentation when present in high concentrations [36]. Table 1 [36],[37] highlights the effects of silver on human health, and it incorporates the following information: In [28], it is impossible to predict how silver and its salts will affect the environment because of how different and unique the disposal body is.Despite the fact that silver can be found in all surface water, it is only found in extremely low amounts in the great majority of instances. It is impossible to predict what the Silver and its salts have an adverse effect on the environment. will be because of the varied composition and chemical changes connected with the disposal body. Sensitive aquatic plants have been seen to develop poorly when exposed for 5 days to concentrations ranging from 3.3 to 8.2 g Ag/litre, while sensitive aquatic plants have died when exposed for longer periods of time to values greater than 130 g Ag/litre [38]. When exposed to nominal water concentrations ranging from 0.5 to 4.5 g Ag/liter, the majority of the afflicted species exhibit growth retardation [38]. Terrain plants can only accumulate a given amount of silver from their soil, and as a result, the amount of garbage containing concentrated silver created by agricultural methods is restricted [19],[38]. It is recommended that silver exposure be avoided during the germination stage of plants, according to Ratte [39], because this is the most sensitive time. Increased concentrations of Ag have a detrimental effect on lettuce, however increased concentrations of Ag in maize will cause the plant to die if the concentration is greater than 9.8mg Ag/litre (Zea mays). Plants such as tomato (Lycopersicon esculentum) and bean (Phaseolus spp.) can be harmed by a spray with 100-1000mg Ag/liter of water. However, it has been claimed that treating soil with silver-laden waste could increase the yield of particular crops [38],[40],[41]. Mucilage, skin, and eyes are all treated with silver during argyria treatment. The most often used method of determining the affected area is based on the color of the area [37].

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Table 1.Silver's Influence On Health In Various Forms[42]

Source of Silver	Health effects
Medicinal	
Silver nitrate - oral ulcerations; Silver nitrate solution - varicose veins; Silver nitrate - topical for gingival bleeding	Argyria*, silver deposits in organs, and abdominal pain
Silver acetate - antismoking gum, lozenges, and tablets	Argyria
Colloidal silver protein - allergy and cold medication	Argyria and high blood-silver levels
Silver protein - nose drops	Argyria
Colloidal protein - eye drops	Argyrosis
Colloidal silver and silver compounds	Argyria, argyrosis
Silver coated pills - mouth freshener; Silver coated acupuncture needles; Silver in water - hemodialysis therapy	Argyria
Occupational	
Soluble	Elevated blood-silver levels; Argyrosis; Argyria; abdominal pain; nosebleed; respiratory irritation; allergic response.
Metallic	Argyro-siderosis of the lungs
Insoluble	Severe circulatory and respiratory symptoms; Argyrosis

The Methods Used To Recover Silver

The silver on x-ray films are recovered at the end of the process with a purity of 99.8% silver in order to be reused for the same or different applications, and the silver is then sold back to the market in the form of bars or billions [43].The hydrometallurgical process begins with leaching, which is the initial stage. In order To thoroughly remove the metal from the polymer substrate during the removal process, it is necessary to provide a series of acidic or caustic solutions over a period of time. Widespread use is made of the technique of shredding and treating X-ray film with sodium hydroxide in order to liberate silver from the film and dissolve it in the solution [21]. For the purpose of separating and concentrating the silver, purification and separation processes Precipitation and solvent extraction, as well as adsorption and ion-exchange, are all employed in conjunction with each other. Techniques such as electro-refining, chemical reduction, and crystallization can be used to remove metal ions from solution[27].

Electrolysis

Electrolysis, or more specifically electrowinning, is that the most generally used and universally applicable method for silver recovery within the photo processing industry [44].It is possible to produce silver with a purity more than 98 percent using the electrolysis method, which includes the application of a direct current between two electrodes. But Ajiwe and Anyadiegwu [45] investigated the recovery of silver from X-ray films. The anode is made of the crude metal to be processed, and the cathode is made of the pure metal to be processed. It has been suggested that silver can be recovered by percolating wastewater via ditches with immersed steel and copper electrodes, followed by internal electrolysis, which can then be utilized to recover the metal. A catalytic agent, cassava cyanide, was introduced to the ditches in order to speed up the procedure. It is possible to remove products in a continuous manner using electrolysis, and the silver-deficient liquid can be recycled [46]. Silver is deposited at the cathode as metallic silver under the influence of a strong current that can reach 300 Amperes[47]. Silver that is purer than 98% can be created using this technique. Pure metal is deposited on the cathode after raw metal is processed on the anode [46].

Metallic Replacement

In addition to cementation, the procedure of replacing metallic materials is also known as rebarbing .In terms of wastewater recovery, Iron, zinc, and copper are some of the more active metals in terms of recovery compared to silver, which makes it more cost-effective than silver recovery. More active metals release ions into solution while less active metals replace them in the solid state [19], causing the more active metals to lose their activity. But the approach introduces impurities of active metals such as Fe^{2+}, Zn^{2+}, and Cu^{2+} into the effluent and silver sludge, necessitating the use of a time-consuming and expensive remediation procedure [18]. As part of the process, there are typically two redox half reactions ,the reduction of the more active metal ion and the oxidation of the less active metal ion are the two reactions that take place [19]. The pH range of 5-7.6 has been recommended for optimal process performance[19]. According to Abdel-Aal and Farghaly When they used Zn metal powder in conjunction with 6 percent nitric acid in their research, they were able to achieve 98 percent Recovery of silver at 90°C with a retention time of 50 minutes was tested[21].

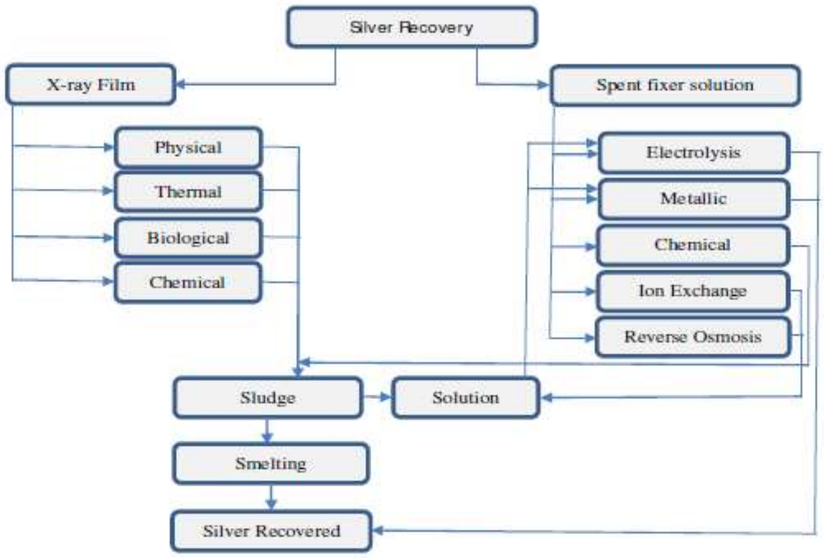
Chemical Precipitation

The definition of precipitation is the formation of a solid from a solution[48]. This method is among the most popular and well-researched ones now accessible in the silver recovery sector. The term for it is chemical precipitation. With sulphide precipitation, silver may be recovered with a recovery efficiency of more than 90% from photographic chemical effluent at concentrations as low as 0.1-1 mg Ag/l [18]. [18] But strict control must be maintained over the precipitation procedure and the amount of sulphide added to the mixture in order to prevent the production of hazardous hydrogen sulphide gas. In this experiment, precipitating agents like the chemicals sodium sulphide, sodium dithionate, potassium borohydride, and 2,4,6-trimercapto-s-triazin were all utilized to recover silver from photochemical processing waste [18]. Bas and colleagues [18] examined ethylene glycol to see how it affected hydrogen peroxide levels because it is used as a stabilizing component in the silver recovery process. [14] [20].Khunprasert and colleagues [20] conducted a study on the usage of oxalic acid, malonic acid, and acetic acid in the silver recovery process, and their findings were published in the journal Chemical Engineering. According to the conclusions of the study, oxalic acid has the best rates of recovery and efficiency of any chemical that has ever been discovered. The recovery process took a total of 20 minutes at 100°C and 5 percent (w/v) oxalic acid to complete. Researchers discovered that increasing the acid concentration of the solution as well as the working temperature were both excellent strategies of enhancing the effectiveness of the procedure. The use of oxalic acid as a leaching agent, according to some sources, is commercially viable because it requires less than 1g/Kg of X-ray to complete the leaching action. It is found naturally in plants such as spinach and rhubarb, albeit in less amounts, and it is effective in extending the shelf life of goods [14],[15],[16]. Among other things, this chemical is used in the manufacturing of medicinal products.

Table 2A Comparison of Several Methods of Recovery[42][49]

Fig. 3 General Method for the Recovery of Silver From X-ray Waste [50]

	Recover y efficiency	Advantages	Disadvantages
Electrolysis	>90%	• 98% purity is achievable • The cathode can be cleaned and reused • Does not produce any new pollutant	• High capital cost of equipment • Operating cost due to electricity requirement • Special electrical and plumbing requirement • Requires monitoring and servicing to ensure high efficiency • Agitation is required to improve efficiency
Metallic replacement	>95%	• Low initial investment cost • Low operating cost • Low maintenance cost due to no mechanical parts and electrical connections • Relatively high recovery efficiency	• Not as efficient as other process • Impurities by presence of other metals in the silver sludge • Recovered silver in the form of sludge need further treatment • Unit must be replace each time it is expended
Chemical Precipitation	>99%	• Low silver concentration in effluent • Easy to monitor performance	• Require stringent control measure to avoid emission of poisonous hydrogen sulphide



Conclusion

The following findings can be obtained from the laboratory testing of two distinct methods for recovering silver from radiography films: - the silver obtained is of a satisfactory quality. Silver can be purified by electrically refining, and both procedures are easy and inexpensive.

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