



HAL
open science

Sonochemical reduction of Pu(IV) in aqueous nitric solutions

Matthieu Viot, Laurent Venault, Philippe Moisy, Vincent Morosini, Sergey I. Nikitenko

► **To cite this version:**

Matthieu Viot, Laurent Venault, Philippe Moisy, Vincent Morosini, Sergey I. Nikitenko. Sonochemical reduction of Pu(IV) in aqueous nitric solutions. Migration 2013 -14th International Conference on the Chemistry and Migration Behaviour of Actinides and Fission Products in the Geosphere, Sep 2013, Brighton, United Kingdom. <hal-04765301>

HAL Id: hal-04765301

<https://hal.science/hal-04765301v1>

Submitted on 4 Nov 2024

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



HAL Authorization

SONOCHEMICAL REDUCTION OF Pu(IV) IN AQUEOUS NITRIC SOLUTIONS

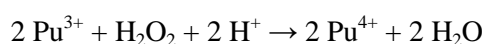
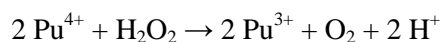
M. Viro¹⁾, L. Venault²⁾, P. Moisy²⁾, V. Morosini¹⁾, S. I. Nikitenko¹⁾

¹⁾ ICSM-UMR5257 CNRS/CEA/UM2/ENCSM, Site de Marcoule,
BP 17171, 30207 Bagnols sur Cèze, France

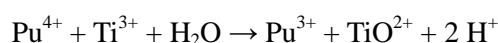
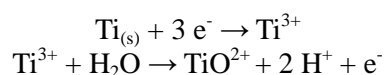
²⁾ CEA/DEN/DRCP, Radiochemistry and Process Department,
BP 17171, 30207 Bagnols sur Cèze, France

The propagation of ultrasound ($16 \text{ kHz} < f < 1 \text{ MHz}$) in a liquid media may lead to acoustic cavitation which is the nucleation, growth, and rapid implosive collapse of vapour filled micro-bubbles. In homogeneous systems, the implosion of these bubbles goes with the *in-situ* generation of transient extreme conditions in terms of temperature and pressure which promote the formation of excited species and free radicals in solution [1,2]. In heterogeneous systems, acoustic cavitation involves the production of micro-jets and shock waves at the vicinity of the treated solid boundary which can be related to several applications such as erosion, extraction, cleaning, dissolution, etc. [1-3]. Because of these particular occurrences, power ultrasound has been showed to enhance the yields and accelerate the reaction rates of numerous homogeneous and heterogeneous chemical systems.

In this study, the behaviour of Pu(IV) in aqueous nitric solutions is considered under 20 kHz ultrasound irradiation. The experiments are performed at $\sim 25^\circ\text{C}$ under Ar flow in a 50 mL home-made reactor and the reactions are followed using a UV-Vis spectrometer. In the presence of HNO_2 scavengers (ex: sulfamic acid or hydrazinium nitrate), ultrasound irradiation of Pu(IV) leads to the accumulation of Pu(III) in solution. Pu(IV) reduction rate is found to be function of the acoustic intensity delivered to the solution. The reduction follows a first-order reaction mechanism and is not complete. In addition, a steady-state is observed for prolonged irradiation. The behaviour of Pu(IV) and Pu(III) in this system is attributed to reactions with sonochemically generated hydrogen peroxide, H_2O_2 , in agreement with the following reactions that can also be observed without ultrasound application [4].



The kinetics observed for Pu(IV) sonochemical reduction are however found to be faster than the kinetics related to H_2O_2 generation under ultrasound at similar conditions. The various investigations show that this difference can be attributed to the mechanical erosion of the ultrasonic tip resulting from the acoustic cavitation generated at the vicinity of the solid surface. The erosion of the tip produces titanium particles and their subsequent solubilization in the sonicated solution (checked with SEM and ICP-AES) may be involved for reactions with Pu(IV) in agreement with the following equation.



[1] K. S. Suslick, D. J. Flannigan, *Ann. Rev. Phys. Chem.* 59, 659 (2008).

[2] T. G. Leighton, *The Acoustic Bubble*, Academic Press: London (1994).

[3] M. Viro¹⁾, T. Chave, S. I. Nikitenko, D. G. Shchukin, T. Zemb, H. Mohwald, *J. Phys. Chem. C* 114, 13083 (2010).

[4] J. M. Cleveland, *The Chemistry of Plutonium*, American Nuclear Society, La Grange Park (1979).