Reduction of uranium (VI to IV) by hydrogenation using Adams’ catalyst

Avinash Sahu, Tessy Vincent*, J.G. Shah, P.K. Wattal

Process Development Division, Nuclear Recycle Group, Bhabha Atomic Research Centre, Mumbai 400 085, India
Tel. +91 22 25595488; Fax: +91 22 25505340; email: tessyv@barc.gov.in

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ABSTRACT

Plutonium uranium reduction extraction using U(IV) as universal reductant for Pu partitioning is the only technology practiced internationally to recover U and Pu from spent nuclear fuels. Uranous requirement of Indian reprocessing plants is met by the electrolytic reduction of uranyl nitrate with 50–60% conversion. Though the current requirement can be met with this method, it increases the load on uranium purification cycle. In addition, it is a batch process with slow kinetics. In order to achieve higher conversion of uranyl nitrate to uranous nitrate, catalytic reduction method using hydrogen in presence of Adams’ catalyst (PtO2) was tried. Parametric studies have been performed in an autoclave to evaluate the effect of U(VI) concentration, the role of hydrazine nitrate and pressure. It is observed that kinetics is improved at higher pressures. The studies revealed that near total conversion of uranium from (VI to IV) can be achieved by the catalytic reduction route.

Keywords: Uranyl nitrate; Adams’ catalyst; Hydrogenation; Uranous; Reduction

1. Introduction

Reprocessing flow sheet-based on plutonium uranium reduction extraction process essentially involves dissolution of used nuclear fuels followed by solvent extraction cycles. The first extraction cycle, comprises of co-decontamination and U/Pu partitioning. Partitioning involves the selective stripping of Pu from U. This is accomplished by the reduction of extractable Pu(IV) to the non-extractable Pu(III) oxidation state. Among the various reducing agents, U(IV) is most widely accepted and employed. In Indian reprocessing plants, uranous nitrate [U(IV)] is produced externally by conventional electrolytic reduction of U(VI) using titanium substrate insoluble anode with hydrazine nitrate as uranous nitrate stabilizer. Hydrazine nitrate scavenges the nitrous acid generated by autocatalytic decomposition of nitric acid and prevents the re-oxidation of U(IV) to U(VI) [1]. Limited conversion of 50–60% leading to increase in uranium processing load is the major drawback of the existing electrolytic route. Other limitations includes poor kinetics, frequent recoating of electrode and secondary waste generation during decontamination of anode. Use of cation exchange membrane to separate catholyte from anolyte could enhance percent conversion. However, developing the membrane...