Fabrication of simulated americium transmutation targets via infiltration

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Introduction

The long-term radioactivity and heat load of spent fuel is mainly caused by the minor actinides (MAs). A possible solution to reduce the radiological hazard of spent fuel is to partition the MAs and to transmute them into lighter, short-lived elements in fast spectrum reactors.

Infiltration-produced MA loaded microspheres are a promising route and were successfully demonstrated for Am-loading of inert matrix matrices [Richter et al. 1997]. Uranium based matrices were less explored for infiltration and also loading with elevated Am contents proved to be difficult. In a previous PhD thesis, internal gelation (IG) was used for direct production of mixed oxide fuels [Schreinemachers et al. 2020]. Since IG may also offer an elegant route for the creation of porous microspheres enabling good infiltration [Pillon et al. 2003], this route was explored both with and without pore formers.

Objectives

- **Reducing the long-term radiotoxicity of spent fuel via recycling Am**
- **Developing innovative, dust free and remote fabrication routes for Am targets**
- **Fabricating UO_2 microspheres via internal gelation and Nd(III)-doping (surrogate of Am) via infiltration**
- **Obtaining single-phase $U_{1-y}Nd_yO_{2-x}$ targets**

Methods

Ammonium diuranate (ADU, $3UO_3{\cdot}2NH_3{\cdot}4H_2O$) microspheres are prepared via internal gelation. Further processing involves thermal treatment (calcination) to convert the microspheres into an oxide form. The effect of calcination temperature and use of pore-formers on the microstructure of the microspheres was investigated by gas adsorption and pycnometry, microscopy and X-ray diffraction. Nd(III)-doping of selected host microsphere matrices via infiltration was investigated and tuned. After infiltration, a re-calcination and sintering treatment was applied to obtain $U_{1-y}Nd_yO_{2-x}$ microspheres.

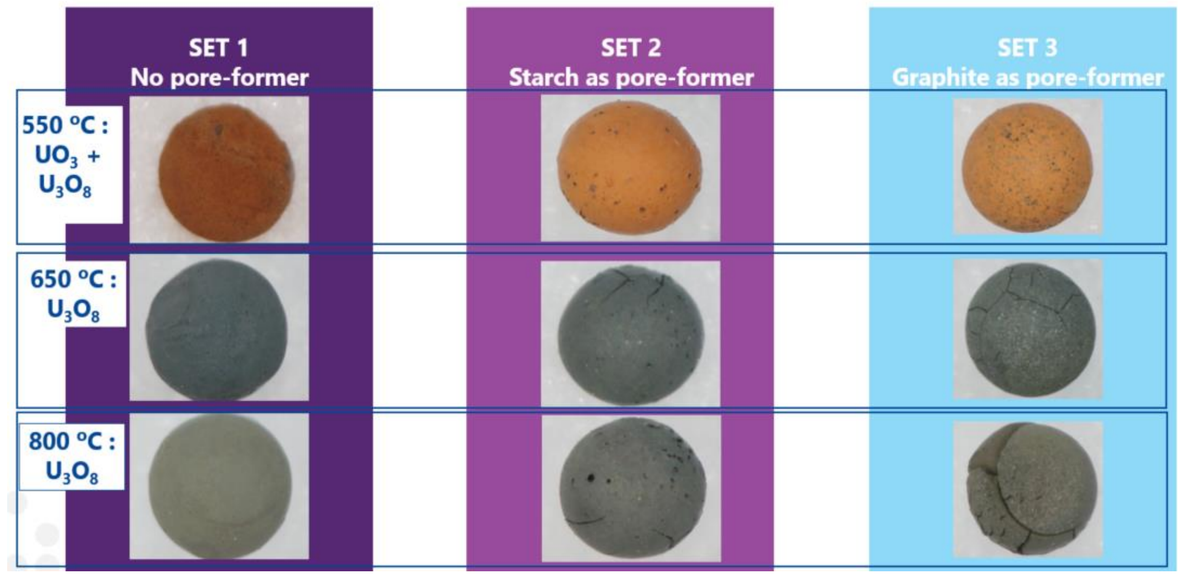

Figure 1. Optical microscopy pictures of infiltration host matrices; at 550 °C calcination the composition is mixture a of UO_3 and U_3O_8, 650 °C and 800 °C their compositions are U_3O_8 with & without pore-formers.

Results and Discussion

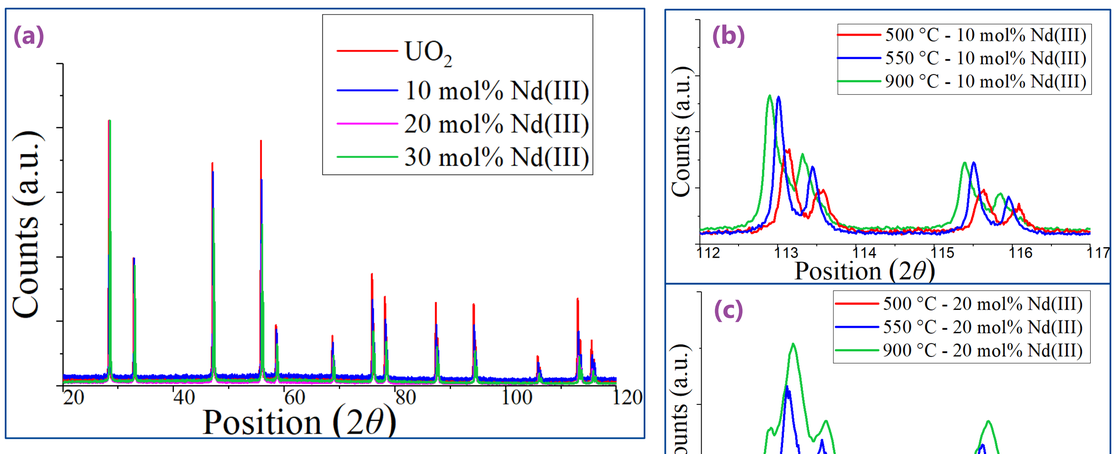

Figure 2. XRD diffractograms of infiltrated microspheres.
(a) Pure UO_2 and Nd(III)-doped UO_2. Comparison of high angle peaks of microspheres calcined at different temperatures and infiltrated with
(b) χ(Nd)=10 mol%,
(c) χ(Nd)= 20 mol%,
(d) χ(Nd)=30 mol%.

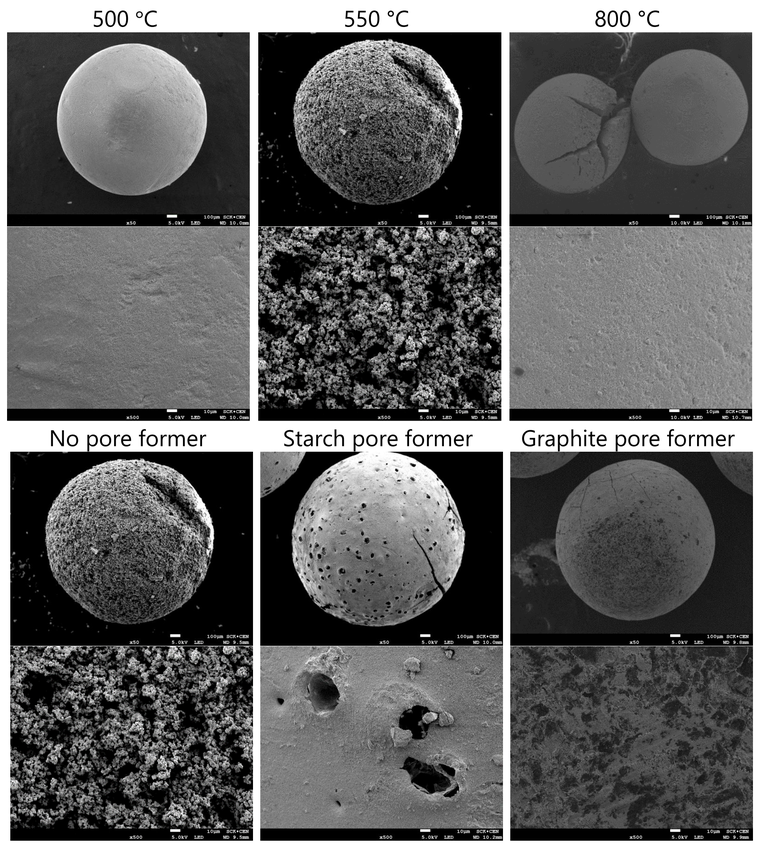

Figure 3. SEM micrographs of un-doped microspheres: (left) calcined at 500 °C, (middle) at 550 °C, (right) at 800 °C.

Figure 4. SEM micrographs of microspheres calcined at 550 °C: (left) without pore-former, (middle) with starch as pore-former, (right) with graphite as pore-former.

Table 1. Specific surface area (SSA) and density of calcined microspheres

T_{calc}	550 °C			650 °C	800 °C
Pore former	None	Starch	C	None	None
Composition	$UO_3 + U_3O_8$	$UO_3 + U_3O_8$	$UO_3 + U_3O_8$	U_3O_8	U_3O_8
SSA (m^2/g)	24.7 ± 0.1	18.9 ± 0.1	13.3 ± 0.1	10.8 ± 0.1	6.0± 0.1
$\rho_{apparent}$ (T.D%)	66.9	63.8	78.1	74.8	89.3
$\rho_{pycnometry}$ (T.D%)	87.8 ± 0.4	90.6 ± 0.5	90.2 ± 0.2	96.7 ± 0.1	97.6 ± 0.1

- As calcination temperature increases (Figure 3), more fine porosity is being removed from the microsphere, explaining the reduced SSA as function of calcination temperature (Table 1).
- According to SEM, pore former addition significantly effects microstructure (visible big pockets), however, SSA of microspheres without pore former addition stays the highest. (Figure 4).
- The infiltration treatments applied on 500 °C and 550 °C calcined microspheres appears much more effective for reaching the target dopant concentration (Figure 5) and obtaining single solid solutions (Figure 2).
- Lab infrastructure is being prepared to perform infiltration of Am(III) solution according to the present results.

Conclusion

- Infiltration process conditions were optimized for transmutation fuel target fabrication.
- Lower calcination temperatures (e.g. 500, 550 °C) seem most appropriate to obtain a porous microstructure, suitable for infiltration.
- Fabrication of Nd-doped (5-30 mol%) UO_2 microspheres via infiltration was achieved.
- Excellent sphericity and density microspheres have been obtained.
- The further development of the infiltration process is ongoing for $(U,Am)O_2$ production.

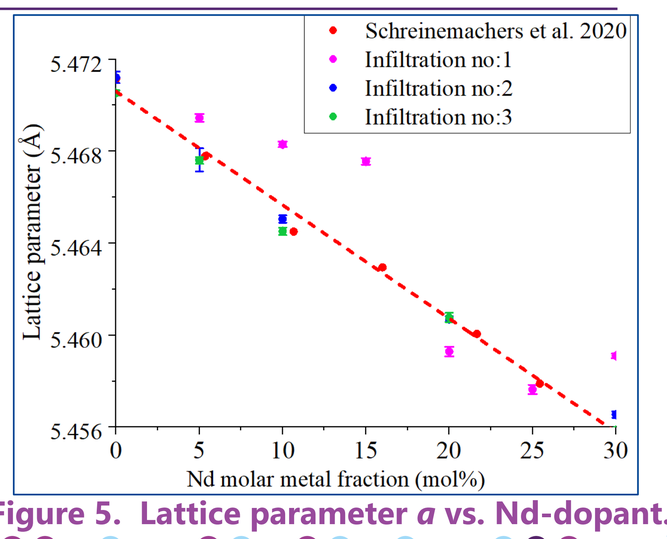

Figure 5. Lattice parameter *a* vs. Nd-dopant.

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