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Original Article

INVESTIGATION OF ACTIVATED CARBON ADSORBENT ELECTRODE FOR ELECTROSORPTION-BASED URANIUM EXTRACTION FROM SEAWATER

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ABSTRACT

To support the use of nuclear power as a sustainable electric energy generating technology, long-term supply of uranium is very important. The objective of this research is to investigate the use of new adsorbent material for cost effective uranium extraction from seawater. An activated carbon-based adsorbent material is developed and tested through an electrosorption technique in this research. Adsorption of uranium from seawater by activated carbon electrodes was investigated through electrosorption experiments up to 300 minutes by changing positive potentials from +0.2V to +0.8V (vs. Ag/AgCl). Uranium adsorption by the activated carbon electrode developed in this research reached up to 3.4 g-U/kg-adsorbent material, which is comparable with the performance of amidoxime-based adsorbent materials. Electrosorption of uranium ions from seawater was found to be most favorable at +0.4V (vs. Ag/AgCl). The cost of chemicals and materials in the present research was compared with that of the amidoxime-based approach as part of the engineering feasibility examination.

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1. Introduction

Under the concern over greenhouse gas emissions, nuclear energy is considered important to meet future energy demands. Projection of world nuclear generating capacity until 2035 shows that it will constantly increase to between 44% and 99% from the present value for low and high energy

demand scenarios, respectively. Nevertheless, the future of nuclear energy depends on the sustainability of fuel supply. Unless breeder reactor technology is widely utilized, the future of nuclear energy is limited by the availability of uranium. According to the OECD (Organization of Economic Cooperation and Development) Nuclear Energy Agency and IAEA (International Atomic Energy Agency), the total

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confirmed uranium reserve was estimated at 7 million tonnes, with an additional 10 million tonnes for speculated uranium resources [1]. Based on the current uranium consumption rate of 0.063 tonnes/y, the uranium supply is expected to last for about 80 years, and up to 250 years if the speculative uranium resources are taken into account. Additional uranium resources are available from unconventional uranium sources, such as phosphate rock, but the resources pool is not well understood [2], and its contribution is not expected to be significant enough to meet the long-term future demand. Uranium is also known to exist in seawater. Although the uranium in seawater exists at a very dilute concentration of 3.3 ppb, the total inventory of uranium in the entire ocean is greater than the terrestrial uranium inventory: the theoretical amount of uranium resource in seawater is about 4.5×10^9 tonnes (U ~3.3 ppb, ocean volume $\sim 1.37 \times 10^9$ km³), which is roughly 1000 times greater than the estimated conventional uranium reserves [3]. The uranium in seawater primarily exists as uranyl tricarbonate complex $[\text{UO}_2(\text{CO}_3)_3]^{4-}$ [4] as shown in Fig. 1, with the presence of other ions at higher concentrations. The typical elements present in this complex in normal seawater are shown in Table 1 [5,6]. Typically, the linear uranyl group are surrounded by six oxygen atoms of the three bidentate carbonate groups. The radius of the oxygen ions at equatorial plane amounts to 4.85 Å, making it the largest ion present in seawater [6].

The idea of recovering uranium from seawater was first introduced in 1960 [7]. Studies on uranium recovery from seawater primarily focused on the development of high efficiency adsorbent materials that were capable of extracting uranium in the presence of other ions. Various adsorbent materials suggested include synthetic polymers [8–13], inorganic materials [14–19], biopolymers [20–23], and carbon-based materials [13,24–34]. Among them the amidoxime-based adsorbent is the current state-of-the-art material, with its affinity in chelating uranyl ions in seawater [35]. However, the cost of extracting seawater uranium using amidoxime-based adsorbent materials is still very high

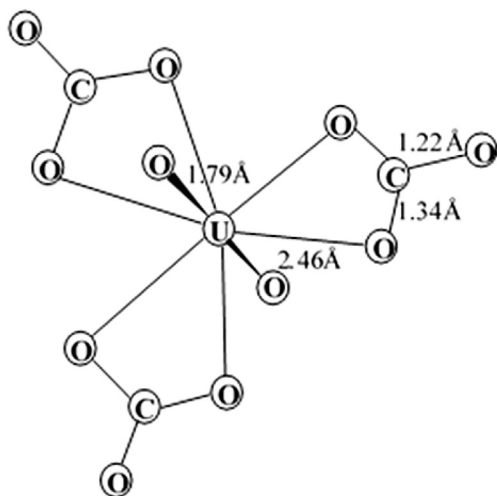


Fig. 1 – Uranyl tricarbonated complex structure $[\text{UO}_2(\text{CO}_3)_3]^{4-}$. Reproduced from Ref. [6].

Table 1 – Typical elements in seawater [5,6].

| Elements | Cations (ppm) | | Anions (ppm) | |
|--------------------|-------------------|--|--|-------|
| Major elements | Na ⁺ | 10770 | Cl ⁻ | 19500 |
| | Mg ²⁺ | 1290 | SO ₄ ²⁻ | 2650 |
| | Ca ²⁺ | 412 | HCO ₃ ⁻ | 140 |
| | K ⁺ | 380 | Br ⁻ | 65 |
| | Str ²⁺ | 8 | F ⁻ | 1.3 |
| Minor elements | | | H ₃ BO ₃ | 260 |
| | Li ⁺ | 0.18 | I ⁻ | 0.06 |
| | Rb ⁺ | 0.12 | MoO ₄ ²⁻ (as Mo) | 0.01 |
| | Cs ⁺ | 0.0004 | VO ₂ (OH) ₃ ²⁻ (as V) | 0.002 |
| | Ba ²⁺ | 0.02 | PO ₄ ³⁻ | 0.07 |
| | Zn ²⁺ | 0.0005 | | |
| | Fe ³⁺ | 0.01 | | |
| | Cu ²⁺ | 0.003 | | |
| | Mn ²⁺ | 0.002 | | |
| | In ³⁺ | 0.02 | | |
| Ge ⁴⁺ | 0.00007 | | | |
| U ⁶⁺ as | | [UO ₂ (CO ₃) ₃] ⁴⁻ | 0.33 | |

compared to that of the conventional uranium mining method [30]. This is mainly due to poor adsorbent performance and high production cost. Recently, use of porous carbon materials (e.g., activated carbon and mesoporous carbon) received attention mainly due to their high efficiency in adsorbing uranium. Porous carbon materials usually have a very large internal surface area, large pore volume, tunable pore size, and are easy to fabricate [31].

In recent studies, use of porous carbon materials combined with the electrosorption technique was shown to have high selectivity for uranium separation. For instance, uranium capture by activated carbon fibers from a 50-hour cumulative adsorption experiment was reported to reach 600 g uranium/kg activated carbon fibers [32]. This is significantly greater than the reported capture efficiency of amidoxime-based adsorbent materials at 2 g uranium/kg adsorbent material [30]. In addition, oxime-functionalized mesoporous carbon materials were reported to have high selectivity for the uranium ion in the presence of competing ions [33]. Similar behavior was also reported for benzoylthiourea-grafted activated carbon [34]. While these interesting performances were noted for carbon materials under different conditions, the ability of carbon materials in seawater has never been tested [36].

The objective of this research is to investigate the performance of porous carbon materials for electrosorption-based separation of uranium from seawater. Electrosorption is a potential-induced sorption technique based on applied voltage to force charged species (ions) to move towards oppositely charged electrodes. The mechanism behind electrosorption is based on electrical double layer formation, with the formation of a region that increased the accumulation of counterions, while coions are driven away from the electrode [37]. The selectivity of ions in electrosorption depends on the surface properties of the electrode and properties of the ion [38]. Both the size of the ion and the pore size of the electrode will contribute to the selectivity process. Since uranium is one of the largest ions in seawater [6], the electrosorption approach is expected to increase the selectivity of uranium ions to be adsorbed by the activated carbon electrode.

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