Annular plutonium zirconium nitride fuel pellets

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One interest in the nuclear field is to have the possibility to fabricate specially shaped fuel pellets (e.g. barrel with a hole) to reach higher burn up's. Complex shapes can be realised with ceramics using the direct coagulation casting (DCC)[1] process, a relatively new shaping method for ceramics.
The direct coagulation process is based on the destabilisation of electrostatically stabilised ceramic suspension by time delay in situ reactions. Enzyme catalysed reactions can be used to created salt to increase ionic strength or/and to shift the pH of the suspension to the isoelectric point of the powder. In both cases the viscosity of the ceramic suspension increases and a wet green body is obtained.
Today the DCC method is well explored for alumina and other oxide powders.
The current project aims for using the DCC process to cast mixed plutonium zirconium nitride annular pellets. Zirconium nitride has been proposed as inert matrix material to burn plutonium or to transmute long-lived actinides in accelerator-driven sub-critical systems or fast reactors[2]. Because of the high thermal conductivity, the high melting point and the hardness of zirconium nitride, the material seems to be predestined for an inert fuel matrix.
To yield the mixed nitride three different routes were investigated. The first route is based on the production of plutonium zirconium oxide microspheres with additions of carbon black, using the well-known sol-gel method. The oxide spheres will be converted into the nitride by carbothermic reduction under nitrogen atmosphere.
The second route uses directly a mixture of plutonium oxide powder, zirconium oxide powder and carbon for the carbothermic reduction.
The third possibility is the use of commercial zirconium nitride, which will be mixed with plutonium nitride produced by carbothermic reduction.
The DCC method will be applied the first time in nuclear field to cast this kind of materials. As enzymatic hydrolysis reaction we use the urea/urease system, which produces carbon dioxide and ammonia in water, which can all be removed during calcination. But before two major problems have to be solved. The recepies of the colloid chemistry for the DCC process had to be adjusted as the enzyme activity suffers by the irradiation of the plutonium.
For this project a joint doctoral thesis between CEA Cadarache, ETH Zurich and PSI has been started mid of 1999, with the goal of fabricating annular (Pu, Zr)N pellets, which will be characterised by XRD, chemical analysis, ceramography, physical properties.
We will report about the production of the first nitride microspheres containing plutonium and the preliminary results of the colloidal forming technique DCC using commercially available zirconium nitride powder.

1 Gauckler, L.J., T.J. Graule, and F.H. Baader, *Ceramic forming using enzyme catalysed reactions.* Materials Chemistry and Physics, 1999. **61**: p. 78-102.
2 Burghartz, M., et al., *Some aspects of the use of ZrN as an inert matrix for actinide fuels.* Journal of Nuclear Materials, 2001. **288**: p. 233-236