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Actinide -Zirconia Based Materials for Nuclear Applications: Cubic Stabilized Zirconia Versus Pyrochlore Oxide*

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Abstract

Concepts about nuclear energy and nuclear materials have changed considerably over the past six decades. Regardless of one's position on the nuclear generation of electric power, there are serious needs for pursuing fundamental and technological science of existing actinide materials. These needs are best addressed by obtaining an atomic and molecular understanding of these actinides and actinide containing materials. Although electro-nuclear energy is considered less polluting in terms of uncontrolled releases (e.g., SO_2, heavy metals, CO_2, etc.) into the environment, its use produces solid wastes, which offer a challenge for scientists. Fortunately, concepts are being developed to appropriately handle[1] these materials after irradiation, reprocessing, etc.

One envisioned strategy for addressing several radionuclides of concern, once they are partitioned, is to transmute ("burn"/"incinerate") them in a dedicated reactor or some specific neutron source system, (e.g., Accelerator Driven System, "ADS") to provide a more suitable material for disposal. The topic of this paper addresses important materials science issues of americium and/or curium materials that are attractive for different nuclear applications, including disposal/storage options.

One disposal approach is to "recycle" americium and/or curium in a uranium- or plutonium-free host, which would avoid production of additional actinides by neutron capture processes. For this application, is would be necessary to provide a structurally and thermochemically stable material for americium and/or curium. This material would need to meet several criteria concerning neutron cross section, chemical inertness with cladding and/or coolant, melting point, phase stability, thermal conductivity, etc. Potential candidates as MgO, Y_2O_3, $MgAl_2O_4$, CeO_2, CeN, and others have already been envisioned for this application[2,3].

We have pursued studies of zirconia-based compounds that can provide either an inert matrix or a diluent in multiphasic systems, for incineration/transmutation of actinides (Pu, Am, Cm, etc.) We have incorporated and studied structural aspects of actinides incorporated in cubic, zirconia-based materials (both with and without yttria stabilization), as well as in zirconia-based pyrochlore oxides. From this work, we concluded that cubic yttria-stabilized zirconia (Y-CSZ) materials, which are well known refractory ceramics, would be suitable for transmutation schemes for americium and/or

curium. Several experiments have already demonstrated that cubic stabilized zirconia materials are very resistant to neutron irradiation and damages related to fission processes[4].

One particularly attractive material is a 25 mol % yttria-stabilized matrix, which offers a stable, single phase (fluorite-type structure) material that can incorporate significant quantities of americium and/or curium oxides. In a study with americium, we examined the structural and phase behavior of the AmO_2-$Zr0_2$-$YO_{1.5}$ ternary system (see Fig. 1), and also determined that the cubic lattice parameters of the products were linear with the americium composition.

Zirconia-based pyrochlore oxides, which also form refractory ceramics, offer another potential matrix for the transmutation of americium and curium or the incineration of plutonium. The particular pyrochlore oxides discussed here have several common aspects with the components of Synroc[5], although they exhibit structural differences. They consist of the actinide plus zirconium, and have the general formula, $A_2B_2O_7$ (where A = actinide and B = zirconium). The "pyrochlore" terminology comes from the structural similarity of the material to the mineral, "pyrochlore". An interesting aspect of these oxides is that they can exhibit a large domain of homogeneity, which is reflected by the linear variation of cell parameter for the americium containing materials up to 60 mole % AmO_2.

The presentation will compare and discuss the structural properties of these fluorite-type and pyrochlore-type zirconia systems. In addition, the fundamental physicochemical aspects of these materials with regard to their application in transmutation/incineration schemes will be compared to other materials that have been considered for such applications.

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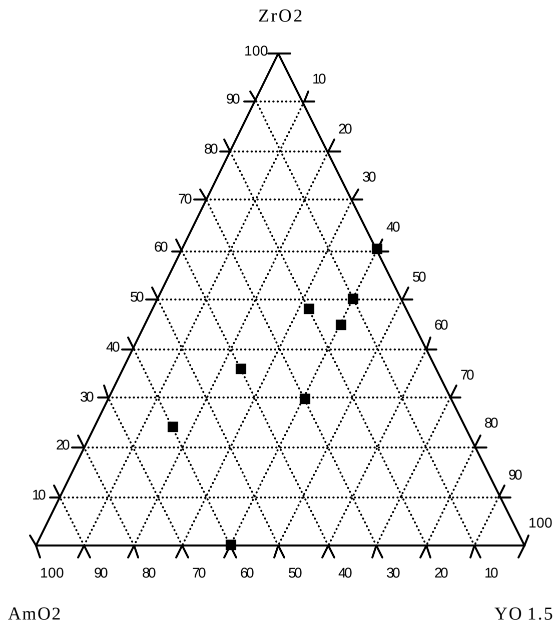

<u>Figure 1</u>: *The pseudo ternary system AmO_2-ZrO_2-$YO_{1.5}$*