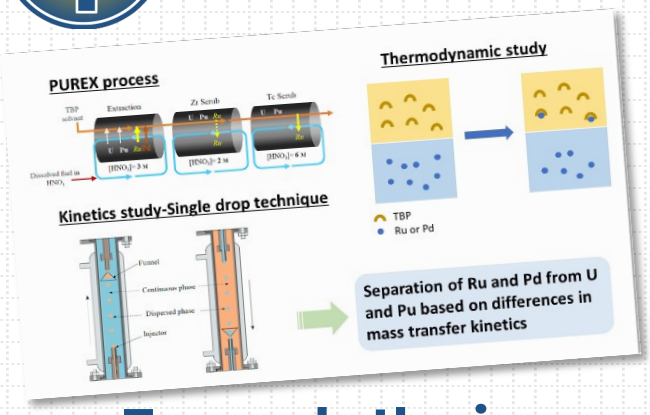
PhD DEFENSE



Marwa KHODER

HYbrid Systems for the Separation Laboratory (LHYS)

Towards the improvement of the separation of Pd and Ru in the PUREX process: Thermodynamic and kinetic studies in the extraction step

Monday, November 30, 2026

9:30 AM

Auditorium ICSM

The objective of this thesis was to investigate the extraction behaviour of the fission products palladium and ruthenium in tributyl phosphate (TBP) systems relevant to the PUREX process, focusing on whether mass-transfer kinetics could enable their separation from major actinides. While ruthenium transfer kinetics have been examined in a few studies, palladium extraction kinetics in TBP/nitric acid media had not, to our knowledge, been quantified. Both metals show moderate extraction by TBP, but their behaviours differ. Palladium extraction may be affected by TBP and diluent degradation products generated through hydrolysis and radiolysis. Ruthenium displays complex speciation in nitric acid and slow interconversion between species, which complicates extraction and stripping and can lead to retention in the organic phase.

For palladium, a distribution coefficient of approximately 0.065 was measured at 3 M HNO_3 in 30% TBP/TPH. Mass-transfer kinetics were quantified using the single-drop technique and compared with published data for U(VI) under comparable conditions. The Pd(II) and U(VI) mass-transfer coefficients were both of the order of 10^{-5} m s^{-1}, indicating that kinetic separation of these species is not feasible under the studied conditions.

For ruthenium, persistent retention in the organic phase after stripping prevented reliable measurement of mass-transfer kinetics. A parametric study showed that ageing of the initial ruthenium solution markedly reduced organic-phase retention and improved the stripping mass balance. Multivariate analysis confirmed ageing as the predominant factor among those investigated. The small retained fraction could not be chemically characterized. Further studies are needed to identify the retained species and reliably quantify ruthenium mass-transfer kinetics.

Keywords: Ruthenium; Palladium; Liquid-liquid extraction; Kinetics; Tributyl-phosphate (TBP); Single drop technique



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