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► To cite this version:

Matthieu Virof, Thomas Dumas, Philippe Moisy, Sergey I. Nikitenko. Recent advances about the multi-scale structural properties and formation mechanism of plutonium oxide nanoparticles in aqueous solution. Congrès SCF 2023, Jun 2023, Nantes, France. hal-04765404

HAL Id: hal-04765404

<https://hal.science/hal-04765404v1>

Submitted on 4 Nov 2024

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RECENT ADVANCES ABOUT THE MULTI-SCALE STRUCTURAL PROPERTIES & FORMATION MECHANISM OF PLUTONIUM OXIDE NANOPARTICLES IN AQUEOUS SOLUTION

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Keywords: plutonium, nanoparticles, colloids, nuclear, ultrasound, sonochemistry

Summary: Plutonium is an anthropogenic radioactive element exclusively formed in nuclear reactors and playing a central role for current and future energetics.¹ In aqueous solution, Pu exhibits an intricate behaviour governed by complexation, redox, disproportionation and hydrolysis reactions. In particular, Pu can be stabilized in acidic solution under four oxidation states ranging from +III to +VI. The proximity of the standard potentials around 1 V/ENH for the associated redox couples gives plutonium the exceptional property to coexist in slightly acidic aqueous solution under these four oxidation states. This complex chemical character is further amplified with the instability of the degrees +IV and + V which can disproportionate and the property of the +IV degree to be highly prone to hydrolysis reactions even in acidic solutions. The latter phenomenon is particularly interesting since hydrolysis of Pu(IV) yields very stable colloidal suspensions of typical emerald-green color.¹ Such a phenomenon has been observed for the first time during the Manhattan project but has been later rather prevented in industrial processing by maintaining Pu flux at high acidity, resulting in a knowledge gap about the physico-chemical properties for these colloids. However, recent evidences of potential environmental migration of radioactive element under the colloidal form increased the attention given to plutonium oxide nanoparticles in addition to their formation mechanism.^{2,3} Furthermore, the question of the structure vs. property relationship for PuO₂ nanomaterials present a strong interest for the development of advanced nuclear energy systems underway. Research about these concrete topics is intensified by the curiosity of scientists who are questioning the physico-chemical property of nanomaterials made out with the element Pu which shows unique properties around the periodic table.⁴

This presentation will summarize the recent advances achieved by our group about the multi-scale structural characterization of PuO₂ nanoparticles and their formation mechanism.⁴ The beginning of the preparation will focus the preparation of reference colloidal suspensions using controlled hydrolysis and sonochemistry (i.e. chemical reactions driven by the propagation of high-intensity ultrasound waves) approaches which allowed the preparation of reference Pu oxide nanomaterials with different sizes.⁵ The thorough characterization of these particles using state-of-the-art laboratory and synchrotron techniques allowed to go deeper in the description of the structural properties of intrinsic Pu(IV) colloids which can be now described as spherical and crystalline nanoparticles showing a PuO₂-like structure.⁶ New insights about the formation mechanism of these nanoparticles in aqueous solution will be finally discussed with the recent evidence of the contribution of hexanuclear oxohydroxo buildings during the process.^{7,8}

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