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Removal of metal ions from wastewater using EB irradiation in combination with HA/TiO_2/UV treatment

A.A. Zaki[a,*], Naima A. El-Gendy[b]

[a] Hot Laboratory and Waste Management Center (HLWMC), Atomic Energy Authority, P.O. Box 13758, Inshas, Cairo, Egypt
[b] Department of Polymer, National Center for Radiation Research and Technology(NCRRT), Atomic Energy Authority, Nasr City, P.O. Box 19, Cairo, Egypt

HIGHLIGHTS

- A combined electron beam (EB) and humic acid (HA/TiO_2) treatment process was developed.
- The ion metals Cu^{2+}, Sr^{2+}, and Co^{2+} were effectively removed from wastewater.
- About 50 kGy of EB was needed to remove the Cu^{2+}, Sr^{2+}, and Co^{2+} from wastewater.
- Mechanisms of interactions between HA and metal ions were suggested and discussed.

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ABSTRACT

The electron beam (EB) irradiation technology was applied for removal of Cu^{2+}, Sr^{2+}, and Co^{2+} ions from wastewater. The aim of this study is to achieve an efficient treatment process of wastewater using EB and introducing a combination of humic acid (HA) as a natural organic polymer and ultraviolet irradiation of a TiO_2 (TiO_2/UV), as a suspended catalyst in the treatment of wastewater solutions (TiO_2/UV + HA). The experimental results showed that the percentage removal of Cu^{2+}, Sr^{2+}, and Co^{2+}ions was 41%, 87% and 75% respectively, at 125 kGy. In the presence of TiO_2 photocatalyst and exposure of the investigated wastewater to ultraviolet rays before irradiation by the EB the percentage removal of Cu^{2+} ions became 51%, while the percentage removal of both Sr^{2+} and Co^{2+} ions was slightly improved; was 87% and 75%, respectively at the same EB dose. On the other hand, by introducing the combination of TiO_2/UV + HA, only an irradiation dose of about 50 kGy led to removal of Cu^{2+}, Sr^{2+}, and Co^{2+} completely from the wastewater. Mechanisms of interactions between HA and Cu^{2+}, Co^{2+} and Sr^{2+} metal ions were suggested and discussed.

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1. Introduction

The handling of wastewater appeared to be one of the most important problems in the last few decades. Aqueous wastes may be treated using ion exchange, sorption, chemical precipitation, evaporation, reverses osmosis, filtration and solvent extraction [1]. The main features and limitation of these treatment processes are described elsewhere [2].

Occupational or environmental exposure to cobalt is associated with health effects on various organs, including the respiratory tract, the skin, the red blood cells, the thyroid gland and the myocardium [3]. Historically, effects on the thyroid gland were observed as an undesirable side-effect of cobalt therapy used for its polycythemic action, often in association with iron, in the treatment of various anemias [4].

The major industrial uses of strontium include the production of glass for color television picture tubes, ferrite magnets, and refining of zinc [5–8]. Copper is an essential micronutrient that forms part of several proteins involved in a variety of biological processes indispensable to sustain life [9]. At the same time, it can be toxic when present in excess, the most noticeable chronic effect being liver damage [10]. Radiation technologies are frequently applied for purification of wastewater and groundwater contaminated with organic pollutants [11,12]. Humic acids (HAs) are macromolecular yellow-to-black colored natural organic matter derived from the degradation of plant, algal, and microbial material [13]. Although their formation mechanism and chemical structures are not well understood, they are known to be high in carbon content (50–60%) of both aliphatic and aromatic character and rich in oxygen-containing functionalities such as carboxyl,

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* Corresponding author. Tel.: +20 2 35620445; fax: +20 2 4620796.
E-mail address: aazaki7@yahoo.com (A.A. Zaki).

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phenolic, alcoholic, and quinoid groups [14,15]. The absorption of light by chromophores existing in humic acid (HA) may lead to alterations of HA structure and composition which is reflected by the changes in absorption, fluorescence, chemiluminescence, NMR, IR and EPR spectra [16,17]. Photocatalytic oxidation using TiO_2 is gaining wide attentions as an advanced water treatment technology. TiO_2 photocatalyst initiates upon absorbing UV photons with generating conduction band (CB) electrons and valence band (VB) holes within the particle [18,19]. In particular, the hydroxyl radicals generated through the reaction of VB holes account for the strong oxidizing power of the TiO_2 photocatalytic system. Although TiO_2 has been widely used as a photocatalyst, the band gap excitation of TiO_2 proceeds only in the presence of UV light ($\lambda < 388$ nm) [20]. Sensitized TiO_2 by humic acid (HA–TiO_2), ruthenium-based complexes or various dyes has been extensively investigated in order to extend the spectral response into the visible region [21].

The aim of this work is to achieve an efficient and energy saving process for treatment of wastewater and removal of Cu^{2+}, Sr^{2+} and Co^{2+}ions from waste solutions using a combination of electron beam irradiation (EB), HA sensitized UV-irradiated Ti_2O catalyst (HA–TiO_2).

2. Materials and methods

2.1. Materials

All chemicals used in this work were of analytical grade. All aqueous solutions were prepared with distilled deionized water, which was purified by a Millipore Milli-Q water (resistivity = 4.4 MΩ cm, TOC = 0.1 mg/l) purification system.

Chemical powders used to simulate wastewater were supplied from Merck; $CuSO_4{\cdot}5H_2O$, $CoCl_2$, $SrCl_2{\cdot}4H_2O$. Stock solutions of 100 mg/l from different metal ions were prepared using distilled deionized water. Humic acid (HA) in sodium form and TiO_2 powder, Degases P25 (80% anatase and 20% rutile), were purchased from Aldrich. The TiO_2 powder had a BET surface area of 55 m^2/g and an average particle diameter of 30 nm. HA was initially dissolved in alkaline solution (pH ≈ 12) at 100 mg/l and subsequently adjusted to pH 5.0 using 1 N HCl. All experiments were done at room temperature (298 ± 1 K) otherwise is mentioned. The HA filtrate was stored as stock solution and its concentration was routinely checked.

2.2. Electron-beam research facility

Experiments to study the electron beam (EB) treatment process were carried out using NCRRT-Accelerator. The NCRRT-Accelerator is a direct accelerator delivered from High Voltage Company, USA; 1.5 MeV, beam current of 25 mA, power of 37.5 kW and variable scan width up to 90 cm. This accelerator is used to produce an EB with high energy which can be used for different applications in different fields.

2.3. Electron beam irradiation (EB) radiation of HA molecules

The stability of HA, 50 mg/l, was studied by applying electron beam irradiation (EB) at doses of 0–32 kGy, and its ultraviolet (UV) absorbance was checked. A Unicam UV4 double beam spectrophotometer (product of Unicam Co. Ltd., England) was used to measure absorption spectra of the EB irradiated and unirradiated HA.

2.4. Effect of pH on the uptake of HA by Ti_2O

Uptake studies of HA by Ti_2O, humic acid–titanium dioxide sensitization (HA–Ti_2O), were performing by mixing 500 mg/l Ti_2O with 50 mg of HA for pH ranging from 2.0 to 13.0. The mixture was shaken in a thermostatic shaker at room temperature (298 ± 1 K) for 2 h at 200 rpm. The percentage uptake of HA by Ti_2O was calculated according to the formula $(C_0 - C_e/C_0) \times 100$, where C_0 and C_e are the initial and equilibrium concentrations of HA, respectively. The dissolved humic acid was spectrophotometerically measured using the UV–vis spectrophotometer, Shimadzu-UV 2550, at $\lambda = 254$ nm.

2.5. Photocatalytic TiO_2/HA/metal ion process

Photocatalytic TiO_2 powder (Degases P25) is used in photocatalytic experiments due to its high UV-light efficiency. The TiO_2 suspension was prepared by simultaneous sonication and shaking in an ultrasonic cleaning bath (Branson 3210). Solution of HA was then added to a 100 mg/l TiO_2 suspension to give a desired concentration of HA and added to metal ion solutions. A removable 30 cm long UV-florescent lamp, model DESAGA Abnehmber was used to produce light for irradiating the TiO_2/HA/metal ion solution. It has two wavelengths 254 nm and 396 nm, power of 30 W and the incident light is 60 W/m^2.

2.6. Procedures of experimental setting up

The liquid solution samples were divided into three groups. The first group samples were irradiated for different doses in the range 25–150 kGy. The second group samples were exposed to UV-lamp with photocatalytic TiO_2 powder (Degasses P25) suspended in solution for 2 h (checked well during irradiation) then directly moved to the EB irradiation. The third group samples were mixed with HA and exposed to UV-lamp with TiO_2 photocatalytic for 2 h then directly moved to EB to complete the irradiation. After EB irradiation the samples were centrifuged at 5000 rpm for 10 min before measurement on an atomic absorption spectrophotometer (AAS) as well as the metal ions in the samples of the other two groups. The AAS utilizes air acetylene flame, is a Boch Scientific model 210 VGP, USA.

2.7. Sorption mechanism

Fourier transform infrared spectroscopy (FTIR) was used to investigate the sorption mechanisms of the studied metal ions onto HA. For FTIR experiments, 1 g mass of finely powdered HA placed in 250 cm^3 beaker, 100 cm^3 of 0.025 M metal salt solution was added and adjusted to pH 5.0. The suspension was magnetically stirred at 333 K for half an hour, and then left for 24 h to saturate with the metal ion. Resulting suspensions of HA + M complexes were filtered and rinsed with distilled water. Finally the solid complexes were dried at 353 K for 4 h. The dry complexes were mixed with 50 mg KBr and compressed to obtain a pellet. The pellets were dried at 378 K for 2 days and then, analyzed using a MIDAC 1700 model FTIR spectrometer. The base line was corrected and scanning was performed from 4000 to 400 cm^{-1}.

3. Results and discussion

3.1. Effect of EB irradiation on HA molecules

Humic acid (HA) substances effectively absorb non-ionizing radiation, such as ultraviolet (UV), visible or infrared radiation. Much less is known about their sensitivity to ionizing radiation, such as EB, γ- or X-rays. The UV absorption spectra of EB irradiated HA are illustrated in Fig. 1. As shown from the figure, that the humic acid solution at pH 5.0 has a maximum absorbance in the UV region at 320 nm [22]. This absorption belongs to the aromatic rings of HA and a considerable decrease in the absorbance of peak occurred with the increase in the EB irradiation dose. The observed decrease in absorption (Fig. 1) is attributed to degradation and precipitation of HA molecules.

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