

In-situ, Real Time Detection of Impurity Transport in Plutonium Electrorefining

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Electrorefining is the primary method used to purify plutonium (Pu) metal across the National Nuclear Security Administration complex. Present day operations rely on constant potential control in a two-electrode molten salt system, with periodic open circuit potential (back electromotive force, BEMF), measurements to determine the run endpoint. Limited real-time process visibility necessitates conservative operating conditions to minimize impurity carryover, resulting in long run times to purify a full-scale anode.

Lawrence Livermore National Laboratory developed a new method that can monitor impurity transport in real time during electrorefining while being controlled in a constant current state [1]. Under conditions where Pu is the only species transported, the system response follows Ohm's Law, producing a linear current-voltage relationship with the slope corresponding to the cell resistance. Deviation from the linear behavior provides an in-situ indicator of impurity transport and can be used to adjust current output in real time, enabling constant current operation while maintaining product purity targets. Proof of concept was demonstrated using a surrogate system with tin as the actinide surrogate and copper as an impurity in a LiCl-KCl-CaCl₂ molten salt system. A measurable deviation from linear behavior occurred when current surpassed 20 A, which correlates to a change in reaction potential between the electrodes confirming that tin is no longer the only constituent being transported. The newly designed method was implemented in a Pu electrorefining cell. The system was run at a conservative value of 30 A, reaching the endpoint in approximately 40 hours. Future work will focus on parameter optimization and full automation to minimize operator involvement. If proven viable, this approach could be the first step to in-situ real time process monitoring.

[1] Zhang, C., Parkes, A.M. and Holliday, K.S., 2023. First principles optimization of plutonium electrorefining. *Journal of Nuclear Materials*, 577, p.154327.

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