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Distribution behavior of U(VI), Am(III) and Eu(III) on diglycolamide based extraction chromatographic resin in perchloric acid medium

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Abstract An attempt has been made in the present work to investigate the role of anion for the uptake of Am(III)/Eu(III)/U(VI) by extraction chromatography (EXC) resin incorporating tetra-*n*-octyl-3-oxapentanediamide, commonly referred to as tetra-octyl diglycolamide (TODGA). In contrast to the nitric acid, perchloric acid medium favors extraction of trivalent metal ions even at low acidity (pH 2) and is almost insensitive to the acidity up to 5 M. Exceptionally large distribution coefficients (10^5–10^6) in the wide range of perchlorate concentration (10^{-2}–5 M) is quite unusual and is by far the largest reported in the literature for Am(III)/Eu(III). Thermodynamic data suggests the possibility of inner sphere/cation exchange mechanism involving TODGA aggregates at higher acidity but outer sphere/cation exchange mechanism at low acidity for Eu(III). There is a possibility of employing TODGA based EXC resin for the remediation of liquid waste (contaminated with long lived transuranics like $^{241/243}$Am and ^{245}Cm) in the wide range of acidity.

Keywords Americium · Europium · Uranium · Extraction chromatography · Distribution coefficient

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Introduction

Notwithstanding some major mishaps, nuclear energy is destined to play an increasingly important role globally due to significantly lower green house gas emissions and economical considerations [1, 2]. It is imperative however to develop inherently safer reactor designs and technologies to manage the spent nuclear fuel (SNF)/high level liquid waste (HLLW) efficiently. It has been recognized that the principal cause of the radiotoxicity of SNF/HLLW is the presence of transuranics like ^{237}Np, ^{239}Pu, $^{241/243}$Am and ^{245}Cm (which are long lived alpha emitters) [3–5]. The P&T process (partitioning of long-lived radionuclides followed by transmutation) envisages the complete removal of these transuranics (minor actinides) from HLLW and their subsequent burning in the fast reactors/accelerator driven systems as mixed (oxide or metallic) fuel [6–9]. This process leads to the generation of energy and at the same time would alleviate the need for long term surveillance of geological repositories. Alternatively, partitioned minor actinides can be vitrified in suitable matrix (like synroc) and disposed in geological repository. The separation of these transuranics from HLLW reduces the mass/volume of radiotoxic elements (to be discharged after vitrification) significantly, thereby saving the precious space and services in the geological repository.

Development of new extractants/technology for actinide partitioning is one of the challenging areas of research for separation scientists and technologists. Diglycolamides (DGAs), malonamides and carbamoyl methyl phosphine oxides are currently the focus of radiochemists engaged in developing the flow sheets for the partitioning of minor actinides from HLLW [8, 10–14]. High affinity displayed by DGAs towards trivalent lanthanides and actinides is particularly intriguing and has been attributed to their