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乙醛肟还原钚的动力学及其在Purex流程铀钚分离中的应用

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摘要 采用分光光度法研究乙醛肟浓度、酸度、NO^-_3浓度、Fe(Ⅲ)浓度和温度等对乙醛肟还原Pu(Ⅳ)反应的影响，得到了反应速率方程和相应的物化参数。实验表明：提高乙醛肟浓度和温度、降低酸度皆有利于加快乙醛肟与Pu(Ⅳ)反应的速率，而NO^-_3浓度和Fe(Ⅲ)浓度却对反应的速率影响不大；25 ℃时，该反应的速率常数为（39.51±0.05） $(mol/L)^{1.1}\cdot min^{-1}$，反应活化能$E_a$=（88.96±9.43） kJ/mol。乙醛肟反萃Pu(Ⅳ)的单级实验和模拟Purex流程1B槽的串级实验结果表明：在以乙醛肟为还原剂的8级反萃、6级补萃的串级实验中，铀的收率大于99.99%，钚的收率为99.99%，铀中去钚的分离系数达到1.05×10^4，钚中去铀的分离系数达到2.7×10^5。

关键词 乙醛肟 Pu(Ⅳ) 还原 反应动力学

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Kinetics of Pu Reduction by Acetaldoxime and Application of Acetaldoxime to Separation of Pu From U in Purex Process

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Abstract The effects of concentrations of C_2H_4NOH, H^+, NO^-_3, Fe(Ⅲ) and temperature on the reaction rate of the Pu(Ⅳ) acetaldoxime were studied by spectrophotometry, and then the rate equation and the corresponding parameters were obtained. The reduction rate of Pu(Ⅳ) can be improved by either increasing the acetaldoxime concentration and the temperature, or decreasing the concentration of acid. However, the effects of concentrations of NO^-_3 and Fe^{3+} on the reduction rate of Pu(Ⅳ) are negligible. It is found that the rate constant is (39.51±0.05） $(mol/L)^{1.1}\cdot min^{-1}$ at 25 ℃, and the activation energy is （88.96±9.43） kJ/mol. In the counter current cascade experiments with acetaldoxime used as reductant (in which 6 stages for supplemental extraction, 8 stages for stripping, and ratio of flow is 1BS∶1BF∶1BX＝1∶4∶1), the recoveries are more than 99.99% for U and 99.99% for Pu . The separation factor of Pu from U is 1.05×10^4, while that of U from Pu is 2.7×10^5.

Key words acetaldoxime _ Pu(Ⅳ) _ reduction _ reaction kinetics

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