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CATALYTIC AND REDUCTIVE DISSOLUTION OF CeO₂ and (Ce,Ln)O₂ IN MILD CONDITIONS

Xavier Beaudoux,^{a,b} Matthieu Viot,*^a Tony Chave,^a Gilles Leturcq,^b Nicolas Clavier,^a
Nicolas Dacheux,^a Sergey I. Nikitenko^a

^a Institut de Chimie Séparative de Marcoule, ICSM-UMR5257 CNRS/CEA/UM2/ENCSM, Site de Marcoule, BP 17171, 30207 Bagnols sur Cèze, France.*matthieu.viot@cea.fr

^b CEA/DEN/MAR/DRCP, Direction de l'Energie Nucléaire, Département de Radiochimie et Procédés, Site de Marcoule, BP 17171, 30207 Bagnols sur Cèze, France.

Ceria-based oxides are very interesting materials for automotive catalytic converters. In the presence of noble metals (Pt, Pd, Rh...) deposited at their surface, the catalytic performance of these oxides is highly improved. Although used in catalytic amount by car, automotive industry alone is estimated to consume about 40% of platinum group metals.¹ Due to the continuous rising demand for these metals and their related price increase, the recycling of such materials appears as a viable alternative. Conventional treatment procedures based on pyrometallurgical or hydrometallurgical processes are often time and energy consuming, and generate high volumes of corrosive and toxic effluents. Due to the higher solubility of Ce^{III} compounds compared to Ce^{IV} and the highly positive redox potential for Ce^{IV}/Ce^{III} couple, the reductive dissolution of ceria-based oxides can be considered.

In this study, we investigate the catalytic and reductive dissolution of ceria-based oxides in the presence of Pt nanoparticles (Pt NPs, 2.5 wt.%) deposited at their surface through the in situ sonochemical reduction (Ar, 18 W.cm⁻², 20 kHz) of H₂PtCl₆ salt (1 mL, 4.5 g.L⁻¹). Experiments are carried out at 40°C in diluted HNO₃ or H₂SO₄ to provide protons for dissolution, and HCOOH as reducing agent. [N₂H₅][NO₃] can be used as anti-nitrous reagent or reducing agent. Preliminary dissolutions are performed on CeO₂. The incorporation of trivalent (Nd, Gd), and tetravalent (Tb) lanthanides in various proportions in the oxide towards dissolution rate is then evaluated. Dissolution progress is checked with ICP-AES, UV-Vis spectroscopy, and SEM.

Whatever the oxide and tested conditions, dissolution rates are found to be very weak in the absence of Pt NPs even under sonication. However, the sonochemical deposition of Pt NPs at the surface of CeO₂ (54 m².g⁻¹) catalyse its reductive dissolution in 0.25 M HNO₃ – 1 M HCOOH – 0.2 M [N₂H₅][NO₃]. Surprisingly, after Pt deposition, dissolution rates are found to be higher under stirring than under ultrasound (~30% against 17% in 7 h) which usually increases dissolution rates through surface effects.² A sonochemical denitration phenomenon consuming the reducing species is highlighted. This phenomenon is absent in 0.125 M H₂SO₄ / 1 M HCOOH and leads to equivalent rates of dissolution whatever the processing conditions. Similar observations are reported for mixed oxides. The incorporation of trivalent and tetravalent lanthanides into ceria matrix results however in faster dissolution rates for oxides due to the weakening of the fluorite structure for +III lanthanides and higher redox potential for +IV lanthanide. Dissolution yields are also increasing with the percentage of incorporation of lanthanide in the ceria matrix. As a function of dissolution progress, aggregation of Pt is observed. Lanthanide/Pt partitioning can be contemplated by filtration or centrifugation after dissolution to recover Pt.

References

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