Gamma irradiation and actinide doping effect in nanocrystalline $Gd_2Hf_2O_7$

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Abstract

Ceramics materials play an important role in the development of high energy radiation waste forms for nuclear waste immobilization. A suitable ceramic material must be stable, should accommodate radiation induced damages and not lose its shape at high radiation doses or actinide incorporation. A ceramic oxide that stood out is $A_2B_2O_7$ pyrochlore as it's radiation tolerant, flexible and negligible of leaching.

Motivation

- Due to the planet's instability of greenhouse gases, our priority is to promote a cleaner source of power through nuclear technology.

Purpose

- Test the effect of gamma radiation and actinide doping on phase behavior, structure and crystallinity of $Gd_2Hf_2O_7$ (GHO) through X-ray diffraction (XRD) and Raman spectroscopy
- Demonstrate that GHO can withstand high doses of gamma radiation up to 12800 Gy and accommodate relatively very high concentrations of uranium ion (~ 30 %) without any change in phase, structure, and crystallinity

Goals

- Test the effect of ion radiation (alpha, positron) damage
- Study the role of actinide doping on the ion radiation induced damage
- Design highly durable and especially radiation tolerance ceramics which are not prone to amorphization and swelling.

Project Impact

- The study of ceramics will create a versatile number applications of nuclear power through the development of fuels, treat some difficult wastes through separation technologies, structural applications, etc.

Methods

Synthesis procedure:

-Gadolinium hafnate nanocrystals were synthesized on a two- step process. First, the single-source complex precursor of $Gd(OH)_3 \cdot HfO(OH)_2 \cdot nH_2O$ through coprecipitation. Secondly, we control the growth of GHO NCs through molten-salt synthesis.

Sample characterization:

-X-ray diffraction (XRD) and Raman was utilized to evaluate the crystalline phase of these synthesized NCs.

Results

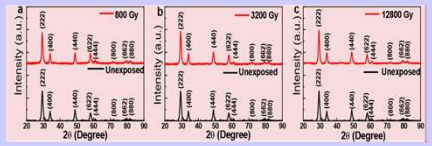


Fig.1 XRD pattern obtained from $Gd_2Hf_2O_7$ compound before and after gamma radiation exposre with doses of 800, 3200 and 12800 grays (Gy).

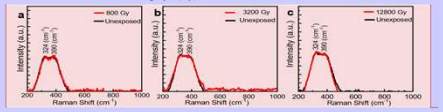


Fig.2 Raman pattern obtained from Gd2Hf2O7 compound before and after gamma radiation exposure with doses of 800, 3200 and 12800 grays (Gy).

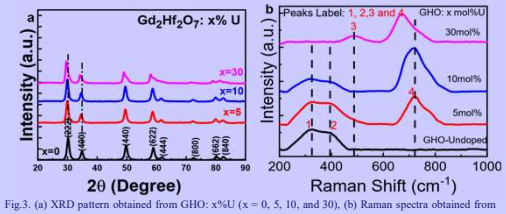


Fig.3. (a) XRD pattern obtained from GHO: x%U (x = 0, 5, 10, and 30), (b) Raman spectra obtained from GHO: x%U (x = 0, 5, 10, and 30).

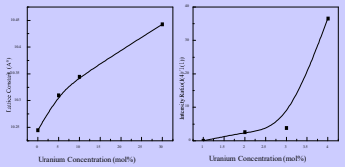


Figure 4. (a) lattice parameter shown along the ordinate obtained from (222) plane values, (b) Ratio of Intensity between Raman modes of the distorted HfO6 (I4) and the main F_{2g} Raman mode of the weakly ordered pyrochlore I(1).This serves as indicator of Uranium induced anion disordering in GHO as a function of Uranium

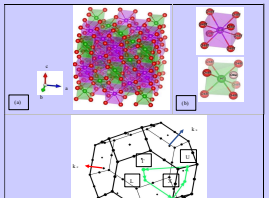


Figure 5. (a) The crystal structure of the $Gd_2Hf_2O_7$ pyrochlore (space group Fd3m), (b) polyhedral of the Gd and Hf sites that compose $Gd_2Hf_2O_7$ illustrating the O sites (O_{48f} and O_{8a}) and its vacancy (O8a), and (c) the first Brillion zone of the $Gd_2Hf_2O_7$. The path in the reciprocal k-space, used in the electronic band structure calculations, is shown by green arrows. Points are as follows: Γ = (0, 0, 0), L = (½, ½, ½), W = (¼, ½, ¼), X = (½, ½, 0), and U = (5/8, 5/8, 0).

Results - con't

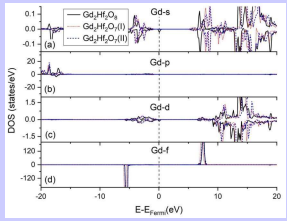
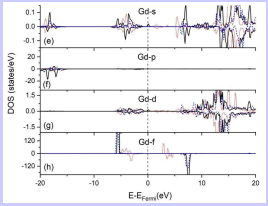


Figure 6.Gd DOS spectra per s, p, d, and f atomic orbitals for $Gd_2Hf_2O_8$ and $Gd_2Hf_2O_7$ structures, with the O_{8a} sites simulated via ghost atoms (I) and being vacant (II). (a)-(c) stable structures and (d)-(f) cation antisites.



Future Goal

- Investigate the resistance to amorphization in Gd2Hf2O7: x mol% U (x =5, 10, 15 and 30) samples in the near future.
- Address whether or not GHO can be an engineering material as a universal nuclear immobilization host. If not, under what parameters is GHO limited to becoming a universal host.

Conclusions

GHO shows the resistance to amorphization high doses of gamma radiation up to 12800 Gy. Furthermore, all purposes we have presented support the idea of GHO being a potential material as a nuclear waste immobilization host. An important remark to note is that weakly ordered (not properly structured) GHO can uptake uranium up to 30 mol% with a structural transformation.

References

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