

Article

Study on the Leaching of Mercuric Oxide with Thiosulfate Solutions

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Abstract: Mercury is receiving more concern due to its high mobility and high toxicity to human health and the environment. Restrictive legislations and world-wide efforts have been made on mercury control, especially the release and disposal of mercury-contaminated wastes. This paper describes a novel technology for detoxifying mercury-containing solid wastes with thiosulfate salts. Various parameters which may potentially influence mercury extraction from mercuric oxide with the thiosulfate leaching system—including reagent concentration, solution pH, and temperature—have been examined. The experimental results show that virtually all mercuric oxide can rapidly dissolve into the thiosulfate leaching system under optimized experimental conditions, indicating that thiosulfate is an effective lixivant for recovering mercury from mercury-containing solid waste.

Keywords: mercury; mercuric oxide; thiosulfate; leaching; detoxification

1. Introduction

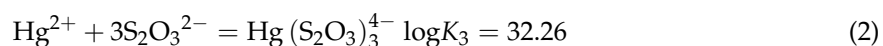
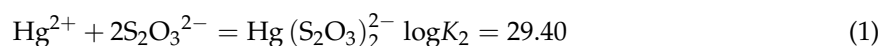
Mercury has attracted great public concern due to its high hazards and adverse effects to both human health and the environment. Toxicology studies have proven that mercury—both inorganic and organic—can cause serious health effects, though organic mercury is generally more toxic [1–4]. However, mercury is highly mobile, and mercury contamination can become much more widespread than that of other heavy metals [5,6]. Mercury may undergo complex physical, chemical, and biological transformations in the environment—e.g., the atmospheric transport of Hg^0 , the photochemical oxidation and subsequent deposition of mercury on water and land, and the methylation of Hg^{2+} by reducing bacteria in anoxic habitats and its absorption and accumulation by organisms, which results in high mercury concentration in fish and chronic low level exposure to humans through the food chain [7,8]. More than two thousand people were affected and 1043 died due to Minamata Disease caused by mercury pollution from a local chemical plant in Japan in the late 1970s [9].

Mercury emission may come from natural, anthropogenic, and reemitted resources. Typical natural mercury emissions include volcanic emission, forest fires, and degassing from mercury mineral deposits. Re-emitted mercury resources, such as those mercury-contaminated sites or systems, are also important in the global mercury emission cycle [10]. Anthropogenic mercury emission mainly results from coal and oil combustion, urban discharge (municipal and medical wastes), and the mercury mining/mineral processing industry [11]. The extensive exploration of cinnabar ore in the Wanshan area (Guizhou, China) has resulted in severe mercury pollution to the local aquatic system and land [12]. In artisanal gold mining, mercury is used to extract gold from ore, which always results in severe mercury releases to the local environment (surrounding rocks, soils, and sediments) [13,14]. It has been reported that ionic forms of mercury are strongly adsorbed by soils and sediments and are desorbed slowly. Most mercury ions are absorbed by organic matter (mainly fulvic and humic acids) in acidic soils. As a result, the following mercury compounds are commonly found in mercury-contaminated

solid wastes: Hg_2Cl_2 , $\text{HgO}/\text{Hg}(\text{OH})_2$, HgS , and methyl-mercury compounds such as CH_3HgCl and CH_3HgOH [15].

To detoxify mercury-containing wastes, technologies such as amalgamation, adsorption, precipitation (sulfides/selenides), thermal treatment, and stabilization/solidification through vitrification or chemical formation have been developed [16]. For example, elemental mercury can also react with elemental sulfur to form mercury sulfide (HgS), which is kinetically stable at room temperature [15]. A more stable and insoluble compound is HgSe , also known as tiemannite. The technologies and processes using HgS or HgSe for mercury stabilization are commonly used in HgO -containing wastes and solid wastes containing a large amount of mercury [17]. In adsorption treatments, adsorbents (mainly activated carbons) are used to stabilize Hg^0 [18]. Thermal desorption, retorting, and roasting—which all employ heating (at low or high temperatures)—have been commonly used to treat mercury contaminated soil, sediments, and other solid wastes [19–21]. For stabilization/solidification processes, the vitrification process is a high-temperature treatment technology designed to immobilize contaminants by incorporating them into a vitrified end product [22–25]. For solid mercury-contaminated wastes, water-based technologies (or leaching technologies) such as soil washing that use both physical and chemical separation methods to reduce contaminant concentrations in wastes have also been commonly used [26]. These processes can be used independently or in conjunction with other treatment technologies. Chemicals such as HCl , HNO_3 , and H_2O_2 have been used as lixivants for the remediation of mercury-contaminated sediments [27]. Thiosulfate salts are also suggested as the leaching reagent for the extraction of mercury from mercury-containing soil. Issaro et al. [28] reported that $50\% \pm 5\%$ of mercury can be extracted from contaminated soil after leaching with $0.01 \text{ M Na}_2\text{S}_2\text{O}_3$ for about 24 h. Lu et al. [29] examined the effect of thiosulfate on mercury removal, and reported that $\text{Na}_2\text{S}_2\text{O}_3$ can be used as a chemical aid for improving trace mercury removal. However, since mercury may be present in various forms in contaminated wastes, detailed information on leaching behavior and mechanisms of different mercury components in the thiosulfate leaching system is lacking. As one of the typical inorganic mercuric compounds, mercuric oxide was chosen for research in this paper. The leaching behavior of HgO in thiosulfate solutions under different experimental conditions (temperature, thiosulfate concentration, liquid–solid ratio, and initial pH) was examined, and the potential use of sodium thiosulfate for the remediation of mercury-contaminated wastes is discussed based on experimental results.

Elemental mercury is virtually insoluble in water ($50 \mu\text{g}/\text{L}$ at 298 K), while mercuric oxide (in the form of Montroydite in nature)—a kind of alkaline oxide—has a solubility of $15.8 \text{ mg}/\text{L}$ in water at 298 K , and has good solubility in diluted hydrochloric acid, dilute nitric acid, and alkaline cyanide. The aqueous speciation and coordination of Hg have been well-documented [30–36]. The oxidation states of Hg in aqueous systems are 0, +1, and +2, of which $\text{Hg} (2+)$ is the most stable in typical aerated water. Mercury speciation and coordination in the aquatic system without the presence of other strongly complex ligands is mainly controlled by hydrolysis reactions. The Eh–pH diagram of the Hg – H_2O system at room temperature has been calculated by many researchers [37]. Basically, at low pH, Hg^{2+} is octahedrally coordinated by six water molecules, forming the hex-aquaion $\text{Hg}(\text{H}_2\text{O})_6^{2+}$, with Hg – O bond lengths of 2.34 – 2.41 \AA [33]. As the pH increases, Hg^{2+} hydrolyzes to form HgOH^+ and $\text{Hg}(\text{OH})_2$, of which two Hg – O bonds are shortened to distances of 2.00 – 2.10 \AA , while the remaining bonds are lengthened to approximately 2.50 \AA . The distorted octahedral coordination is indicative of the tendency for $\text{Hg} (2+)$ to form mononuclear linear, double-coordinated complexes. When sulfur species are present, mercury sulfide (HgS) is stable in a wide range of pH at low potentials. At high potentials, sulfur will occur as more stable species, such as HSO_4^- (low pH) and SO_4^{2-} (high pH) [30,38]. If sulfur occurs as non-stable species, such as in the form of thiosulfate, complex reactions between Hg^{2+} and $\text{S}_2\text{O}_3^{2-}$ may happen in aqueous solution, and the complexing reactions can be represented as follows [39]:



the complexes can only form conditionally, depending on the solution potential, pH, temperature, and concentrations of relevant species.

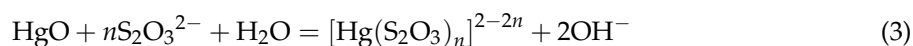
2. Experimental

Reagent grade mercuric oxide (particle size: P80–16 μm), sodium thiosulfate, sulfuric acid, and sodium hydroxide were used as received from local suppliers. De-ionized water was used in all experiments. Leaching tests were carried out in a sealed beaker, and mixing was provided by a mechanical agitator with glass impellers. Temperature was controlled with a water bath. Equilibrium pH was adjusted by direct addition of concentrated H_2SO_4 solution (50% *v/v*) or NaOH solution (2 mol/L). The solution pH and potential were measured with a glass pH probe during leaching. For each test, a designated amount of mercuric oxide was leached in 500 mL solution, and 10 mL of slurry were taken out for sampling at different time intervals. The slurry was filtered through a conical separator with filter paper. The mercury content in the aqueous sample was analyzed by flame atomic absorption spectrophotometer.

3. Results and Discussion

3.1. Effect of pH

Tests of leaching HgO in $\text{Na}_2\text{S}_2\text{O}_3$ solutions under different initial pH were first conducted at indoor temperature (20 ± 1 °C). In these tests, 0.4 g HgO was leached in 500 mL aqueous solution with 0.01 molar $\text{Na}_2\text{S}_2\text{O}_3$. Variations of solution pH and mercury extraction at different leaching times are shown in Figures 1 and 2, respectively. In all three cases, the solution pH increases from the initial pH to around 11.5, and then keeps nearly constant after that. These results are consistent with reports of the dissolution chemistry of mercury species in thiosulfate solutions in the literature [33]. The reaction of HgO and $\text{Na}_2\text{S}_2\text{O}_3$ in aqueous solution can be represented as follows:

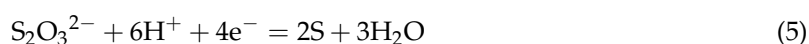


where n may be 2 or 3, depending on the $\text{Na}_2\text{S}_2\text{O}_3$ concentration. The Gibbs values of Equation (3) with ligand numbers of 2 and 3 were also calculated, and the results were -22.05 kJ/mol and -36.32 kJ/mol, respectively, indicating that the complex reactions are spontaneous. The dissolution of HgO in the $\text{Na}_2\text{S}_2\text{O}_3$ solution results in the release of OH^- ions, which leads to an increase of pH when leaching begins until equilibrium is established (about 5–10 min under the experimental conditions). Accordingly, the dissolution of HgO results in increasing Hg concentration in the solution with increasing leaching time. It was also observed that for different initial pH, higher Hg extraction is obtained with initial pH 9.67 than those of initial pH 7.00 and initial pH 10.96 at the condition of $\text{Na}_2\text{S}_2\text{O}_3$ concentration 0.01 M. This phenomenon is probably related to the stability of thiosulfate. The potential decomposition reactions at low and high pH are expressed below:

At alkaline conditions,



At acidic conditions,



According to Reactions (1) and (2), to dissolve 0.4 g HgO entirely, at least 5.5×10^{-3} mole $\text{Na}_2\text{S}_2\text{O}_3$ will be needed to form the thiosulfate complex, theoretically. In other words, the total amount of $\text{Na}_2\text{S}_2\text{O}_3$ (0.01 M) used in these tests is merely enough for complex reactions. As a result, the mercury extraction under the defined experimental conditions is sensitive to the change of initial pH in this case. Further tests indicate that when a high concentration of $\text{Na}_2\text{S}_2\text{O}_3$ (0.1 M) is used for leaching, the initial solution pH exhibits little effect on the leaching rate.

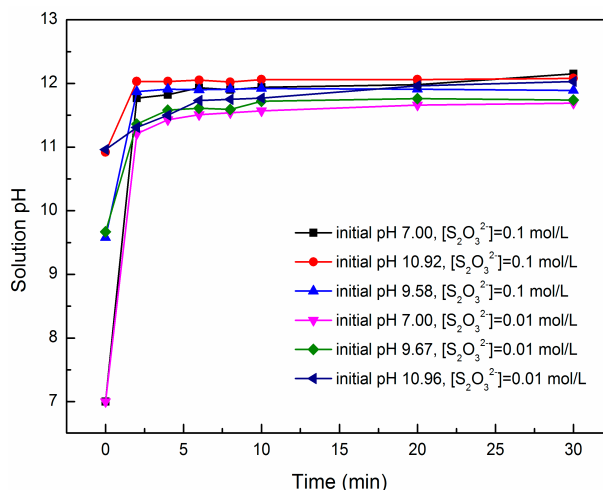


Figure 1. Variations of pH on Hg extraction in 0.01 mol/L and 0.1 mol/L Na₂S₂O₃ solution (Initially, 0.4 g HgO was mixed with 500 mL Na₂S₂O₃ solution, 20 ± 1 °C).

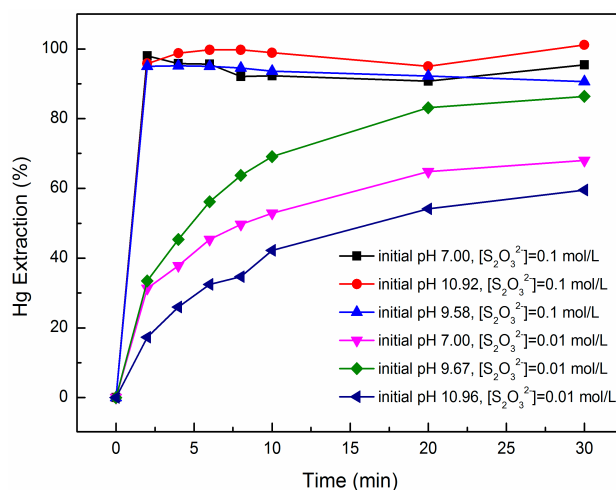


Figure 2. Effect of initial pH on Hg extraction in 0.01 mol/L and 0.1 mol/L Na₂S₂O₃ solution (Initially, 0.4 g HgO was mixed with 500 mL Na₂S₂O₃ solution, 20 ± 1 °C).

3.2. Effect of Na₂S₂O₃ Concentration

To determine the effect of Na₂S₂O₃ on HgO dissolution, tests were carried out with 0.4 g HgO in 500 mL solutions with different concentrations of Na₂S₂O₃. For all tests, the pH was uncontrolled during leaching. Similar to the tests above, the solution was initially around 7.0 and reached constant (around 11.5) after 5 min. Variations of Hg extraction with time are shown in Figure 3. At low Na₂S₂O₃ concentration (0.01 mol/L), the Hg extraction increases relatively slowly with increasing leaching time, and the final Hg extraction is only 70% after leaching for 30 min. At a higher concentration of Na₂S₂O₃ (0.05 mol/L), the dissolution rate of HgO increases significantly—the Hg extraction reaches about 95% after leaching for 5 min and then keeps nearly stable after that. When the concentration of Na₂S₂O₃ in solution further increases to 0.1 mol/L, the HgO dissolution rate is slightly higher than that of 0.05 mol/L Na₂S₂O₃ in solution, and virtually all HgO dissolves within 5 min.

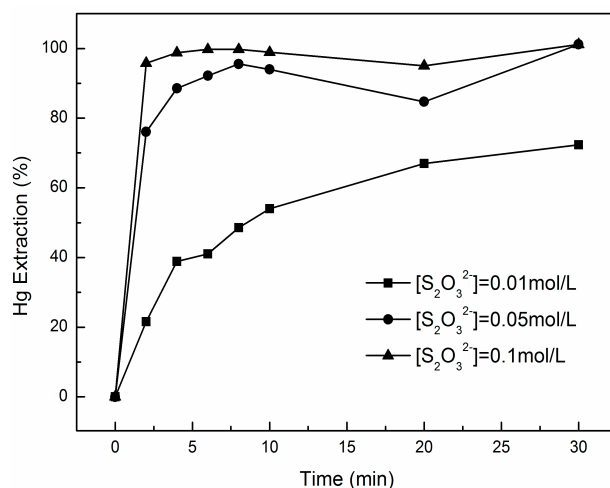


Figure 3. Effect of $\text{Na}_2\text{S}_2\text{O}_3$ concentrations on Hg extraction (Initially, 0.4 g HgO was mixed with 500 mL $\text{Na}_2\text{S}_2\text{O}_3$ solution, 20 ± 1 °C).

3.3. Effect of Temperature

To determine the effect of temperature on HgO leaching with $\text{Na}_2\text{S}_2\text{O}_3$, tests were carried out with 0.4 g HgO in 500 mL 0.01 mol/L $\text{Na}_2\text{S}_2\text{O}_3$ solutions. For all tests, the pH was uncontrolled during leaching. Variations of Hg extraction with time are shown in Figure 4. It shows that the initial leaching rate of HgO increases with an increase of temperature, when temperature varies from 293 K to 323 K. The final Hg extraction after 40 min at the higher temperature (303 K) is close to 95%, much higher than that at 293 K (about 70%). However, the difference of final Hg extractions at 303 K, 313 K, and 323 K is insignificant. Considering the potential effect of the decomposition of $\text{S}_2\text{O}_3^{2-}$ during leaching, a simple estimation of the initial leaching rate of HgO under different temperatures was conducted. Assuming that

$$k_T = \frac{dX_{\text{HgO}}}{dt} = -\frac{d\frac{C_{\text{Hg}}}{C_{\text{Hg-max}}}}{dt} \approx \frac{\Delta\frac{C_{\text{Hg}}}{C_{\text{Hg-max}}}}{\Delta t} \quad (6)$$

where k_T is the initial leaching rate of HgO at temperature T (K); X_{HgO} is the fraction of unreacted HgO; t is leaching time (min); C_{Hg} is the molar concentration of dissolved Hg in leachate (mol/L); and $C_{\text{Hg-max}}$ is the maximum molar concentration of dissolved Hg in leachate (mol/L).

$$k_T = A \exp\left(-\frac{\Delta E}{RT}\right) \quad (7)$$

$$\log k_T = -\frac{\Delta E}{2.303RT} + \log A \quad (8)$$

The Arrhenius equation in terms of the reaction rate constant k_T can be written in Equation (7). Here the ΔE is the apparent activation energy for the leaching Reaction (3), and A is the constant. If the leaching rate in the first two min is used for the calculation, the plot of $\log k_T$ vs. $1000/T$ is shown in Figure 5, and a straight line is obtained when the temperature varies from 293 K to 323 K. The negative value of the line's slope indicates that leaching Reaction (3) is endothermic, and increasing temperature promotes the leaching of HgO with $\text{Na}_2\text{S}_2\text{O}_3$ between 293 K and 323 K. The apparent activation energy can be calculated according to the slope of the line, giving a value of 23.3 kJ/mol, indicating that the initial leaching of HgO in $\text{Na}_2\text{S}_2\text{O}_3$ solution is under mixed control and both the diffusion of the lixivant $\text{S}_2\text{O}_3^{2-}$ and the complexing reaction between mercury and $\text{S}_2\text{O}_3^{2-}$ exhibit effects on the dissolution of HgO.

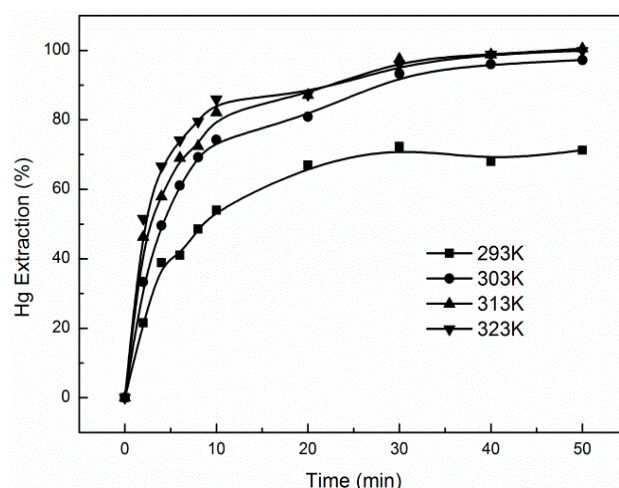


Figure 4. Effect of temperature on Hg extraction with $\text{Na}_2\text{S}_2\text{O}_3$ (Initially, 0.4 g HgO was mixed with 500 mL 0.01 mol/L $\text{Na}_2\text{S}_2\text{O}_3$ solution).

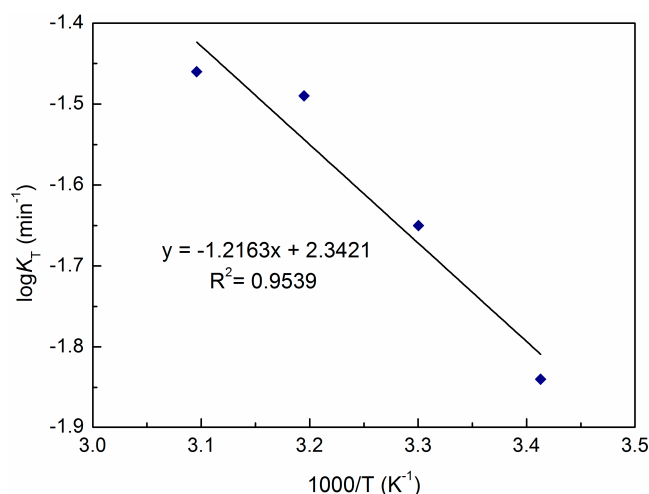


Figure 5. Plot of $\log k_T$ vs. $1000/T$ (Initially, 0.4 g HgO was mixed with 500 mL 0.01 mol/L $\text{Na}_2\text{S}_2\text{O}_3$ solution).

4. Conclusions

The leaching behavior of HgO in $\text{Na}_2\text{S}_2\text{O}_3$ solutions was investigated in the paper. Effects of initial pH, $\text{Na}_2\text{S}_2\text{O}_3$ concentration, and temperature on the dissolution rate of HgO in $\text{Na}_2\text{S}_2\text{O}_3$ solution were examined. The test results indicate that the solution pH exhibits a significant effect on HgO leaching when the molar ratio of $\text{Na}_2\text{S}_2\text{O}_3$ to total mercury is relatively low (e.g., less than 3). An increase of $\text{Na}_2\text{S}_2\text{O}_3$ concentration may potentially increase the dissolution of HgO due to the increase of the molar ratio of $\text{Na}_2\text{S}_2\text{O}_3$ to mercury. Increasing temperature can slightly increase the dissolution rate of HgO in $\text{Na}_2\text{S}_2\text{O}_3$ solutions when the temperature varies from 293 K to 323 K. The apparent activation energy can be calculated according to the slope of the line, giving a value of 23.3 kJ/mol, and the initial leaching of HgO in $\text{Na}_2\text{S}_2\text{O}_3$ solution is under mixed control. Under optimum experimental conditions, virtually all mercuric oxide can dissolve in $\text{Na}_2\text{S}_2\text{O}_3$ solutions within five minutes, indicating that the thiosulfate leaching system is an effective way to detoxify mercury solid wastes containing mercuric oxide.

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Author Contributions: Chao Han wrote the original manuscript and helped with leaching tests and data collection. Feng Xie supervised experimental work and data analysis. Feng Xie and Wei Wang revised the manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

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