Fabrication of simulated americium transmutation targets using internal gelation

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Introduction

The long-term radioactivity and heat load of the spent fuel waste fraction after uranium and plutonium recycling is mainly caused by the minor actinides (MA). In particular, americium, constituting a mass fraction less than 0.1 wt.% of spent UO_2-based fuel, dominates the heat emission for several thousands of years. A possible solution to reduce the radiological hazard of spent fuel (SF) is to partition the MA and to transmute the minor actinides into lighter, short-lived elements in fast spectrum reactors or dedicated accelerator driven systems.

Objectives

- **Develop innovative methods regarding conversion of Am content in SF into suitable transmutation targets.**
- **Convert aqueous feeds into solid oxide targets.**
- **Fabricate microspheres via IG and achieve Nd(III) (simulant of Am) doping via infiltration.**

Methods

In this contribution we present how the internal gelation (IG) process (Fig. 1.a-b-c) has been used to fabricate zirconium oxide-based microspheres as a matrix for the transmutation of actinides. By using neodymium as a surrogate for americium, the infiltration of microspheres (Fig. 1.d-e-f) was additionally investigated as an alternative approach towards americium target fabrication. The materials were characterized using analytical techniques such as optical microscopy, thermogravimetry, and gas pycnometry.

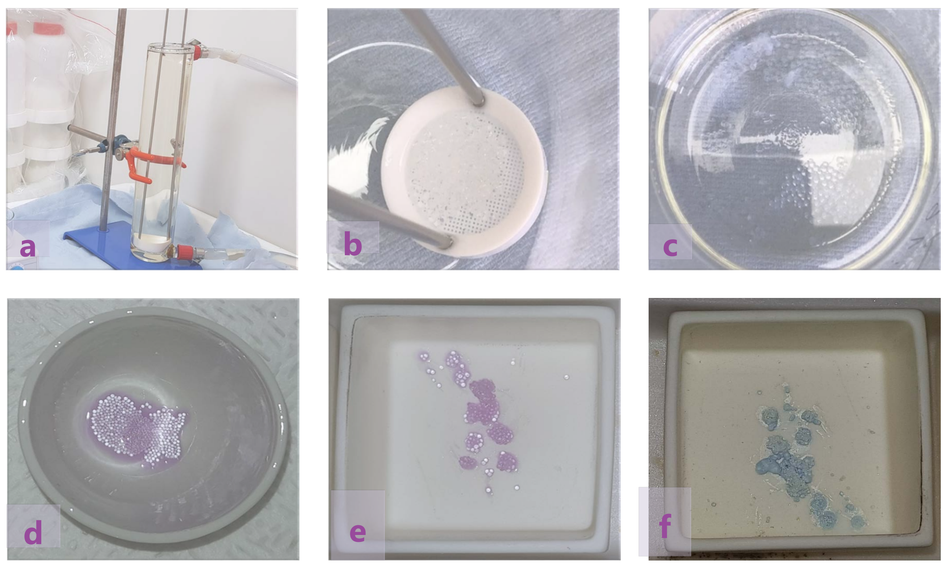

Figure 1. The main steps of the IG experiment: (a) Double jacket column in which gelation takes place; (b) Sieve for collection of the microspheres; (c) Microspheres during washing. Infiltration of 2.4 M $Nd(NO_3)_3$(aq.) into $(Zr_{0.86}Y_{0.14})O_{1.93}$ microspheres: (d) 24 h after drying; (e) Before calcination; (f) After calcination at 900 °C and 2 h isotherm time.

Results and Discussion

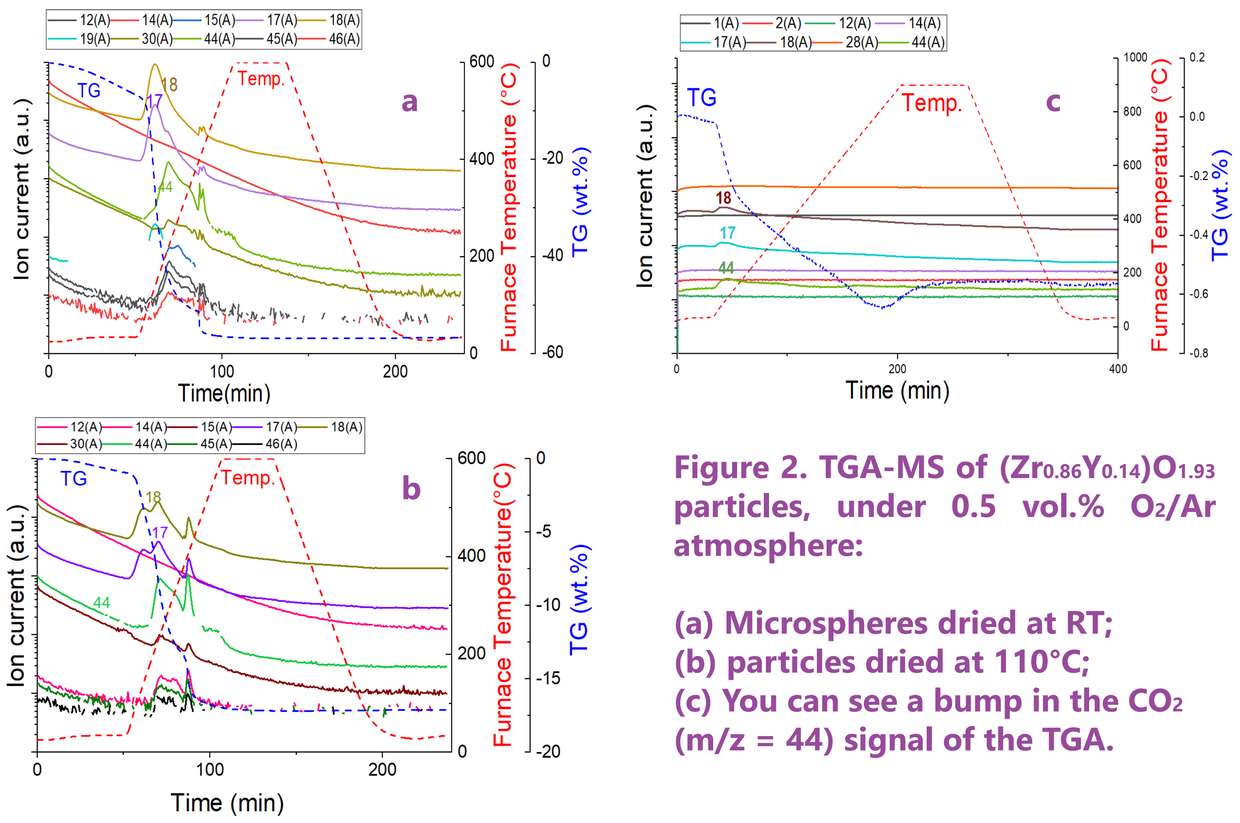

Figure 2. TGA-MS of $(Zr_{0.86}Y_{0.14})O_{1.93}$ particles, under 0.5 vol.% O_2/Ar atmosphere:

(a) Microspheres dried at RT;
(b) particles dried at 110°C;
(c) You can see a bump in the CO_2 (m/z = 44) signal of the TGA.

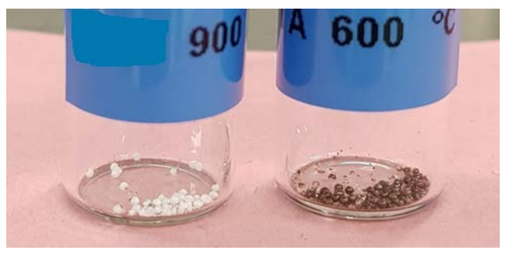

Figure 3. Microspheres calcined in air at 600 °C (right, black) and after second heat treatment in air at 900 °C (left, white).

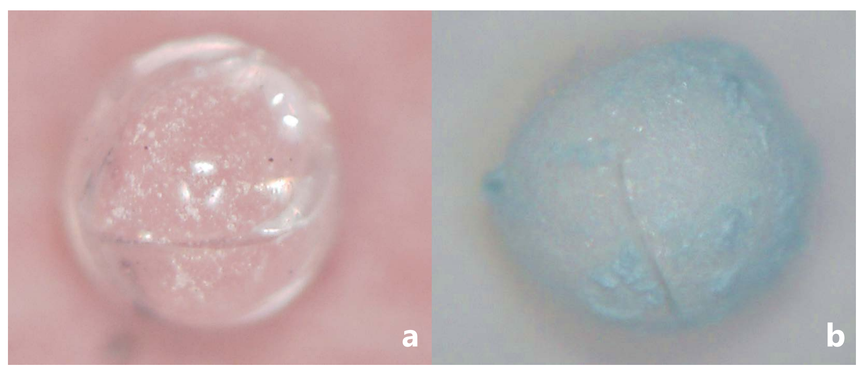

Figure 4. Optical microscopy pictures of (a) un-doped $(Zr_{0.86}Y_{0.14})O_{1.93}$ microsphere; (b) Nd(III)(aq.) solution infiltrated $(Zr_{0.76}Y_{0.14}Nd_{0.10})O_{2-x}$ microsphere.

- For the yttria-stabilized zirconia (YSZ) microspheres dried at 110 °C (Fig. 2.b) a similar profile in mass change was observed as for the RT dried ones (Fig. 2.a), but the relative mass difference of the first mass loss step was significantly lower. This shows that the drying procedure at 110 °C already removed a substantial amount of volatile species from the particles. According to the MS signals, the mass loss in both samples is mainly due to water release (indicated by m/z = 18), and CO_2 release (m/z = 44).
- The color of YSZ microspheres (Fig. 3) was black after calcination at 600 °C and after application of second heat treatment at 900 °C the color was observed as white. There were some remaining carbonaceous impurities from the IG fabrication not entirely removed at 600 °C. A small bump in the m/z= 44 signal of the TGA in the Fig. 2.c is observed. Hence, a higher temperature under oxidizing conditions was required to complete the calcination process.
- The average density of un-doped microspheres (Fig. 4.a) measured as 5.0 ± 0.2 $g.cm^{-3}$ (85.4 % T.D) and Nd(III) doped ones (Fig. 4.b) as 5.8 ± 0.1 $g.cm^{-3}$ (96.9 % T.D) by gas pycnometer.
- XRD is still required to study the crystallographic properties of un-doped and Nd-doped YSZ particles.
- N_2 gas adsorption analysis preliminary tests were carried out, but without successful results yet because of the small quantities. Further investigation is needed with higher sample mass for specific surface area analysis and porosity measurements on calcined microspheres.

Conclusion

- 600 °C as calcination temperature is not high enough to remove organic residuals originated from the gelation agents in the YSZ spheres, 900 °C is appropriate.
- The average density and % T.D of YSZ particles is increased and a blue color change was observed after infiltration with neodymium. Neodymium infiltration enhanced the densification.
- Un-doped and neodymium-doped UO_2 microspheres ($(U,Nd)O_2$) will be fabricated via internal gelation and characterized in the upcoming weeks with the aim of comparing them with $(Zr,Y,Nd)O_2$ microspheres and to study their size, porosity and density after sintering.

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