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Thoria based inert matrix fuel production via sol–gel process

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ABSTRACT

There is an important concern that uranium reserves will not be sufficient for new nuclear facilities in the future. Thorium could offer a potential alternative fuel because it is three times more abundant than uranium. Thorium based nuclear fuels are important candidates for the future of nuclear fuels. The main interest is in considering thorium based fuels for burning plutonium in conventional reactors. Thoria based inert matrix fuels (IMF) are also candidates for burning plutonium and the transmutation of minor actinides. In this study, inert matrix fuel production was examined by external gelation process. Metal nitrate solution of each material (thorium nitrate, cerium nitrate, aluminum nitrate and magnesium nitrate) were mixed for the feed solution. The metal nitrate solution was 50% of total feed solution. After mixing the metal nitrate, solution polyvinyl alcohol (PVA, 10%) and tetrahydrofurfuryl alcohol (THFA, 40%) were added into mixed metal nitrate solution during stirring. PVA, THFA and metal nitrate solution were dropped through a nozzle into gelation medium ammonia solution. Then external gelation process was carried out. Cerium (Ce) was used to simulate Plutonium (Pu). Obtained gels from the external gelation process were aged, washed and dried. The powders were calcinated at 1273 K for 6 h. After calcination, the powders were pressed at 500 MPa with a manual press. Pellets were sintered at elevated temperatures (1473, 1573, 1673, 1773, 1873, and 1973 K) for 4 h. The densities of sintered pellets were measured by immersion method. Sintered pellets characteristics were determined by scanning electron microscopy, x-ray diffraction analysis and energy dispersive x-ray analysis and the results were discussed.

1. Introduction

Ceramic nuclear fuels are fabricated conventionally through the powder metallurgical route in nuclear fuel production. Uranium oxide nuclear fuels have been used commercially in nuclear reactors. The nuclear fuels, for near future, must provide proliferation-resistance, less nuclear waste and prevent the increasing of plutonium stockpiles and minor actinides (MAs). An important option to reduce the excess plutonium is to utilize this plutonium in advanced fuel types, such as thoria-based IMFs. Because of these type of fuels are uranium-free fuel and the plutonium consumption rate is very high (Schram and Klaassen, 2007). In recent years, the investigations showed that thoria based nuclear fuels especially IMFs can solve these problems. In addition, the interest of thoria based nuclear fuels have been increased because of abundance of thorium and shortage of uranium (Pai et al., 2004). Thoria based inert matrix fuels have a great potential for more efficient destruction of plutonium and MAs relative to mixed oxide (MOX, U–Pu) fuels (Carmack et al., 2005). This type of fuel can consume 70–80% of the Pu whereas MOX fuels can reach only 20–30%. The best option for disposition and solve the problem of plutonium surplus is using in existing Water Reactors (IAEA-Tecdoc-1516, 2006). IMF, a promising strategy, could be used in the existing commercial light-water reactors. In near future fast neutron reactors (FRs) may also be considered for the effective incineration of minor actinides (Degueldre and Yamashita, 2003). We think that thoria based nuclear fuels especially IMF will be used to overcome the problem of excess Pu and MAs transmutation. At the other hand thoria based IMFs have multi-component ceramic structures. So the production of this type of multi-component ceramic oxide fuels are difficult to obtain homogeneous structure. Therefore sol–gel route were used to obtain the good quality powders and more homogeneous pellets that are required especially in multi-component ceramic structures. Also,

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Table 1
The percentage of final desired oxide composition.

Material	%
ThO_2	80
CeO_2	5
MgO	5
Al_2O_3	10

sol–gel based nuclear fuel fabrication is more amenable from powder metallurgical route to remotization and automation.

1.1. Sol–gel process for nuclear fuel fabrication

Sol–gel based processes have been developed for the fabrication of nuclear fuel materials (U, Pu, Th). This process has the advantage of being free from the use of radioactive powder that could get air borne and being amenable to remotization and automation. Sol–gel process involves gelation of the droplets of a sol (colloidal solution), or solution by hydrolysis resulting in condensation and polymerization of the desired fuel material into gel form (Nagarajan and Vaidya, 2012). In nuclear technology, the sol–gel process has been used to obtain homogeneous structure of U, Pu, Th oxides for pellet fabrication. Also, the sol–gel process has been used for the production of microspheres of ceramic nuclear fuels. The main areas for sol–gel application in nuclear fuel fabrication are plutonium fuel production, nuclear fuel for MA transmutation and fuel production for thorium based reactor systems. There are some advantages of sol–gel process such as not requiring of handling, suitable for remotization, fewer mechanical steps and homogeneous structure for multi-components. In this study, the production of Th based multi-component ceramic structure were aimed. In nuclear fuel, homogeneous structure is important for nuclear safety. The sol–gel process were preferred to obtain homogeneous structure.

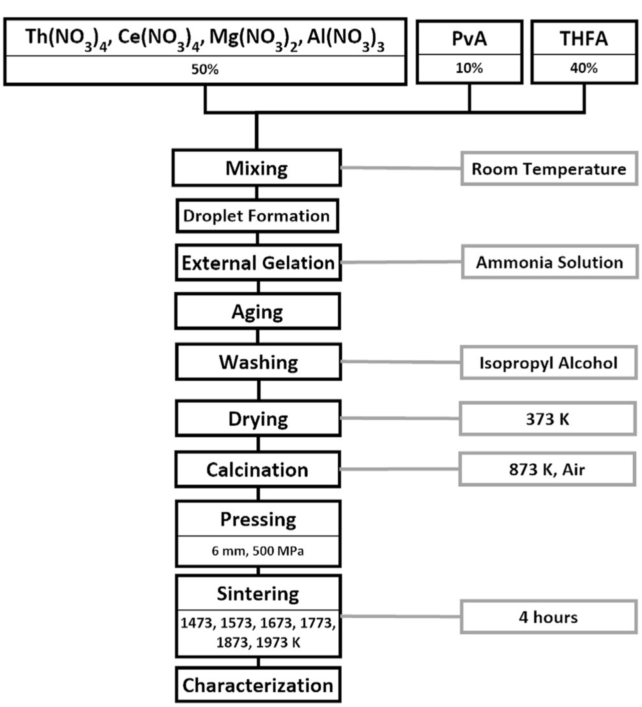

Fig. 1. The flow sheet of the external gelation process.

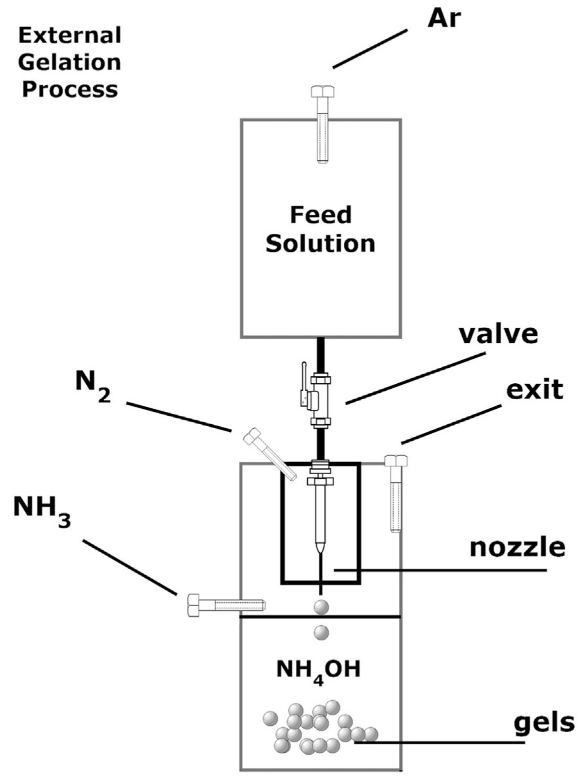

Fig. 2. External gelation process assembly.

1.2. Thoria based inert matrix fuel

Thorium is a viable alternative nuclear fuel to uranium for the near future. The use of thorium fuels can provide a safe, sustainable and proliferation-resistant source of nuclear power. Thorium can be mixed with radioactive waste from current uranium fueled reactors to reduce the stockpile of nuclear waste. Thorium oxide is an attractive matrix from a waste minimization point of view (Barrier et al., 2006). The actinide production is very low when using thoria as a support for burning plutonium. In addition, thorium dioxide is a chemically inert matrix so that it can be used as an inert matrix for final disposal (Hubert et al., 2001). Also, thorium dioxide is chemically more stable and has higher radiation resistance than uranium dioxide. The use of thoria based fuels requires the addition of fissile materials like uranium and plutonium. IMFs in other words, uranium-free fuels are a new initiative concept in which the uranium is replaced by elements not producing actinides under neutron irradiation (Matzke, 1999). The main object in IMFs is reaching higher burn-up for fissile atoms due to better behavior than standard fuels under neutron

Table 2
The geometrical densities of sintered ThO_2 and IMF pellets.

Temperature (K)	ThO_2 (g/cm^3)	IMF (g/cm^3)
1373	4.92	3.13
1573	4.89	3.17
1673	5.13	3.17
1773	5.25	3.20
1873	5.36	3.20
1973	5.50	3.23