The Future of Nuclear Energy: Electrochemical Reprocessing of Fuel Takes Center Stage

by Bethany Kersten, Krista Hawthorne, Mark Williamson, Rohan Akolkar, and Christine E. Duval

Nuclear power generates approximately 10% of the world's electricity, and in 2020, nuclear energy surpassed coal in US electricity generation.[1,2] Furthermore, the International Atomic Energy Agency predicts that the worldwide nuclear energy capacity will nearly double by 2050.[3] Nuclear power uses energy-dense fuel, does not directly produce greenhouse gas emissions, and provides dependable baseload energy. Despite these advantages and the growing prominence of nuclear reactors in commercial power generation, long-term management of used nuclear fuel (UNF) remains a key challenge for the industry. During nuclear power generation, fuel rods containing enriched uranium are irradiated by neutrons, which causes uranium nuclei to fragment and generate energy through a process called fission. When used fuel rods are removed from the nuclear reactor, they contain both unreacted uranium (U) and elements created during fuel irradiation, including transuranic elements (actinides with a higher atomic number than uranium, e.g., Np, Pu, Am, Cm), lanthanides, noble metals, and others (Fig. 1). A key challenge for UNF management is the radiotoxicity derived from the transuranic elements (TRU), which have extremely long half-lives (e.g., Am-241 has a half-life of 432 years and its decay product, Np-237, has a half-life of 2.14 million years). It takes approximately 300,000 years for UNF to decay to the radiation levels of naturally occurring uranium ore due to the slow radioactive decay of the transuranic elements. However, it is possible to separate uranium and TRU from the bulk of the UNF through reprocessing (recycling), thereby decreasing the decay time of the waste from 300,000 to 300 years.[4,5] Reprocessing recovers the actinide elements for future use as fast neutron reactor fuel and minimizes the waste volume for direct disposal. In addition to the decrease in waste volume, the waste poses a lower threat to the environment if the integrity of the containment vessels were to be compromised, as the elements that are contained have much lower radiotoxicity than the actinides. Direct disposal and reprocessing paths for UNF disposition are illustrated in Fig 1. Recycling components of used nuclear fuel will bring the nuclear fuel cycle closer to being self-sufficient and decrease the demand and environmental impact of uranium mining.

Electrochemical Processing of UNF

Actinide separation from bulk UNF can be achieved via a series of electrochemical operations in high-temperature molten salt electrolytes, with the main separation occurring through electrorefining of the actinides. In the field of nuclear fuel reprocessing, this approach is sometimes referred to as pyrochemical reprocessing, pyrometallurgical reprocessing, or pyroprocessing. This type of UNF separation was first developed in the 1950s, and the technology was greatly expanded in the 1980s as part of the United States' Integral Fast Reactor program. This program proposed a metal fuel-based fast spectrum reactor that incorporated onsite UNF recycling.[7] Electrochemical reprocessing can be used to recycle fuel from a wide variety of reactors, including uranium oxide fuel from light water reactors (LWRs), metal fuel from fast reactors, and molten salt fuel from molten salt reactors. In the case of molten salt reactors, the electrolyte can be tailored to the reactor salt (e.g., UNF from fluoride-based molten salt reactors are reprocessed in fluoride salts). In the case of oxide fuel from LWRs, a two-step process is used: 1) oxide reduction: an electrochemical conversion of oxide fuel to its metallic form, and 2) electrorefining: electrodissolution and electrodeposition of the actinides from UNF. After the electrodeposited actinides are removed from the refiner, any residual actinides and lanthanides that

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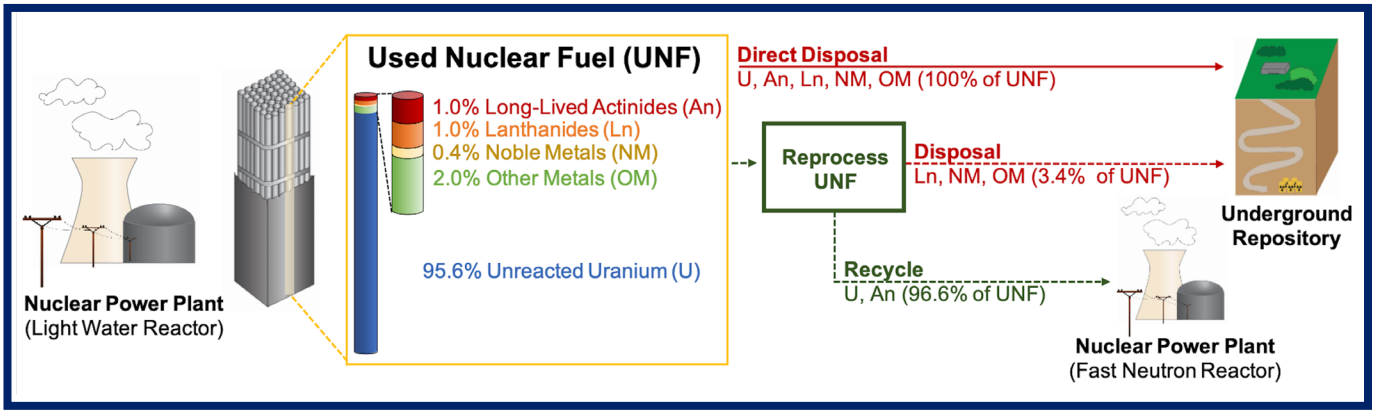

Fig. 1: *An overview of potential paths for the treatment of used nuclear fuel (UNF). Nuclear fuel assemblies (bundles of fuel rods) are placed within the reactor core at the nuclear power plant. The uranium oxide fuel assemblies (100% uranium per metal basis) are bombarded with neutrons during plant operation, generating energy and many new elements. After 1-2 years of service, UNF is removed from the reactor and stored in a pool prior to disposal or reprocessing. The representative UNF composition shown above is based on a used nuclear fuel rod from a pressurized water reactor after 10 years of cooling in a storage pool.[6] Percentages are given as weight percent of total metals. Currently, the US UNF management plan is direct disposal in underground repositories. Alternatively, U and TRU can be recycled for reuse in fast neutron reactors via electrochemical reprocessing. In this scenario, only the remaining fission products, lanthanides (Ln), noble metals (NM), and other metals (OM), are sent to an underground repository as stable waste forms. Reprocessing UNF can drastically reduce the volume of waste buried underground and allow for additional electricity generation through the use of recovered actinide elements as fuel.*

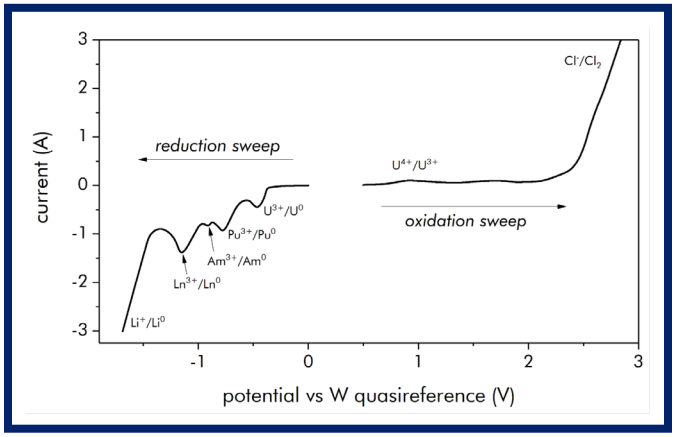

Fig. 2: *Linear sweep voltammogram of a typical electrorefiner molten salt versus a tungsten quasireference electrode. Ln represents a mixture of lanthanides in the electrorefiner. Molten salts are excellent supporting electrolytes for actinide electrodeposition because of their electrochemical stability. In the widely-studied LiCl-KCl eutectic melt, electrolyte decomposition occurs at the negative potential limit via Li metal deposition and at the positive potential limit via evolution of Cl_2, in a window of about 3.3 V. This large window of stability is important for UNF processing because the reduction of actinides to their metallic states occurs within the LiCl-KCl stability window. Adapted with permission from N. Hoyt (Argonne National Laboratory).*

remain in the molten salt are "drawn-down" through electrolysis, resulting in an electrolyte that can be reused. The recovered actinides are recycled, and the lanthanides are prepared for disposal as stable waste forms.

Electrochemical separation of actinides is performed using molten salt electrolytes because of their high ionic conductivity (>1 S/cm) and wide electrochemical stability windows (>3 V).[8,9] Furthermore, exchange current densities for solid metal electrodeposition in high-temperature molten salts are typically in the 10-100 mA/cm^2 range. These properties result in high processing rates, low energy consumption, and absence of parasitic reactions. In the context of the nuclear industry, molten salts offer the additional benefit of radiolysis resistance due to the fast recombination kinetics of the salt, whereas other solvents decompose due to the high radiation field of the UNF.[10,11] Figure 2 shows the electrochemical activity associated with various actinides and lanthanides within the electrochemical stability window of a LiCl-KCl eutectic electrolyte. Note the very negative reduction potentials for the actinide electrodeposition reactions.

Oxide reduction and electrorefining processes have been demonstrated both at the laboratory- and pilot-scales, but several challenges in both processes still remain. In this article, we review the current status of these processes, discuss key technical challenges, and offer our perspective on future prospects as they relate to electrochemical reprocessing of UNF.

Oxide Reduction

Before actinides can be recovered from LWR or other oxide fuels, they must be converted from oxide to metallic form. This transformation is accomplished via solid-state electrochemical conversion. As LWR UNF is predominantly U (>95 wt% metals basis), the oxide reduction process is explained here in the context of UO_2. The process uses a porous stainless-steel basket to contain the chopped UNF. A current collector is placed in the center of the UO_2 bed and serves as the cathode lead in the electrochemical cell. A consumable or inert material is employed as the anode, and a LiCl melt with ~1 wt% Li_2O at 650ºC serves as the molten salt electrolyte (Fig. 3).[12,13,14,15] At the cathode, UO_2 is electrochemically reduced to U (at potentials negative with respect to –2.40 V vs. SHE). At the anode, O^{2-} liberated during the reduction of UO_2 is oxidized to O_2:

$$UO_2 + 4e^- \rightarrow U + 2O^{2-} \quad (1)$$

$$2O^{2-} \rightarrow O_2 + 4e^- \quad (2)$$

In an alternative route, UO_2 can also be chemically reduced through a metallothermic reaction with Li metal:[16]

$$UO_2 + 4Li \rightarrow U + 2Li_2O \quad (3)$$

The Li metal reactant required for Eq. 3 is formed by the *in situ* electrochemical reduction of Li_2O present in the electrolyte. This reduction occurs at a potential of about –2.47 V vs. SHE at 650ºC, i.e., 70 mV more cathodic compared to the potential at which UO_2 is reduced.[18]

$$Li_2O + 2e^- \rightarrow 2Li + O^{2-} \quad (4)$$

Oxide Reduction Challenges

Unanswered questions remain pertaining to the operation of the oxide reduction process and its chemistry. One of the challenges is understanding the transport mechanism of O^{2-} out of the reduced UNF cathode pellet. The O^{2-} diffusion in the solid pellet is slow and limits the overall rate of the process.[18,19] This limitation occurs because diffusion coefficients in solids are orders of magnitude lower than those in liquids Fig.4. Additionally, controlling the Li_2O concentration in the bulk electrolyte is important. Too high of a concentration of Li_2O may result in thermodynamically unfavorable conditions, whereby Li deposition is favored over lanthanide oxide reduction in UNF (the UNF contains lanthanide fission products).[20] Many current research efforts in this field are focused on optimizing cell operating parameters, identifying inert anode materials that maintain their integrity at high processing rates, and optimizing UNF oxide particle sizes and geometries for more efficient O^{2-} out-diffusion.[16,21] A fundamental approach to understanding the O^{2-} out-diffusion process and influencing factors is needed. Precise characterization of the transport within the solid pellets can enable a theory-guided approach towards achieving enhanced oxide reduction rates. Furthermore, understanding the synergistic interplay between the electrochemical (Eq. 1-2) and chemical routes (Eq. 3-4) for UO_2 reduction is an opportunity for improved fundamental understanding of this process.

Electrorefining

Once the metal oxide UNF is reduced to its metallic form in the oxide reduction step, the fuel is moved to the electrorefiner where the metallic actinides are separated from the fission products through

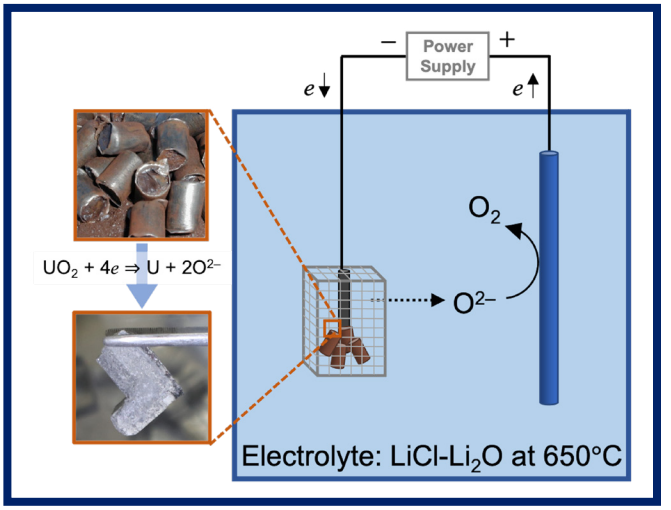

Fig. 3: *Schematic of the oxide reduction cell during operation. UO_2 is electrochemically converted to U, and the reduction front moves gradually inwards from the shell towards the core of the oxide particles. Oxide ions diffuse out of the solid-state pellets and are transported towards the anode where they are oxidized to O_2.*

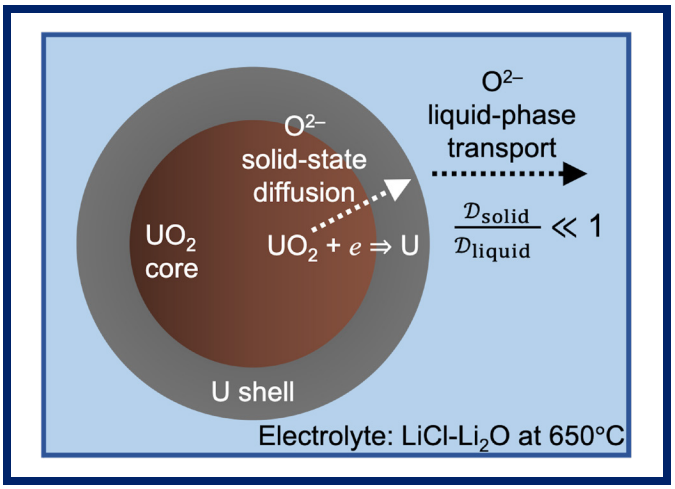

Fig. 4: *During the electrochemical conversion of UO_2 to U, O^{2-} is produced and must diffuse out of the UNF oxide pellet. Transport of O^{2-} is limited by the sluggish solid-state diffusion process within the pellet, which limits the oxide reduction rate.*

electrodeposition in a LiCl-KCl eutectic molten salt electrolyte at 500ºC or LiCl electrolyte at 650°C.[22,23] The anode is a porous stainless-steel basket containing the metallic UNF and the cathode rod material is stainless-steel. At the anode, the actinides and fission products are oxidized to form their soluble chloride complexes. Noble metals and, if present, elements that comprise nuclear fuel cladding do not dissolve and remain in the anode basket. Figure 5 shows a schematic representation of the electrorefining process. The electrorefining operation occurs in two parallel stages. During steady-state operations, uranium is electrodeposited at a uranium-deposition cathode, and at a separate cathode, uranium and the transuranic elements are co-deposited. This co-deposition occurs because polarizing the co-deposition cathode to sufficiently negative potentials results in an alloy deposit of U, Pu, Am, and Np.[9] Once the desired actinides are deposited, the cathodes are removed from the electrorefiner and the metal is separated from residual molten salt in a high temperature vacuum furnace. Finally, the actinides are consolidated into an ingot and are ready for fabrication into nuclear fuel rod form.

Lanthanide Draw-down

Over time, the lanthanide concentration in the electrorefiner electrolyte increases. The increased activity of the lanthanides in the electrolyte moves their reduction potentials to more positive values, which can result in co-deposition of lanthanides during actinide electrorefining.[24,25,26] In order to prevent this, electrochemical separation of lanthanides is performed in a separate process. In this process, called "draw-down", a cathode is polarized to sufficiently negative potentials to facilitate the electrodeposition of the lanthanides. Because draw-down is performed in a molten chloride salt, chlorine gas is evolved at the anode. The recovered lanthanides are then prepared as stable waste forms suitable for disposal. The electrorefiner salt need not be treated in its entirety for lanthanide separation. Instead, the lanthanide fission product concentrations can be actively monitored in the electrorefiner using electrochemical sensing. When the lanthanide concentration reaches a critical level, a portion of the electrorefiner salt can be removed and treated via the draw-down method.

Electrorefining and Draw-down Challenges

Electrorefining processes for UNF have been under operation for decades. Nevertheless, there is an incomplete understanding of the actinide and lanthanide species transport in a molten salt, kinetics of their electrochemical transformations, and the nucleation and growth of the resulting electrodeposits. Actinides and lanthanides exhibit multivalency in the halide environment of a molten salt and many undergo multistep electron-transfer reactions during electrodeposition. Several publications have examined the redox behavior of actinides in LiCl-KCl eutectic salts. Most actinides are found to behave quasi-reversibly or irreversibly during fast scan voltammetry experiments and at the high concentrations (i.e., higher than 2 wt% actinide) typically found in an electrorefiner.[27,28,29,30] This behavior means that the deposition reactions experience kinetic limitations of a measurable degree. In an analogous lanthanide electrochemistry study, it was shown that kinetic limitations during Nd deposition in a $NdCl_3$-LiCl-KCl system result in out-diffusion of intermediate species (Nd^{2+}) causing coulombic efficiency loss. This type of behavior may be prevalent in other multivalent actinide and lanthanide systems and thus deserves investigation.[31] Additionally, such behavior may influence the accuracy of electrochemical sensing as part of process monitoring, which employs voltammetric waveforms to correlate peak current densities to actinide or lanthanide concentrations.[32] Another aspect of the process that is not well-controlled is the formation of the actinide deposits. Uranium typically deposits dendritically, even under low current and high mass transfer conditions. The dendrite morphology is highly dependent on the cell operating conditions, with larger crystals observed at lower deposition currents and thin, branching dendrites observed at high deposition currents.[33] The dendritic nature of the uranium deposits can result in detachment from the electrode surface which may in turn reduce recovery.[34] Understanding the nucleation and growth mechanisms of actinides is thus important.[35] Quantifying these effects will enable theory-guided designs for more efficient electrorefining processes.

Summary

Nuclear energy is an essential component in humankind's transition to carbon-free energy. Recycling used nuclear fuel reduces waste, increases resource utilization, and enables the use of next-generation advanced nuclear reactor technologies. Molten salt electrolysis methods play a key role in the recycling of used nuclear fuel due to their scalability, potential for *in situ* diagnostics and control, and their ability to produce recycled actinides that are suitable for re-use as reactor fuel without generating hazardous waste. Much work is still needed to address the unanswered scientific questions outlined

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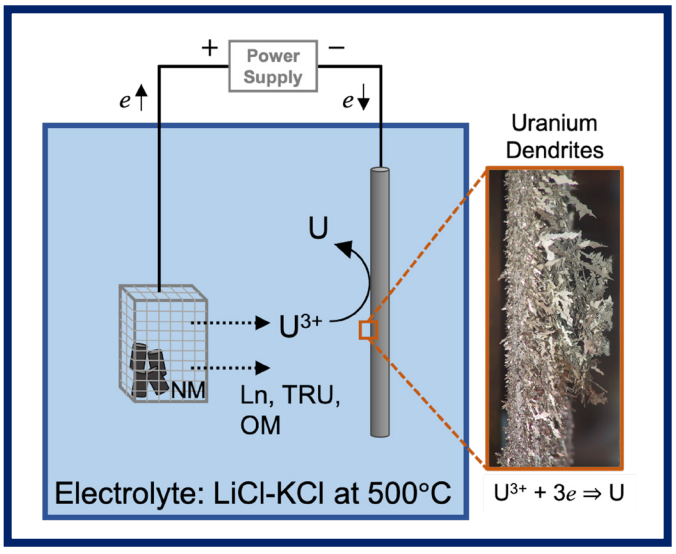

Fig. 5: *Electrorefining involves the anodic dissolution of the elements that constitute UNF and electrodeposition of the actinides onto cathodes. Transuranic elements (TRU), lanthanides (Ln), and other metals (OM) (including alkali and alkaline earth metals) anodically dissolve into the molten salt while noble metals (NM) and fuel cladding materials remain in the anode basket. In electrorefining, uranium is electrodeposited onto one cathode and uranium and transuranics are co-deposited at a second cathode.*

herein and to enhance our understanding of the relevant transport and reaction processes in molten salt based electrochemical technologies for nuclear fuel reprocessing.

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