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Release behavior of uranium in uranium mill tailings under environmental conditions

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ABSTRACT

Uranium contamination is observed in sedimentary geochemical environments, but the geochemical and mineralogical processes that control uranium release from sediment are not fully appreciated. Identification of how sediments and water influence the release and migration of uranium is critical to improve the prevention of uranium contamination in soil and groundwater. To understand the process of uranium release and migration from uranium mill tailings under water chemistry conditions, uranium mill tailing samples from northwest China were investigated with batch leaching experiments. Results showed that water played an important role in uranium release from the tailing minerals. The uranium release was clearly influenced by contact time, liquid-solid ratio, particle size, and pH under water chemistry conditions. Longer contact time, higher liquid content, and extreme pH were all not conducive to the stabilization of uranium and accelerated the uranium release from the tailing mineral to the solution. The values of pH were found to significantly influence the extent and mechanisms of uranium release from minerals to water. Uranium release was monitored by a number of interactive processes, including dissolution of uranium-bearing minerals, uranium desorption from mineral surfaces, and formation of aqueous uranium complexes. Considering the impact of contact time, liquid-solid ratio, particle size, and pH on uranium release from uranium mill tailings, reducing the water content, decreasing the porosity of tailing dumps and controlling the pH of tailings were the key factors for prevention and management of environmental pollution in areas near uranium mines.

1. Introduction

With the rapid development of nuclear power plants worldwide, the demand for uranium is increasing. As the first step of the nuclear fuel cycle, uranium extraction has generated about one billion tons of uranium mill tailings in 4000 mines worldwide (Abdelouas, 2006; Sethy et al., 2011). As of 2010, there were about 6×10^7 tons of uranium-bearing tailings in China (Zhang et al., 2010). As a general practice, these wastes are stock-piled in the environment (Haque and Ritcey, 1983). The disposal of uranium mill tailings has been considered to pose a significant risk to the environment due to the leachable and mobile toxic metals in weathering environments.

Uranium is a naturally occurring radionuclide and heavy metal having been present in the Earth's crust (current average 2.7 ppm) since its formation. Due to both natural and industrial activities, elevated uranium concentrations can be found in environmental matrices. Although most of the uranium is extracted from ores, a certain content of uranium is still left in uranium mill tailings. To better assess the toxicity and environmental impact of uranium, more information concerning its mobility and availability in uranium mill tailings is necessary.

Concentrations of remaining uranium in tailings is always higher than the earth's crust (Othmane et al., 2013). However, it does not necessarily mean the wastes are toxic or metals are released into the environment. The behavior and availability of uranium in uranium mill tailings is dependent on the physicochemical form of uranium and the characteristics of the tailings (Othmane et al., 2013). When uranium mill tailings are exposed to

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atmospheric oxygen and moisture in the tailing pond, the release of uranium is also influenced by the water in contact with the material (Al-Hashimi et al., 1996). The potential release of toxic heavy metals from uranium mill tailings to the surface and ground water are of particular concern (Hu et al., 2016; Sharma et al., 2016; Tuovinen et al., 2016; Yan et al., 2016). There are various interactions that are bound to control the mobilization of uranium in the solution such as dissolution, complex formation, and desorption-sorption at the water-rock interface (Tricca et al., 2000). The concentration and distribution of uranium in tailings determine the rates and amounts of uranium released into solution during leaching (Štrok and Smodiš, 2013). In addition, the redox conditions, pH, and ligands like carbonate and phosphate in water, are all factors that could affect the uranium mobility in the surface and subsurface environment (Fox et al., 2012; Liu et al., 2009; Othmane et al., 2013). However, few systematic studies have been reported on how contact time, liquid-solid ratio, particle size, and pH impact upon uranium release to water, from granite-type uranium mill tailings. The mechanism of uranium release under a wide range of water chemistry conditions is still unclear. Therefore, the impact of uranium mill tailings under certain water conditions should be carefully considered.

Without appropriate treatment of the uranium mill tailings, uranium released from uranium mill tailings poses a potential risk of environmental contamination. Therefore, it is important to understand the release characteristics of uranium, for proper waste management of uranium mill tailings. This paper focuses primarily on the release behavior of uranium from uranium mill tailings and the controlling release mechanisms. Batch experiments considering the effects of contact time, liquid-solid ratio, particle size, and pH were utilized to shed light on the release process of uranium in aqueous solution. This study will be of positive significance in the prevention and control of environmental pollution in uranium mine areas.

2. Experiment and method

2.1. Sampling site

The tailing samples were taken from a tailing impoundment affiliated to a uranium mining and metallurgy plant in the northwest China. Uranium mill tailings containing waste rocks, low-grade uranium ore and residues after uranium extraction were discharged into the impoundment. They were brown-colored solids containing uranium and other heavy metal ions, and were neutralized with lime. The tailing impoundment is surrounded with artificial dams (Fig. 1). The distance from the pond to the nearest city is only 6 km. It is also situated near a densely populated area. The study area has a subtropical humid continental climate. The average annual temperature is 9.5 °C with an extreme low temperature of −7.2 °C and an extreme high temperature of 38.3 °C. The average annual rainfall in this area is about 300 mm from May to September. Tailings dumps are reactive environments that evolved with time through a large variety of physical and chemical processes. Similarly to other tailing sites in the early decades of uranium mining, no engineered cover or bottom liner was used to effectively contain the tailings. Since the tailing dumps have no protective soil cover and because of the large pore size among the tailing particles, the residual uranium in the tailings is directly washed out during rainfall. The elutropic uranium from the tailing dumps transports to subsurface aquifers and groundwater, which is subsequently used for drinking water and irrigation purposes in the locality.

2.2. Sample collection and preparation

Tailing samples from the acidic leaching of granite-type uranium ores were collected from the tailing impoundment. Samples were collected from the tailings located at the sub-surface horizon with a depth of 20 cm from the surface. Collected tailing samples were packed into a water-tight bag. In the laboratory, the tailing samples were allowed to dry at room temperature for 2 weeks. In some cases, the dried uranium mill tailings were passed through a series of sieves in order to determine their fractional size distribution. In other cases, the dried uranium mill tailings were ground, homogenized, and sieved through a 74 μm mesh for further processing.

2.3. Sequential extraction method

The sequential extraction procedure is a method for chemical separation of the major mineral components (Schultz et al., 1998; Sims et al., 2008; Tessier et al., 1979). It is complicated and lengthy. Yet, it provides key information towards understanding

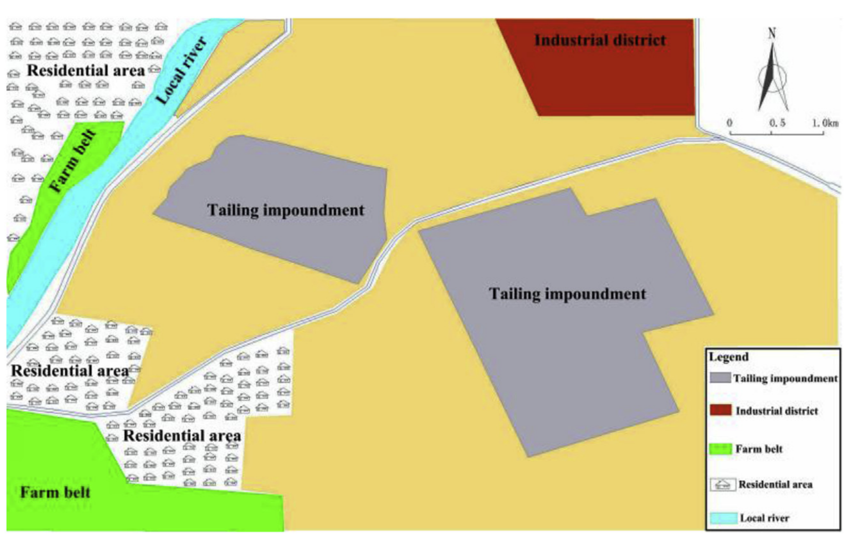

Fig. 1. Location map of uranium mill tailing impoundment.