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Chemical Treatment of X-Ray Film Waste: Silver Recovery and PET Base Recycling via Pyrolysis

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Abstract:
Radiology departments generate significant quantities of medical waste, specifically discarded X-ray films, which pose environmental risks due to their heavy metal content and non-biodegradable components. Managing this waste is crucial to prevent toxic leaching and to recover valuable resources that are often lost through conventional disposal. This study addresses these challenges through a dual-purpose integrated treatment protocol focusing on high-purity silver recovery and the recycling of the polyethylene terephthalate (PET) base. Silver recovery was analyzed using two chemical approaches: acid leaching with nitric acid (HNO_3) and alkaline stripping with sodium hydroxide (NaOH). Stoichiometric analysis demonstrated that the alkaline method achieved a significantly higher silver yield of 31.29 g/kg, compared to 21.08 g/kg for the nitric acid method. Although nitric acid exhibited faster reaction kinetics, the sodium hydroxide approach proved more efficient and environmentally sustainable. Following silver extraction, the remaining PET base was processed through thermal pyrolysis to produce high-value liquid hydrocarbon fuel. Analytical results showed carbon chain distributions ranging from C_6 to C_{20}, closely resembling the profile of commercial gasoline. This research establishes a sustainable circular economy framework for managing hazardous radiological waste. By optimizing resource recovery through the alkaline-pyrolysis pathway, the study offers a viable solution to minimize environmental impacts while maximizing the economic value of medical waste materials.

Keywords: Medical Waste, Silver Recovery, X-ray Films, Pyrolysis, Sodium Hydroxide.

المعالجة الكيميائية لنفايات أفلام الأشعة السينية: استعادة الفضة وإعادة تدوير قاعدة PET عن طريق التحلل الحراري

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الملخص
تتناول هذه الدراسة التحديات البيئية والاقتصادية الحرجة التي تطرحها نفايات أفلام الأشعة السينية الطبية من خلال بروتوكول معالجة متكامل مزدوج الغرض. تركزت الدراسة على الاستخلاص الفعال للفضة عالية النقاء وإعادة تدوير قاعدة البولي إيثيلين تيريفثاليت (PET) الأساسية. تم تحليل استخلاص الفضة بشكل مقارن باستخدام نهجين كيميائيين مختلفين: الترشيح الحمضي باستخدام حمض النيتريك (HNO_3) والتجريد القلوي باستخدام هيدروكسيد الصوديوم (NaOH). أظهر التحليل المتكافئ أن الطريقة القلوية حققت عائدًا أعلى بكثير من الفضـــة بلغ 31.29 جم/كجم، مقارنة بـ 21.08 جم/كجم للطريقة التي تستخدم حمض النيتريك. على الرغم من أن حمض النيتريك أظهر حركية تفاعل أسرع، إلا أن طريقة هيدروكسيد الصوديوم أثبتت أنها أكثر كفاءة واستدامة بيئيًا. بعد استخراج الفضة، تمت معالجة قاعدة البولي إيثيلين تيريفثاليت المتبقية من خلال التحلل الحراري لإنتاج وقود هيدروكربوني سائل عالي القيمة. أظهرت النتائج التحليلية توزيعات لسلسلة الكربون تتراوح من C_6 إلى C_{20}، تشبه إلى حد كبير البنزين التجاري. تضع هذه الدراسة إطارًا اقتصاديًا دائريًا مستدامًا لإدارة النفايات الإشعاعية الخطرة. من خلال تحسين استعادة الموارد عبر مسار التحلل الحراري القلوي، تقدم الدراسة حلاً قابلاً للتطبيق لتقليل الآثار البيئية إلى الحد الأدنى مع زيادة القيمة الاقتصادية لمواد النفايات الطبية.

Introduction

The rapid expansion of the global healthcare sector, driven by population growth and advancements in diagnostic technologies, has led to a critical environmental challenge: the exponential increase in medical waste generation. According to recent reports by the World Health Organization (WHO), high-income countries generate up to 0.5 kg of hazardous waste per hospital bed per day, while developing nations face the dual burden of increasing waste volumes and inadequate disposal infrastructure [1]. Within this complex waste stream, radiological waste—particularly spent X-ray films—presents a significant challenge. Despite the gradual transition toward digital radiography (DR) and Picture Archiving and Communication Systems (PACS), conventional silver-halide radiographic films remain prevalent in dental imaging, mammography, and general radiography in many developing regions due to their cost-effectiveness and high resolution [2]. Additionally, hospitals worldwide hold extensive archives of historical films that have exceeded their statutory retention periods and now require disposal.

These films are not merely general refuse; they are classified as hazardous chemical waste and pose a significant threat to environmental health if not managed properly. Traditional disposal methods, such as open landfilling or uncontrolled incineration, are increasingly untenable. Incinerating X-ray films releases toxic byproducts, including dioxins and furans, into the atmosphere due to their plastic content, while landfilling risks the leaching of heavy metals and chemical compounds into soil and groundwater systems [3]. As a result, the management of X-ray film waste has evolved from a paradigm of simple disposal to one of resource recovery and urban mining, driven by both environmental regulations and economic incentives.

To develop an effective treatment protocol, it is essential to understand the material composition of the waste product. Radiographic films are composite materials designed for durability and optical precision, primarily consisting of a polymeric support base coated with a photosensitive emulsion. The base, which accounts for the majority of the film's mass, is typically made of polyethylene terephthalate (PET). PET is a thermoplastic polymer known for its high mechanical strength, chemical inertness, and transparency, making it a valuable candidate for recycling if it can be effectively separated from the coating [4]. The core functionality of the film lies in the emulsion layer, a suspension of silver halide crystals—predominantly silver bromide (AgBr) and silver iodide (AgI)—embedded within a gelatin matrix.

The silver content in these films is remarkably high compared to natural geological ores. While a typical silver mine might yield 0.05% silver per ton of ore; medical X-ray films can contain between 1.5% and 2.0% silver by weight [5]. This concentration makes spent films an exceptionally rich secondary source of precious metal. However, the silver is tightly bound within the cross-linked gelatin proteins, necessitating chemical intervention to break the protein bonds (proteolysis) in order to release the metal and clean the PET base.

The motivation for chemically treating X-ray films is twofold: mitigating toxicity and recovering economic value. From an ecological standpoint, silver is a heavy metal that exhibits high toxicity in its ionic form (Ag^+). When films are discarded in landfills, the degradation of the gelatin layer over time can release silver ions into the leachate. Research indicates that free silver ions are highly toxic to aquatic microorganisms, algae, and invertebrates, disrupting enzymatic activities and cell membrane functions even at micromolar concentrations [6]. Therefore, the uncontrolled discharge of untreated X-ray film waste is a direct violation of environmental safety standards in many regions.

Economically, silver is a precious metal with fluctuating but generally high market value, essential for industries ranging from electronics (in photovoltaics and conductors) to medicine due to its antimicrobial properties. The concept of Urban Mining suggests that recovering metals from waste is far more energy-efficient than mining virgin ore. Studies show that recycling silver from waste films consumes significantly less energy and produces a lower carbon footprint compared to primary extraction methods [7]. Additionally, the recovered PET base, once cleaned of the emulsion, can be recycled into polyester fibers or new plastic packaging, providing a secondary revenue stream for the treatment process.

Various methods exist for recovering silver from X-ray films, including incineration (burning the film to ash) and biological treatment (using bacteria or enzymes). However, chemical treatment (hydrometallurgy) is widely recognized in recent literature as the most commercially viable method due to its speed, scalability, and high recovery efficiency [8].

The fundamental mechanism of chemical treatment involves "stripping" the emulsion layer by immersing the films in a solution that degrades the gelatin binder. This process breaks the peptide bonds in the gelatin protein structure, converting the solid emulsion into a soluble hydrolysate.[9,10] Once hydrolyzed, the silver particles and emulsion sludge separate from the PET base, typically allowing for silver recovery rates exceeding 95% under optimized conditions.[11] This reaction can be summarized as the breakdown of peptide bonds in the gelatin protein structure. Recent research has evaluated several chemical agents for this process:

Alkaline Leaching: Sodium Hydroxide (NaOH) remains the most documented agent. Research indicates that using NaOH (1.0–2.0 M) at elevated temperatures (80–90ºC) effectively cleaves the collagen structure of the

gelatin. Nakiboglu et al. reported that while effective, this method creates a sludge requiring further purification, yet it remains favored for its cost-effectiveness and high stripping efficiency (>98%) [9,12].
Oxidative Stripping: Agents such as Sodium Hypochlorite (NaOCl) or dilute Nitric Acid (HNO_3) operate by rapidly oxidizing the organic matrix. Studies show this method can significantly reduce processing time (often <20 minutes) compared to alkaline soaking [13]. However, strict pH monitoring is mandatory; acidic conditions can trigger the release of hazardous chlorine gas (Cl_2) or nitrogen oxides (NOx), necessitate fume hoods and scrub systems [14,15].
Complexing Agents: Alternative approaches utilize thiosulfate or other cyanide-free lixiviants to dissolve silver directly. While these agents offer high selectivity for silver without degrading the PET base, they are often cited as economically burdensome for large-scale applications due to reagent costs and complex wastewater treatment requirements [16].
Despite the effectiveness of these methods, challenges remain in optimizing process parameters specifically, balancing the concentration of chemicals, temperature, and residence time to ensure complete removal of the emulsion without degrading the quality of the recovered PET plastic.
While the basic chemistry of gelatin hydrolysis is established, there is a pressing need for optimized protocols that minimize chemical consumption and energy usage while maximizing purity. Many existing industrial processes still rely on overly aggressive conditions that damage the polymer base or generate excessive liquid waste requiring further treatment. Therefore, this study aims to investigate and optimize the chemical treatment of medical X-ray films using nitric acid and sodium hydroxide to achieve sustainable resource recovery.

Material & Methods

Sample Preparation

Sample collection involved obtaining used X-ray films from several clinical and diagnostic health centers. The film types varied, including standard radiographic films (primarily for bone and fracture imaging) and Magnetic Resonance Imaging (MRI) films. The films were sorted, cleaned, and wiped to remove any surface contaminants. The cleaned films were cut into medium-sized squares (≈2 cm×2cm) to facilitate handling and optimize the surface area for the chemical reaction.

Silver Recovery from Used X-ray Films

Silver was quantitatively recovered from the emulsion (gelatin layer) of the used films using two different chemical methods:

Nitric Acid (HNO_3) Stripping Method

Concentrated nitric acid (HNO_3) was placed in a glass beaker. and the film clippings were immersed until the color of the gelatin layer was completely removed. The plastic film base was removed and rinsed in a separate beaker containing distilled water. A solution of NaCl was added to the resulting acidic solution (containing dissolved silver). Immediate precipitation of silver occurred, forming a white precipitate of silver chloride (AgCl), accompanied by effervescence. The precipitate was separated, washed with hot water three times, collected, dried, and finally combusted in a crucible. The dried precipitate was mixed with borax flux before combustion in the crucible to facilitate the melting process and ensure the purity of the recovered silver button.

Sodium Hydroxide (NaOH) Alkaline Stripping Method

100 g of NaOH was added to the hot water for every 1 kg of X-ray film used, with continuous stirring. A hot water bath was used to increase the reaction rate, and a separate beaker with distilled water was prepared for washing. The films were immersed in the NaOH solution until the color was fully removed. Sodium Sulfide (Na_2S) was added to the resulting solution. The solution was covered and allowed to stand overnight. The solution turned black due to the formation of silver sulfide (Ag_2S). The solution was filtered, and the resulting precipitate was separated and dried.

Recycling of the Plastic Film Base via Pyrolysis

Following the silver recovery process, the plastic film base was dried, weighed, and subjected to recycling via pyrolysis:
100 g of the dried plastic base was placed into the pyrolysis apparatus. The plastic was heated in the absence of oxygen to a temperature ranging from 300-450ºC. This process thermally decomposed the polymer molecules in the plastic into a liquid product known as "plastic fuel" or "Pyrolysis Oil."
The experimental work was performed using a custom-designed laboratory-scale batch pyrolysis reactor system. The core setup consisted of a three-necked round-bottom flask (serving as the reactor) heated by a PID-controlled heating mantle. The temperature was precisely monitored using a submerged thermocouple. The reactor was connected to a water-cooled condenser for the collection of condensable volatile products, which were subsequently collected in a liquid collection receiver.

The system was sealed and purged with N_2 for 15 minutes to ensure inertness. The reactor was then heated from ambient temperature to the final reaction temperature of 450 °C at a controlled heating rate of 20 °C/min. Heating was sustained until the cessation of condensation was observed, indicating the completion of the thermal degradation process. The resulting liquid product (pyrolysis oil) was collected and weighed to determine the liquid oil yield, and the total reaction time was recorded.

Results & Discussion

Silver Recovery from Used X-ray Films

The silver yields were calculated based on the stoichiometry of the precipitates (AgCl for the acid method and Ag_2S for the alkaline method).

Nitric Acid (HNO_3) Method

The silver present on the X-ray films (as AgBr) reacted with concentrated nitric acid. Silver was then precipitated as silver chloride (AgCl) using NaCl salt.

$$AgBr + HNO_3 \longrightarrow AgNO_3 + HBr$$

$$AgNO_3 + NaCl \longrightarrow AgCl\downarrow + NaNO_3$$

The resulting silver, after precipitation, drying, and burning with Borax, is shown in Figure 1.

Figure 1. Precipitated silver recovered from used X-ray films via the Nitric Acid method

The weight of the recovered precipitate (AgCl) was 28.01 g. The silver content was calculated using the ratio of the atomic weight of silver (107.87) to the molecular weight of silver chloride (143.32):

$$\text{Silver Yield} = 28.01 \times \frac{107.87}{143.32} = 21.08 \text{ g/Kg}$$

Sodium Hydroxide Method (NaOH)

The weight of the recovered precipitate (Ag_2S) was 35.95 g. The stoichiometry of silver sulfide (Ag_2S) indicates the presence of two silver atoms per molecule. Therefore, the conversion factor is calculated based on the molar mass of Ag_2S (247.80 g/mol) and the mass of two moles of silver (215.74 g):

$$\text{Silver Yield} = 35.95 \times \frac{215.74}{247.80} = 31.29 \text{ g/Kg}$$

Figure 2. Precipitated silver recovered from used X-ray films via Sodium Hydroxide method

The results demonstrate that the Sodium Hydroxide method achieved a significantly higher recovery rate (31.29 g/kg) compared to the Nitric Acid method (21.08 g/kg). This indicates that alkaline stripping is not only environmentally preferable but also more efficient in extracting the total silver content embedded in the gelatin matrix.

The comparative analysis of the chemical treatment methods yielded a critical finding: the Sodium Hydroxide (NaOH) method demonstrated a significantly superior silver recovery (31.29 g/kg), exceeding the yield obtained by the Nitric Acid (HNO_3) method (21.08 g/kg). This result challenges the general trend observed in much of the existing literature [17, 18], which often prioritizes the kinetic and oxidizing advantages of strong acids. The HNO_3 method relies on its dual functionality as both a strong acid and a powerful oxidizing agent. This dual functionality enables:

Rapid Gelatin Degradation: The strong acidity facilitates the swift hydrolysis of peptide bonds in the emulsion layer.

Silver Oxidation and Dissolution: HNO_3 actively oxidizes the freed elemental silver particles, converting them into soluble silver nitrate ($AgNO_3$). While HNO_3 offers rapid kinetics, the aggressive nature of the attack may lead to premature or incomplete detachment of the gelatin, potentially encapsulating some silver particles within the residue and preventing their full dissolution and final recovery.

In contrast, the treatment using Sodium Hydroxide relies solely on the Alkaline Hydrolysis of the gelatin binder.

Complete Gelatin Removal: While the reaction kinetics of alkaline hydrolysis are typically slower compared to an aggressive oxidizing acid [17], this milder mechanism appears to facilitate a more thorough and complete separation of the entire gelatin matrix from the PET base.

Maximal Silver Exposure: This comprehensive removal effectively exposed the maximum amount of embedded silver particles to the subsequent precipitation agent. This led to the significantly higher final yield of 31.29 g/kg, a result that surpasses the typical total silver content range (15 to 25 g/kg) reported in much of the literature [18].

The findings from this study demonstrate that the NaOH alkaline method offers a triple advantage that resolves the traditional trade-off:

Highest Economic Return: Achieving maximal silver yield (31.29 g/kg).

Environmental Stewardship: Aligning with "Green Chemistry" principles [19] by producing a less toxic effluent that is simpler to neutralize and treat than the strong acidic wastes generated by HNO_3.

This research confirms that optimal conditions under alkaline treatment can maximize the final recovery yield, thus achieving high efficiency alongside low toxicity. Therefore, the Sodium Hydroxide method is the optimal choice, effectively removing the need to balance economic return versus environmental stewardship, as it maximizes both.

Pyrolysis of the Plastic Base of Used X-ray Films

Analysis of the fuel sample resulting from the pyrolysis of the used X-ray film plastic base was performed at the Libyan Petroleum Institute – Tripoli, using Gas Chromatography (GC) (ENV 2-4), carbon distribution method. The results are shown in Figure 3 and Table 1.

Table 1. Carbon Distribution of Pyrolysis Oil generated from the X-ray film plastic base**.**

Composition	%Wt	Composition	%Wt
C_5	3.179	C_{16}	**3.172**
C_6	9.735	C_{17}	**2.235**
C_7	11.390	C_{18}	**1.658**
C_8	12.468	C_{19}	**0.984**
C_9	15.730	C_{20}	**0.853**
C_{10}	7.513	C_{21}	**0.430**
C_{11}	8.676	C_{22}	**0.287**
C_{12}	6.541	C_{23}	**0.144**
C_{13}	5.907	C_{24}	**0.093**
C_{14}	5.455	**Total**	**100.000**
C_{15}	3.494		

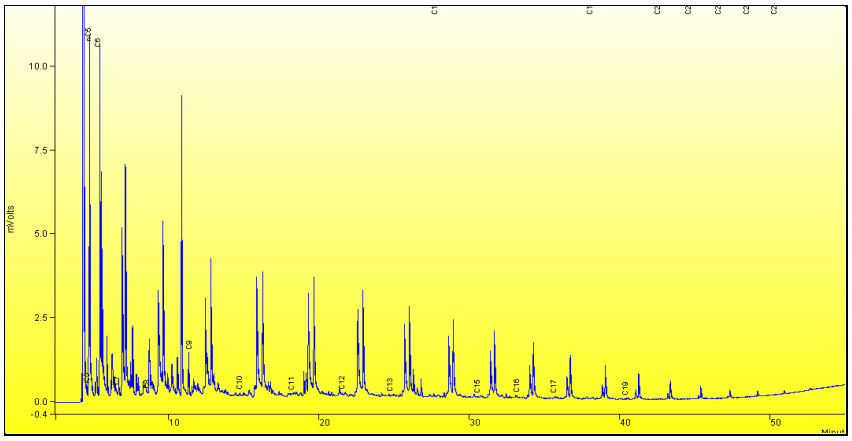

Figure 3. GC spectrum of Pyrolysis Oil generated from the X-ray film plastic base

The treatment of the plastic PET base via Pyrolysis, converting it into valuable hydrocarbon Oils represents a crucial innovation in establishing a fully integrated solution for radiological waste. This conversion is a significant step towards a Circular Economy for healthcare plastics. Instead of sending the non-biodegradable PET to landfills, the Pyrolysis process utilized it as a feedstock for energy production.

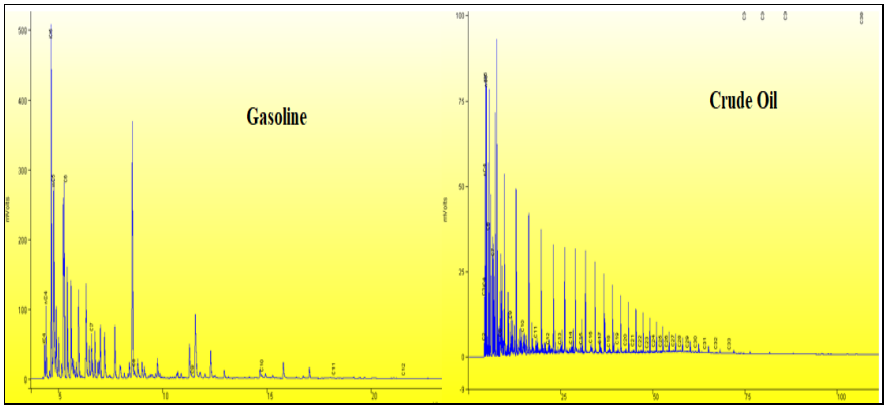

Figure 4. Reference GC spectrum for Crude Oil and Gasoline fuel

- Comparison with Mechanical Recycling: While mechanical recycling of PET is widely researched [20], Pyrolysis offers the unique advantage of creating liquid fuel, which directly addresses energy scarcity and replaces a portion of virgin fossil fuels.
- Fuel Quality: The successful conversion into hydrocarbon oils with properties "similar to petroleum fuel" is a key finding. This suggests the products are viable for direct use or as a blending agent in the fuel industry, significantly enhancing the overall economic viability of the entire waste treatment process.

The selection of the optimal integrated process must consider all inputs and outputs:

- Acid Treatment (HNO_3): Provides the highest immediate economic return from silver but incurs high environmental costs for treating acidic effluent and managing potential NOx emissions.
- Alkaline Treatment (NaOH): Offers a moderate silver yield but is generally more environmentally benign in terms of liquid waste management, aligning better with sustainable development goals.
- Pyrolysis: Provides a highly sustainable solution for the plastic component; however, stringent controls are required for the gaseous emissions generated during the thermal process to prevent the creation of new atmospheric pollutants [21].

Environmental Impact and Limitations: Despite the high recovery rates achieved, the chemical treatment of X-ray films involves reagents that require careful management. The alkaline effluent requires pH neutralization before disposal to ensure environmental compliance, preventing soil and water contamination. Similarly, the use of nitric acid poses atmospheric risks; while the HNO_3 method is kinetic-heavy, it necessitates rigorous NOx emission controls to mitigate atmospheric toxicity. Acknowledging these drawbacks is essential for developing a truly sustainable circular economy model for radiological waste.

Conclusion

This research proposed and evaluated an integrated management model for X-ray film waste, focusing on both precious metal recovery and plastic recycling. The comparative analysis of chemical treatment methods revealed that Sodium Hydroxide (NaOH) is the superior agent for silver recovery, achieving a yield of 31.29 g/kg, significantly outperforming the Nitric Acid method, which yielded 21.08 g/kg. This finding shifts the preference towards the alkaline method, as it combines high extraction efficiency with lower environmental toxicity compared to acid-based treatments. Furthermore, the study successfully demonstrated the viability of the "Zero Waste" concept by converting the stripped PET base into valuable energy. The pyrolysis process transformed the solid plastic waste into a liquid fuel with structural properties resembling gasoline and crude oil (C_6-C_{20} range). In conclusion, the proposed Alkaline-Pyrolysis integrated route offers a robust, profitable, and eco-friendly solution for healthcare facilities, turning hazardous radiological waste into a dual source of precious silver and renewable fuel.

Disclaimer

The article has not been previously presented or published, and is not part of a thesis project.

Conflict of Interest

There are no financial, personal, or professional conflicts of interest to declare.

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