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Technetium (Tc)/Rhenium (Re) solubility and leaching behavior from waste forms: An overview

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Technetium-99 (⁹⁹Tc), a radionuclide generated from nuclear industry is a great environmental concern because of its long half-life (2.13×10^5 years) and high mobility in environment. Therefore, apposite management of ⁹⁹Tc is imperative to control its hazardous radiological impact on humans and other livings. So far, the major strategy implementation has been the solidification and immobilization of ⁹⁹Tc radioactive waste in various matrices as waste forms and disposal in deep geological repository. However, by passing the time, ⁹⁹Tc may leach out/solubilize from the waste forms under different geochemical/environmental conditions. In this minireview, we discuss some key contributions towards the solubility of ⁹⁹Tc and rhenium (Re; a well-known surrogate of ⁹⁹Tc) from different waste forms. Specifically, we review the solubility of ⁹⁹Tc from glass, cement, ceramic, and geopolymer waste forms. The final section (conclusion) presents a short summary and future challenges need be addressed to impede the solubility of ⁹⁹Tc from the designed waste forms. We believe this minireview will be beneficial to provide a significant insight on the solubility of ⁹⁹Tc from aforementioned waste forms and in the design of robust matrices to minimize/prevent ⁹⁹Tc migration in various environments.

KEYWORDS

TC, solubility, leaching, waste form, Re

1 Introduction

Radioactive wastes generated from nuclear power plants' operation and decommissioning of nuclear reactors are serious environmental threat due to their long half-lives and impact of extremely hazardous radiations on geosphere as well as humans (Singh et al., 2021). The proper management of these radioactive wastes including high-level waste (HLW), low- and intermediate-level waste (LILW), and low-level waste (LLW) is necessary to control their radiological effect on the environment. Among several radionuclides present in radioactive wastes, ⁹⁹Tc is one of the significant environmental risk contributors due to its long half-life and highly mobile

behavior in oxic environment (Santikari et al., 2022). Therefore, it is essential to control/prevent the mobility of ^{99}Tc into the environment by immobilizing it in suitable and durable waste forms. In several studies, rhenium (Re) has been often used as a preferred non-radioactive surrogate for ^{99}Tc due to their close proximities in terms of speciation, ionic size, and hydration energy (Kim and Kruger, 2018; Duckworth et al., 2021). Since decades, various waste forms including glass, cements, geopolymer, and ceramics have been tested and used for solidification, immobilization, and retention of ^{99}Tc by disposing these waste forms in the deep geological repository (Um, 2018).

Over the time, ^{99}Tc immobilized in different waste forms (glass, cement, geopolymer, ceramics, etc.) under deep geological repository can be released/leached out from the solid matrices and migrate into environment under different environmental and geochemical conditions (Klein et al., 2021). Once released, ^{99}Tc can exist as Tc(VII) in oxic environment (pertechnetate (TcO_4^-); most common chemical species of Tc in the environment) or it can be reduced to Tc(IV) in the presence of electron donors (Meena et al., 2017). In order to prevent and control the solubility and leaching of ^{99}Tc from immobilized solid matrices and to design durable waste forms, it is necessary to understand the solubility behavior of ^{99}Tc from waste forms under different geochemical conditions. In this minireview, we summaries and discuss some of the key studies on solubility of ^{99}Tc and/or Re from the glass, cement, geopolymer, and ceramic waste forms. Additionally, we highlight the future challenges concerning the solubility of ^{99}Tc from waste forms and the design and formulation of durable waste form for efficient ^{99}Tc immobilization and retention capacity.

2 Solubility of Tc and Re from waste forms

The design and formulation of different solid matrices can significantly affect the retention capacity and solubility of ^{99}Tc in the immobilized waste forms. This section presents the selected studies on glass, cement, and other waste forms (geopolymer and ceramics) used for immobilization of ^{99}Tc and/or Re and their leaching/solubility behavior under different experimental conditions.

2.1 Solubility of Tc and Re from glass waste forms

Glass waste form has been preferably applied for immobilization of various radionuclides, including ^{99}Tc globally (Danilov et al., 2021; Donald et al., 1997; Jantzen, 2011). Therefore, the leaching behavior of Tc/Re from glass waste forms under various experimental conditions is

necessary to address the durability of the designed glass matrices and long-term mobility of ^{99}Tc . Bibler and Jurgensen determined the effects of the redox conditions on the release of ^{99}Tc from borosilicate glass (Bibler and Jurgensen, 1987). The authors prepared two batches of glass waste forms under ambient oxidizing and reducing conditions. The leaching tests were performed according to the materials characterization center (MCC-1) test at 90°C and surface area to volume ratio (SA/V) 100 m^{-1} . The obtained leaching test results revealed that ^{99}Tc leached no faster than the glass-forming elements at ambient oxidizing conditions. Additionally, under high pH and reducing conditions, the normalized ^{99}Tc mass loss was observed as 0.02 g/m^2 only, even at significant dissolution of the glass.

A few studies performed the static leaching experiments to examine the leaching behavior of ^{99}Tc in ^{99}Tc doped borosilicate glass in disposal environment (Lemmens & Wang, 1992; Pirllet et al., 2002, 2004). A mixture of clay with synthetic pore water was used as the leachant to simulate the actual interstitial pore water. All tests were conducted in the glove box at SA/V 100 m^{-1} . The authors suggested that ^{99}Tc mass losses enhanced linearly with the time, and was observed to be faster at 90°C than at 40°C . Moreover, ^{99}Tc concentrations were higher in the oxidizing medium than in reducing medium at both 40 and 90°C .

Xu et al. evaluated Re retention and its stability in iron phosphate glass (Xu et al., 2013). The authors observed $\sim 1.1\text{ wt}\%$ retention of Re in iron phosphate glass. Moreover, when 2 wt% or more Re added, a white spherical inclusion ($\text{Na, K}\text{ReO}_4$) was detected by XRD and EDX mapping. The chemical durability tests were performed according to American Society for Testing and Materials (ASTM) C 1285-02. In this method, 1.5 g of glass powder was mixed with 15 ml of deionized water (DIW) in a Teflon vessel and kept inside an oven at 90°C . The normalized Re-released amount after 7-day Product Consistency Test (PCT) was found to be $< 10^{-2}\text{ g/m}^2$. Normalized Re- released along with other elements are shown in Figure 1.

Stefanovsky et al. developed sodium aluminum phosphate (SAP) glass and sodium aluminum iron phosphate (SAIP) glass and determined Re-released rate in both glasses (Stefanovsky et al., 2019). Re- released rate was evaluated *via* PCT test performed at 90°C for 7 days (d). The normalized Re leaching rates were calculated as 2.55×10^{-6} and 9.12×10^{-9} for SAP glass and 3.90×10^{-8} and 7.90×10^{-8} for SIAP glass in the oxidizing and reducing conditions, respectively. Both SAP and SAIP exhibited lower leach rates than the regulatory values ($\sim 10^{-5}\text{ g/cm}^2\text{d}$) specified for HLW glasses. Especially, a sharp decrease in the Re leach rate in SAP glass synthesized under reducing conditions as compared to the same glass produced under oxidizing conditions, was attributed to the enhanced fraction of Re in the reduced form as Re (0).

Pyo et al. developed a new alkali-alumino tellurite glass to incorporate highly volatile Tc/Re (Pyo et al., 2017) and tested leaching behavior of Re *via* PCT method for 7 d at 90°C . The developed tellurite glass incorporated 7 wt% of Re as Re(VII), in

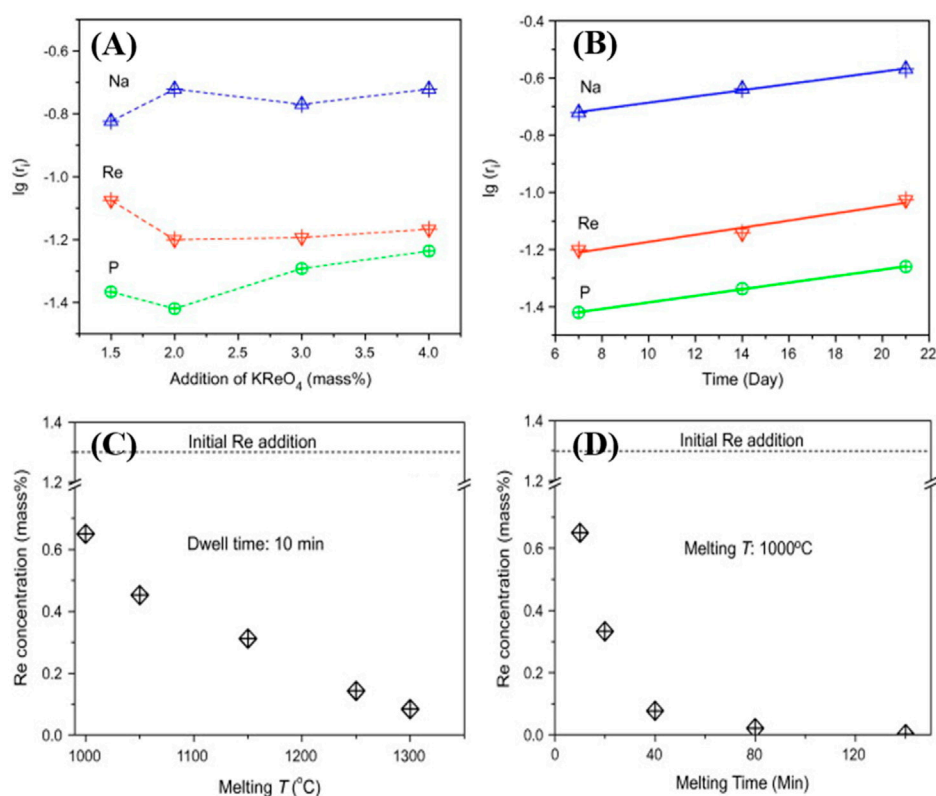


FIGURE 1

Normalized elemental release after 7 days product consistency test (PCT) at different mass% of KReO₄ (A). Elemental release at 2-mass% of KReO₄ vs. PCT duration (B). Concentrations of Re in 2-mass% of KReO₄ samples at different melting temperatures (C) and times (D). Reproduced with permission from the reference Xu et al., 2013. Copyright from Elsevier 2013.

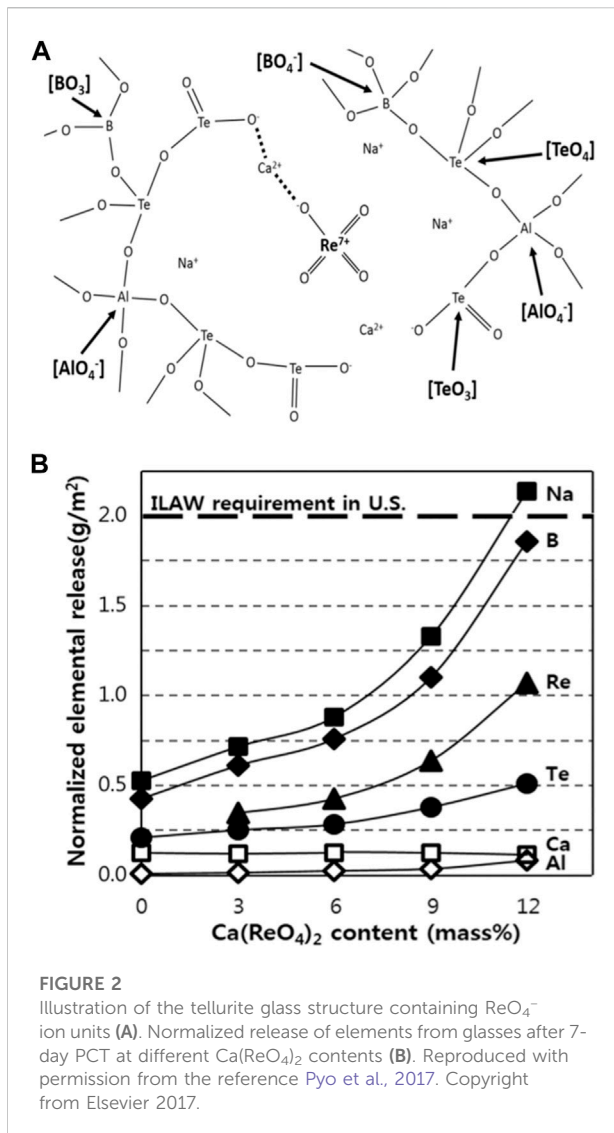
which ReO_4^- tetrahedra were linked to the glass network by bonding between non-bridging oxygens and Na^+ or Ca^{2+} ions (Figure 2A). The normalized elemental release data for Re and Te were observed as $\sim 1 \text{ g/m}^2$ and 0.5 g/m^2 , respectively, which is lower than the safety standard ($<2 \text{ g/m}^2$) for the immobilized low activity waste requirement (Figure 2B).

2.2 Solubility of Tc and Re from cementitious waste forms

Cement is one of the known matrices used for efficient immobilization of various radioactive wastes due to its durable properties; chemical and thermal stability as well as corrosion resistivity (El-Kamash et al., 2006; Faiz et al., 2017; Goo et al., 2021). In this section, we discuss the solubility/leaching of Tc/Re from cementitious waste forms in different testing conditions. Simner et al. reported a Dynamic Leaching Method (DLM) to investigate ^{99}Tc leaching behavior from the saltstone monolith (cementitious material) samples at the Savannah River Site (SRS), in the United States of America (USA) (Simner et al., 2017). In

order to simulate the transport of groundwater through saltstone, DLM applied a flexible-wall permeameter for ^{99}Tc leaching under the elevated hydraulic gradient. This method enabled a continuous flow of permeant through the monolith and analysis of the leachate to determine ^{99}Tc leaching as a function of pore volumes exchanged within the saltstone monolith. Approximately six pore volumes were passed through the monolith. ^{99}Tc concentrations in the leachate were found to be $\sim 1 \times 10^{-8} \text{ mol/L}$, which indicated that ^{99}Tc leaching in saltstone was controlled by the solubility of $\text{TcO}_2 \cdot x\text{H}_2\text{O}$ compounds.

Santikari et al. investigated the leaching and transport of ^{99}Tc from cementitious waste forms in field lysimeters (Santikari et al., 2022). The authors tested two cementitious waste forms; a) slag-grout of 45%:45%:10% mixture of fly ash, blast furnace slag, and cement, respectively, and b) cement of 55%:45% mixture of cement and fly ash, respectively. The study used a duplicate source of each waste form buried in four lysimeters for approximately 10 months to compare the leaching behavior of ^{99}Tc under natural meteorological conditions in South Carolina, United States. Cumulative ^{99}Tc activity in the effluent was low



(2×10^5 Bq) until ~ 300 ml of effluent produced from each lysimeter, and then it was enhanced at a rate of $\sim 3,000$ Bq/mL for all the lysimeters.

Cantrell et al. studied the solubility control of Tc from saltstone by $\text{TcO}_2 \cdot x\text{H}_2\text{O}$ and revealed that the Tc-release in water from saltstone under anoxic conditions can be considerably altered by the solubility of a $\text{TcO}_2 \cdot x\text{H}_2\text{O}$ phase (Cantrell et al., 2013). The authors prepared three reactors and a control with different saltstone sample compositions. All three reactors were composed of 45%, 45%, and 10% of blast furnace slag (BFS), fly ash (FA), and Portland cement (PC), respectively. However, ^{99}Tc content was altered in reactor first ($0 \mu\text{Ci}/\text{kg}$), and kept similar in the reactors second and third ($170 \mu\text{Ci}/\text{kg}$). The obtained data suggested that the solubility of $\text{TcO}_2 \cdot 1.6\text{H}_2\text{O}$ acquired an equilibrium within 2 weeks with $\sim 1.5 \times 10^{-6} \text{M}$ ^{99}Tc concentration. The starting redox potential (E_h)

value was slightly below -100 mV, which lowered considerably between 0–14 d and almost stabilized below -400 mV by 20 d. The authors proposed that the concentrations of ^{99}Tc at equilibrium are expected to vary with $\text{TcO}_2 \cdot 1.6\text{H}_2\text{O}$, as the saltstone pore fluid evolved over a time. Moreover, the solubility of $\text{TcO}_2 \cdot x\text{H}_2\text{O}$ can be dropped considerably due to lowered pH of the samples via carbonation over the times in an actual disposal scenario.

2.3 Solubility of Tc and Re from geopolymer and ceramic waste forms

Geopolymers and ceramic matrices have also been largely used for the incorporation and immobilization of various radionuclides. This section presents key contributions on the solubility/release of Tc and Re from geopolymer and ceramic waste forms. Pierce et al. reported an alkali alumino-silicate geopolymer, DuraLith geopolymer to encapsulate ^{99}Tc liquid radioactive waste (Pierce et al., 2010). The secondary waste simulant of the Hanford Tank Waste Treatment and Immobilization Plant was used to prepare the geopolymer monoliths and three methods developed for the U.S. Environmental Protection Agency (EPA) were applied to assess the stabilization of the DuraLith geopolymer. Diffusivity of ^{99}Tc was found to be as $3.76 \times 10^{-12} \text{cm}^2/\text{s}$ and $9.93 \times 10^{-9} \text{cm}^2/\text{s}$, respectively, for 63 and 2 d cumulative leaching time in batch 1 batch 2 geopolymers. In addition, Xu et al. investigated the effect of blast furnace slag grades on fly ash based geopolymer waste forms (Xu et al., 2014). Ground granulated blast furnace slag (GGBFS) was used to improve the disadvantage of the fly ash based geopolymer and was classified into three grades, 80, 100, and 120. The authors prepared geopolymer mixed with different grades of GGBFSs using the Hanford secondary waste (HSW) simulants spiked by Re. The toxicity characteristic leaching procedure (TCLP) was also applied for the leaching test on fly ash geopolymer waste forms after 28 d of curing at room temperature. Based on the results, the authors suggested that all heavy metals and toxic elements present in the HSW simulant can be more efficiently immobilized by the geopolymer waste forms than Re.

Neeway et al. investigated ^{99}Tc (Re used as surrogate) and I solubility from a sodalite-bearing ceramic waste form using single-pass-flow-through (SPFT) tests (Neeway et al., 2016). The granular fluidized bed steam reforming (FBSR) materials were derived from non-radioactive Hanford low-activity waste (LAW) simulant, radioactive LAW simulant, and the actual radioactive Hanford waste from Tank SX-105. The release rate of Re was determined in the range between $16 \times 10^{-4} \text{g}/\text{m}^2\text{d}$ and $24 \times 10^{-4} \text{g}/\text{m}^2\text{d}$, which is up to 3 times larger than that of the network-forming elements (Na, Al, and Si). Moreover, the release of Re, I, and ^{99}Tc from the FBSR samples exhibited similar behavior. Based on results, the authors presumed that the

solubility of Re and ^{99}Tc was controlled by the mineral phase (sodalite), which was produced during the FBSR process and incorporated Re into the cage structure of the mineral.

Hartmann et al. investigated the crystallographic and hydrodynamic data of ^{99}Tc -based ceramic waste form (Hartmann et al., 2014). Pyrochlore ($\text{Nd}_2\text{Tc}_2\text{O}_7$), perovskite (SrTcO_3), and layered perovskite (Sr_2TcO_4) were synthesized using anhydrous crystalline TcO_2 . Chemical durability of $\text{Nd}_2\text{Tc}_2\text{O}_7$ pyrochlore was measured and compared with ^{99}Tc containing borosilicate glass in close compliance with ASTM C1220. Pyrochlore was insufficiently sintered and did not produce ceramic, however, the specific weight loss of the porous $\text{Nd}_2\text{Tc}_2\text{O}_7$ was determined as $1.48 \times 10^{-7} \text{ g/mm}^2\text{d}$; lower than that of the ^{99}Tc -containing borosilicate glass ($6.43 \times 10^{-7} \text{ g/mm}^2\text{d}$). Moreover, the relative ^{99}Tc -release was evaluated as 0.67% for $\text{Nd}_2\text{Tc}_2\text{O}_7$ pyrochlore, compared to 1.026 for the ^{99}Tc -containing borosilicate glass.

Alekseeva et al. reported the chemical stability (the leaching tests) of the phosphate-based ceramic ($\text{NaRe}_2(\text{PO}_4)_3$) produced by spark plasma sintering (Alekseeva et al., 2021). In this study, the authors used Re as a surrogate of ^{99}Tc . The leaching experiments were conducted for 28 d in distilled water at room temperature under the static mode. The Re-leaching rate was evaluated as $1.3 \times 10^{-5} \text{ g/cm}^2\text{d}$ after 28 d. In another approach, Singh et al. demonstrated the immobilization and leaching behavior of ^{99}Tc from magnesium potassium phosphate (MKP) ceramics using SnCl_2 as a reducing agent (Singh et al., 2006). The designed MKP ceramic waste forms were characterized by XRD and SEM-EDX. Based on results, the authors proposed that ^{99}Tc could be immobilized in a tetravalent state (Tc(IV)) in MKP ceramics because of reducing environment and microencapsulation in the ceramic matrix. The leaching tests were performed using PCT method for 36% elution-loaded waste forms. ^{99}Tc -leaching rate was evaluated between $1.1 \times 10^{-3} \text{ g/m}^2\text{d}$ and $8.5 \times 10^{-3} \text{ g/m}^2\text{d}$ at ambient temperature (25°C), in agreement with ASTM C 1285-94.

3 Conclusion and perspective

In summary, this mini review highlights and summarizes some of the key developments on solubility and release of ^{99}Tc and Re (surrogate of ^{99}Tc) from glass, cement, geopolymer, and ceramic waste forms using different leaching test methods and experimental conditions. In the section two (2) of this mini review, we emphasize on the leaching/release and retention capacity of ^{99}Tc and Re immobilized in these waste forms under various physicochemical testing parameters and geochemical conditions. In section 2.1, we presents solubility and leaching of Re and Tc from glass waste forms. Section 2.2. Discusses the

solubility of Tc and Re from cementitious waste forms, whereas, section 2.3. Describes a few (key) contributions on solubility of Tc and Re from geopolymer and ceramic waste forms. In order to impede the solubility and release of immobilized ^{99}Tc from various waste forms disposed in underground repository and to minimize the mobility of ^{99}Tc in environment/geosphere, future studies and researches (both experimental and theoretical approaches) should be focused on to acquire a wide range of datasets (at different testing conditions) necessary for the design of robust and durable waste forms. This may include more fundamental structural information of candidate waste forms using a various advanced techniques, such as HRTEM, EXAFS, etc. By establishing a comprehensive database for relevant waste forms, it will be easier to design and select most suitable candidates for Re/Tc immobilization. We believe that this mini review will be beneficial to the readers to understand the solubility and release behavior of ^{99}Tc from aforementioned waste forms and to correlate its migration in environment under various geochemical conditions.

Author contributions

BKS, JK, DP, and KK contributed in writing and WU supervised this mini review. All authors discussed, finalized the contents to the article, and approved the submitted version.

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