

Treatment of spent nuclear fuel debris contaminated water in the Taiwan Research Reactor spent fuel pool

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ABSTRACT

The cleanup of Taiwan Research Reactor (TRR) spent fuel pool is a significant milestone in the decommissioning project of TRR. Due to the water was polluted by the ruptured spent nuclear fuel rods, the fission products and actinides were required to be removed from the water prior to transport the water to the radioactive liquid wastes treatment plant. The inorganic water purification media such as aluminosilicate and polyferric sulfate (PFS) were employed as adsorbent and coagulant with respect to their radioactive resistance and efficient. Of the available information, both of the applied adsorbent and PFS were first time applied in the treatment of radioactive liquid wastes. The full scale treatment units including adsorption unit and precipitation bag were established commissioned and successful operated in the treatment of the water in TRR spent fuel pool. Accumulative 600 m³ of water in the TRR spent fuel pool contained 95 Ci of radionuclides had been treated by the combination of adsorption and coagulation processes, and had been transported to further the liquid radioactive wastes treatment plant.

1. Introduction

The cleaning of Taiwan Research Reactor (TRR) spent fuel pool is the phase one task for TRR decommissioning project. TRR is a Canada Deuterium Uranium research reactor which uses heavy water as a moderator, light water as a coolant, graphite as a reflector, and natural uranium metal as fuel. Unfortunately, approximately 926 m³ of water was contaminated by the ruptured of some spent nuclear fuel rods. Hence, all the challenge during the cleaning in the TRR spent fuel pool was raised from the event; fission products and actinides were the major radionuclides presented in the water. The previous cooling water purification system of the TRR spent fuel pool was also phase out. Several small ion exchangers were employed to reduce the radioactivity during the removal of spent fuel, equipment and wastes from the pool periods, but the concentration activity of β -emitters gradually increased to 4000 Bq·mL⁻¹ in the water. Both of the suspended solids and sludge contained fuel particles and actinides often retarded the underwater handling and clogged filters. At the Institute of Nuclear Energy Research (INER) in Taiwan, the acceptance criteria for liquid radioactive wastes treatment facilities using evaporation and ion exchange processes are gross $\alpha < 37$ and $\beta < 370$ Bq·mL⁻¹ (Huang et al., 2012). To meet the acceptance criteria pretreatment units should be established and approximately 90% of β -emitters, major contributed by ¹³⁷Cs and ⁹⁰Sr, in the water of TRR spent fuel pool have to be removed

before feeding into the evaporation facility.

During the last decades, considerable efforts have been directed towards the development of improving processes for the removal of ¹³⁷Cs and ⁹⁰Sr from radioactive waste streams. Furthermore, Fukushima nuclear disaster is thus driving more studies and papers to find the restoration ways for the enormous quantities of radioactive contaminated water and soil (Hu et al., 2012; Kozai et al., 2015; Liu et al., 2014; Masson et al., 2015; Onishi, 2014; Parajuli et al., 2013; Tsuji et al., 2014; Yoshihara et al., 2014). The US EPA has approved the following Best Available Technologies (BATs) and Small System Compliance Technologies (SSCTs) for radionuclides removal from surface and ground water: ion exchange, reverse osmosis, lime softening, enhanced coagulation/filtration, greensand filtration, barium sulfate coprecipitation, electrodialysis/electrodialysis reversal, performed hydrous manganese oxide filtration, activated alumina (U.S. EPA, 2005). Wherein, adsorption method for removing radionuclides from a water stream is highly effective and economical process provided the correct adsorbents are applied in environment that favors adsorption. The selection of appropriate adsorbents is important to the successful removal of cesium and strontium from these liquids (Liu et al., 2014; Tu et al., 2015; Yousefi et al., 2015; Zhu et al., 2014). However, applying the ion exchange resins or some organic composite adsorbents to treat liquid radioactive wastes will suffer material radiolysis during long term storage or disposal, and usually require further stabilization (Wang and

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Wan, 2015). The ion-exchange capabilities and radiation resistance of zeolites have attracted considerable attention pertaining to their application in liquid radioactive waste treatment (Dyer et al., 2000; Wang and Peng, 2010; Koshy and Singh, 2016). In the previous work, the effectiveness of diatomite-based ceramic depth filters in the adsorption of ^{137}Cs and ^{90}Sr ions was demonstrated (Huang et al., 2012). However, as employing the diatomite-based ceramic depth filter to treat water in TRR spent fuel pool, the clogged and the lower adsorption capacity are the major problems. In the previous work, a granular calcium-type aluminosilicate had been found that it can efficiently remove ^{137}Cs and ^{90}Sr from water (Lin et al., 2014).

Coagulation/flocculation is a common process in water treatment, especially for the destabilization of colloids suspensions and for the removal of suspended solids (Ojovan and Lee, 2014). Conventional iron based coagulants such as the $\text{Fe}_2(\text{SO}_4)_3$ or the FeCl_3 are widely used chemicals. However, several factors including such as temperature, pH and dosage can influence the treatment performance (Zouboulis et al., 2008). The polyferric sulfate (PFS) is a pre-polymerized Fe(III) chemical, containing a range of hydrolysis and polymeric species and gradually replace the conventional iron based coagulants (Jiang and Graham, 1998; Moussas and Zouboulis, 2009). Based on our available information, there is almost no report indicated that the PFS had been employed to treat the liquid radioactive wastes. Combination or cascaded of adsorption and coagulation process for the treatment of liquid radioactive wastes was widely performed in nuclear industries.

In this study, a granular synthetic aluminosilicate adsorbent was applied to remove the ^{137}Cs and ^{90}Sr , and then the suspended solids were precipitated using PFS coagulants combined with $\text{Ca}(\text{OH})_2$ in the water of TRR spent fuel pool. A specially designed unit which could be assembled and water immersed was used to pack the adsorbent and to adsorb the radionuclides from the water.

2. Experimental procedure and instrument

2.1. Materials

The selected adsorbent is a kind of calcium-rich aluminosilicate powder which is blended with sodium metasilicate and then granulated to a stripe shape product (namely GWC and produced by JMAX innovation Corp., Taiwan). Natural zeolite (NZ) in the form of mordenite with a particle size of 0.8–1.9 mm was obtained from Nitto Funaka Trading Co., Ltd. (Japan). Inorganic adsorbents, DT-30 and CsTreat[®], were purchased from Diversified Technologies Services, Inc. (US) and Selion Technology Inc. (Finland), respectively. The SEM image and physical properties of GWC was shown in Fig. 1 and Table 1, respectively. Before beginning the adsorption process, the adsorbent was

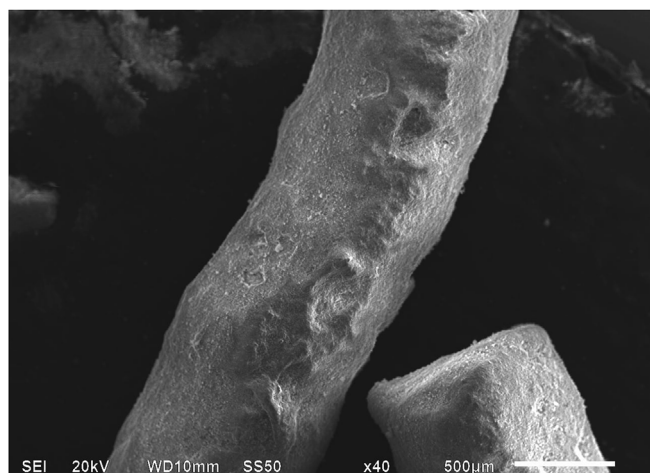


Fig. 1. SEM image of GWC surface.

Table 1
Physical properties of GWC.

Item		GWC
SEM-EDS elements	Na (wt.%)	3.80
	Al (wt.%)	15.89
	Si (wt.%)	68.34
	K (wt.%)	0.69
	Ca (wt.%)	11.27
Particle size (mm)		1.0–1.5
Particle density ($\text{g}\cdot\text{mL}^{-1}$)		1.51
Bed density ^a ($\text{g}\cdot\text{mL}^{-1}$)		0.66
Max. Cs^+ adsorption capacity ^b	($\text{mmole}\cdot\text{g}^{-1}$)	0.34
	($\text{mg}\cdot\text{g}^{-1}$)	45.66
Max. Sr^{2+} adsorption capacity ^b	($\text{mmole}\cdot\text{g}^{-1}$)	0.40
	($\text{mg}\cdot\text{g}^{-1}$)	35.34
Water absorption capacity (wt.%)		45–55
BET Specific surface area ($\text{m}^2\cdot\text{g}^{-1}$)		112
Crystallinity ^c		Poor/amorphous
Color		White to light gray

^a The packed bed density including voids.

^b Obtained from Langmuir isotherm.

^c Observed by XRD pattern.

washed by demineralized water to remove powder. The general formula of the PFS (industrial grade, Jongmaw Chemical Co. Ltd., Taiwan) is $[\text{Fe}_2(\text{OH})_n(\text{SO}_4)_{3-n/2}]_m$ where $n > 2$ and $m = f(n)$. The specifications of the PFS are 1.0–2.0 of pH (1% water solution), 1.2–1.3 $\text{g}\cdot\text{cm}^{-3}$ of density and $> 5\%$ of Fe(III) concentration. The alkali which is applied to neutralize PFS is $\text{Ca}(\text{OH})_2$ (95 wt.%, reagent grade, nacalai tesque, Inc., Japan).

2.2. Instrumentation

The morphology, elemental composition weight percentage and crystallinity of the material were examined by SEM (JEOL JSM-6510, Japan) equipped with an EDS (OXFORD INCAx-act, UK) and XRD (Bruker corp. D8 Advance, USA), respectively. The specific surface area of the material was measured by using the BET method (Quantachrome Instruments NOVA-4200E, USA). The turbidity of water was measured by the turbidity meter (SUNTEX TC-7100) with a detector (TC-500) of ranging from 0 to 500 NTU.

2.3. Radiochemical analysis and instrument

The concentration of U was analyzed using ICP-OES (HORIBA Jobin Yvon, Japan). The activity of ^{137}Cs was measured over a period of 1000 s using a gamma spectrometer with high-purity germanium detector (HPGe, GC4020) coupled to a personal computer analyzer running Genie 2000 software. The HPGe detector was enclosed within CANBERRA 747 shielding a 10-cm-thick layer of lead, coated with 1 mm of tin and 1.6 mm of copper, to reduce the amount of background radiation. The activities of ^{90}Sr , gross α and β were measured using a low-background liquid scintillation analyzer (LSA, Tri-Carb[®] 2910TR, Packard USA), in which ^{90}Sr was characterized using the Cerenkov counting method.

2.4. Adsorption and coagulation processes

2.4.1. Adsorption test

A weighted GWC adsorbent was washed using ultrapure water several times, before being poured into a packed Pyrex column (inner diameter of 2.5 cm and length of 30 cm). The TRR pool water was then fed into the column with a constant volumetric flowrate ($10.27\text{ mL}\cdot\text{min}^{-1}$) using a peristaltic pump (LongerPump 300F) with an adjusted volumetric flow rate, whereupon the permeate water was collected for analyses.

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