Prospects of functionalized porous materials towards extraction of spent nuclear fuel radionuclides

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Abstract. Radioactive waste levels have continued to increase due to the growth and development of nuclear energy, industrial or medical radioactive use. In this regard, conventional radioactive waste generated by nuclear power plants cannot be ignored. Effective management of nuclear radioactive waste plays a vital role in alleviating negative impacts on the society and environment. Despite the progress that has been made concerning separation and recycling of spent nuclear fuel through the PUREX process, several gaps in knowledge still exist especially towards the development of a robust separation system based on solid-phase extraction using porous materials. Solid phase extraction is being viewed as one of the most convenient and effective approaches in the removal of cations in radionuclide solutions. This is due to its ability to increase the selectivity and sensitivity of the method as it permits discriminatory binding of analyte to a solid support where the analyte can be collected and thereafter eluted using small quantity of a different solvent. The review covers the current methods used in aqueous nuclear reprocessing, highlights their deficiencies and introduces the potential of applying solid-phase extraction in management of nuclear waste. This study gives the prospects of functionalized porous sorbent materials as solid support in solid-phase extraction of spent nuclear fuel elements.

1. Introduction

Nuclear power continues to stand out as one of the low-carbon energy options amid increased concerns caused by global warming [1]. This however, comes with a significant increase in the production of radioactive waste thereby raising concerns on radioactive waste management particularly the management of spent nuclear fuel (SNF) [1-4]. This is due to their high toxicity which can last for several years in the environment. Spent nuclear fuel discharged from nuclear power generation is estimated to contain about 94% uranium (U), 1% plutonium (Pu), 5% fission products (FPs) as well as a small quantity of minor actinides (MAs) [5]. The high proportion of uranium and plutonium in the SNF makes it attractive for reprocessing [5]. Lanthanides (Lns) forms the bigger proportion of the fission products that is known to be very harmful due to their ability to emit neutron...
particles which normally interferes with nuclear chain reactions over time [4]. The presence of minor actinides in spent nuclear fuel is the main contributor to its long-term radiotoxicity [6].

Several studies have been performed on the separation of lanthanides as well as minor actinides not only to reduce their quantities and toxicity levels, but also to improve the recovery of uranium for recycling in nuclear fuel [7-9]. In general, two major actinides namely U and Pu are usually separated into the solvent phase whereas the other radionuclides are left in the aqueous phase as high active waste/highly active raffinate [10, 11]. However, the variety of elements in the spent nuclear waste stream present a major challenge to efficient recovery and reprocessing of U and Pu as well as the conversion of high active waste into appropriate waste forms for storage [10, 11]. Although substantial advances have been achieved, development of better separation technologies with increased efficiency are still needed. This review covers a brief overview of the known spent nuclear fuel management strategies and looks at the challenges experienced in aqueous reprocessing methods. In addition, it explores the opportunities presented by selected porous materials namely silica, metal-organic frameworks (MOFs) and porous organic polymers (POPs) in solid-phase extraction of radioactive waste. The overall goal is to demonstrate the prospects of using functionalized porous materials as sorbents in extraction of spent nuclear fuel elements.

2. Overview on management of spent nuclear fuel strategies

Several strategies are currently being used to effectively manage nuclear waste [12-14]. These include direct disposal in which the nuclear fuel is utilized just once in the reactor then stored for several years in a repository. The other strategy involves reprocessing, where uranium and plutonium in the waste are extracted and reused. Various countries such as France, United Kingdom, Russia, China, and Japan have implemented strategies to reprocess their spent nuclear fuel [13, 14]. Countries including Sweden and the United States (US) have chosen a once-through approach where the spent nuclear fuel is directly disposed of in designated repositories [14].

2.1. Nuclear fuel reprocessing.

The current reprocessing process involves isolating the major actinides namely plutonium and uranium from the rest of the spent nuclear fuel elements (minor actinides and fission products). This is done in order to facilitate storage of reprocessed uranium as a potential key asset or to directly reuse the materials as a new fuel [5, 15]. In general, reprocessing of uranium and plutonium significantly improves utilization of fissile material by an increase of up to 25%. Despite this, there are a number of concerns arising from this approach. These include high cost of reprocessing, increased quantity of newly created high active waste as well as increased chances of proliferation [5]. It has been reported that there is an increase in radiotoxicity particularly in mixed oxide fuels (MOX) where reprocessed uranium and plutonium are both used as fuel in thermal nuclear reactors due to the presence of americium and curium in the mixture [16]. It is therefore important to properly store these highly active wastes resulting from reprocessing process. In order to reduce the prolonged radiotoxicity, improve the energy utilization and reduce the heat load of the final repository, more advanced reprocessing options known as partitioning and transmutation (P&T) have been developed [16].

2.1.1. Pyrochemical and Aqueous reprocessing. Pyrochemical and aqueous reprocessing processes are the two main techniques in which partitioning of spent nuclear fuel is done [16, 17]. Pyrochemical methods are also referred to as the dry route. They enjoy the advantages of being radiation-resistant and have a low risk of criticality [17]. They are yet to be applied on an industrial scale. On the other hand, aqueous reprocessing methods, also known as the wet route are highly developed and are already being utilized at an industrial level. This has made them famous and the ideal method in partition and transmutation studies [5].

There are various aqueous reprocessing techniques for partitioning which include chromatographic separation, ion exchange separation, and supported liquid membrane extraction. Out of these, liquid-liquid extraction which is also known as solvent extraction is currently the most commonly used
method in reprocessing of spent nuclear fuel [18]. Well known Plutonium Uranium Redox EXtraction (PUREX) method is an example of a solvent extraction process. PUREX is presently being used in industrial reprocessing of uranium and plutonium. Most of the current methods under development utilizes the PUREX process as their starting point. Examples of processes which takes this approach includes Trivalent Actinide-Lanthanide Separation by Phosphorous reagent Extraction from Aqueous Komplexes (TALSPEAK) process which was developed in USA and the DIAMide EXtraction (DIAMEX)/Selective Actinide EXtraction (SANEX) process [18, 19]. Figure 1 shows the TALSPEAK and DIAMEX/SANEX process [18].

![Figure 1. TALSPEAK and DIAMEX/SANEX process.](image)

2.1.2. Challenges in aqueous processes. Organic solvent is used as the extract in aqueous processing. It consists of the extractant ligand which is normally brought into contact with the aqueous phase of the used nuclear fuel solution [9]. Figure 2 shows the schematic representation of the process. In brief, an organic solvent represented by the blue colour carries molecules in the organic extractant, represented by blue symbols, is mixed with the aqueous phase, represented by the yellow colour, and contains the metal cations marked with the black symbols in order to enhance the contact surface among the two phases i.e. organic and aqueous phases. Additionally, the phases are separated further where the metals extracted are moved to the organic phase.

![Figure 2. Schematic representation of solvent extraction process.](image)

The organic extractant is carefully selected to ensure it is capable of selectively separating the elements of interest from the aqueous solution into the organic phase [11]. The overall goal for determining the choice of solvent extractant and molecular diluent is the ability to minimize the total quantity of secondary waste generated at the end of a separation process. To accomplish this, the extractant should satisfy the CHON principle i.e. it should only have elements namely carbon (C), hydrogen (H), nitrogen (N), and oxygen (O). Various solvents such as N,N'-dimethyl-N,N'-dioctylhexyloxyethyl malonamide (DMDOHEMA), N,N,N',N'-tetraoctyldiglycolamide (TODGA) have been used in combination with molecular diluents such as n-dodecane or kerosene to separate actinides from the lanthanides [11, 20]. However, third-phase formation has been reported in cases where a high concentration of nitric acid solution is present in the feed solution [11, 21]. The use of ionic liquids (ILs) has proved to be a better alternative in minimizing third-phase formation in comparison to molecular diluent. Different studies involving application of ionic liquids as a substitute for molecular diluents in the separation of actinides have been carried out actinides [22-25]. Despite this, the implementation of the CHON principle in choosing a suitable ionic liquid limits their choice.
3. Solid-phase extraction

Solid-phase extraction (SPE) can be explored to overcome most of the challenges encountered in liquid-liquid extraction. It is viewed as one of the most convenient and effective approaches in the removal of cations in radionuclide solutions [26-28]. This is due to its ability to increase the selectivity and sensitivity of the method as it permits discriminatory binding of analyte to a solid support where the analyte can be collected and thereafter eluted using small quantity of a different solvent [28]. This method involves an aqueous solvent comprising of a mixture of various elements being passed through a column containing the sorbent material [9]. The element of interest is extracted and remain adsorbed in the sorbent material whereas the rest of the elements and liquid flows through the column. Figure 3 shows the schematic representation of the process [9].

![Figure 3. Schematic representation of the Solid-phase extraction.](image)

This approach has many advantages such as it increases the enrichment factor, prevents emulsion formation, improves the safety of the radioactive samples, reduces the volume of reagents used and hence the overall costs as well as being flexible which makes it easier for process automation [29-31]. This makes it a promising alternative which needs further studies. In order to explore fully the potential of adopting SPE in nuclear reprocessing, it is vital to choose appropriate sorbent material which are easy to design and stable enough capable of withstanding extreme conditions witnessed in spent nuclear fuel.

The application of solid materials to substitute solvent extraction helps in minimizing degradation of chemical diluent which reduces its performance as well as the elimination of secondary waste formation [32]. Besides, using sorbent materials results into faster reaction kinetics, elimination of phase modifiers and permits the use of insoluble extractant molecule in the organic segment [32]. Moreover, utilization of these materials significantly decreases the process of actinide separations in comparison to solvent extraction methods. This results into reduction of cost as well as the time needed to perform the separation process. Lastly, using sorbent materials for nuclear waste separations gives rise to the possibility of using the sorbed radionuclides as a precursor for the waste form or a transmutation matrix. This further saves on time as well as the cost related to additional processing such as stripping or oxide conversion [33].

A wide range of stationary phases exists for SPE which includes using ionic interactions, hydrophobic or polar [32]. In the case of radionuclides, the affinity between the stationary phase and the analyte is carried out by anion exchange or by formation of surface complexes. Resins are distinguished by the nature of the support carrying the affinity function, the link between the support and the function, and the affinity function itself [32]. In this regard, various materials have been used as a solid support for resins.

3.1. Potential of porous materials as sorbent materials

The use of porous materials as solid support plays a crucial role in separation of radionuclides due to their unique structure [33]. The structure has clearly evolved over time starting from the inorganic open frameworks of silicates, metal–organic frameworks (MOFs) up to the current porous organic polymers (POPs) [33-36]. Conventional porous materials, namely zeolites as well as activated carbons
are known for their robustness [33]. However, their inability to be fine-tuned with a number of organic compounds to accomplish the intended binding affinity calls for additional research. On the other hand, MOFs are porous materials composed of organic linkers coordinated with inorganic metal clusters [34]. They are known to exhibit excellent designability of their structure, high specific surface areas, high porosity, tunable chemical features making them promising sorbent materials in sequestration of radionuclides [33]. However, their hydrolytic instability particularly in highly acidic solution poses a challenge especially in nuclear fuel reprocessing considering the extreme conditions involved in this environment. In addition, their application in adsorption of transuranic radionuclides such as neptunium, plutonium, americium and curium, is still being studied [33]. POPs are porous materials which are covalently linked to polymers and have high surface areas [33]. These materials can be categorized into two main classes namely amorphous and crystalline. Amorphous POPs include porous polymer networks (PPNs), porous aromatic frameworks (PAFs) and hyper crosslinked polymers (HCPs) while crystalline POP is made up of covalent organic frameworks (COFs) [37-39]. As a result of the covalent bond linkages in their structures, POPs have high chemical and thermal stability which makes them attractive as sorbent materials in nuclear reprocessing. In addition, POPs are flexible in their structural design. They can therefore be tailored to suit a specific need which is crucial in promoting selectivity and sorption capacity. Therefore, this can be studied further and be applied in the design of new sorbent materials for use in nuclear reprocessing.

3.2. Design of sorbent materials using functionalized porous materials

The design and development of advanced sorbent materials employed in the solid-phase extraction play a vital role in improving selectivity and hence separation of minor actinides intra-group such as americium and curium from the fission product [31]. One of the main drawbacks in developing the sorbent is leaching of the stationary phase during the impregnation of the chelator-containing organic phase [32]. This has necessitated the usage of functional solid-phase extractants within the porous solid. In general, it has been noted that the most effective solid-phase extractants come from impregnation using organophosphorus compounds, such as phosphonates, alkylphosphates, and also nitrogen-containing reagents, for instance, diamides, malonamides, among others [32]. Besides, a significant number of them have been developed using novel ligands such as task-specific ionic liquids particularly in carbon based porous materials [40, 41]. The use of functionalized carbon-based materials, porous silica, and polymer resins as solid support materials for radionuclide separation is well-studied [41-43]. However, the potential of modification of other porous materials such as zeolites, metal–organic frameworks, porous organic polymers among others for application as sorbent material in solid-phase extraction of nuclear waste has not been fully explored despite their high affinity towards radionuclide adsorption. Their efficiency could further be improved by integrating an extractant ligand onto their surface by impregnating or coating just as it has been done for other solid support materials [41-43]. Therefore, there is need to extend the study to cover these porous materials for effective separation of nuclear waste. This can promote the development of solid-phase extraction system capable of removing spent nuclear fuel elements from aqueous solution for industrial application.

4. Conclusions

The challenges experienced in aqueous processing of nuclear waste can be solved through adoption of solid-phase extraction using appropriate sorbent material. Several porous materials namely silicates, carbon, zeolites, metal–organic frameworks and porous organic polymers have shown promising potential in development of advanced sorbent materials for separation of radionuclides. Porous organic polymers have stood out due to their high chemical and thermal stability which are desirable in spent nuclear fuel environment. In addition, the efficiency of these materials can further be improved by modification using compounds such as organophosphorus, nitrogen-containing compounds and ionic liquids. This can help in eliminating leaching of the stationary phase during the impregnation of the chelator-containing organic phase as currently used in aqueous reprocessing. This can lead to
development of a robust radionuclide solid-phase separation system that can be utilized in separation of spent nuclear fuel at an industrial scale as well as a rapid remediation strategy during nuclear accidents.

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