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CANDU FUEL: *Evolution towards Optimal Performance*

Spark plasma sintering of uranium nitride fuels

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ABSTRACT – The commercial application of uranium nitride fuels is currently considered by Rosatom, Westinghouse and other actors in the nuclear community. Historically,issues related to manufacturability, swelling, compatibility with water and cost of nitrogen-15 haveconstituted showstoppers for this development to cometo fruition.

Recently, spark plasma sintering has proven to be a viable route formanufacturing of high density uranium nitride pellets with adequatemicrostructure. Moreover, the ability of the SPS process to deliver industrialscale throughput now appears to be within reach.

In this paper, we review the progress made in spark plasma sintering of uraniumnitride fuel at KTH and discuss the commercial application of this process tomanufacturing of fuels for the Canadian market.

Introduction

The use of high density uranium nitride fuel (UN) significantly improves neutron economy, due to the 40% higher density of actinides that can be accommodated in the same fuel rod, as compared to UO_2. This feature may allow to improve the breeding ratio of fuels for fast reactors, or to increase residence time of fuel for water cooled reactors [1].

Whereas UN fuels have been used in liquid metal cooled fast reactors, such as BR-10 in Russia, and foresees to use (U,Pu)N in BN-800, BREST-300, as well as in BN-1200, the application of nitride fuels in water cooled reactors has never been realised. The two major reasons are

- The poor tolerance of uranium nitride when exposed to water and steam [2, 3].
- The cost for enrichment of N-15, which is necessary to avoid neutron absorption in N-14.

Since it is impossible to guarantee that all fuel rods in a power reactor remain intact during operation, a nitride fuel used for commercial power production must feature an improved resistance to corrosion by steam. Westinghouse is working on developing a water tolerant nitride fuel for light water reactors [4]. Currently, this effort is carried out within a DOE funded project and the global CARAT consortium. In this paper, we will discuss a novel method for manufacture of UN pellets andthe commercial application of UN fuelin CANDU reactors.

1. UN fuel in CANDU reactors

The application of UN fuel to CANDU reactors has not been considered in the open literature, albeit one may imagine that the work of AECL on corrosion tolerance of UN in water was motivated by the known advantages of using UN fuel [3]. We have used the Monte Carlo code Serpent in conjunction with the JEFF3.1 nuclear data library and s(α,β) functions for in-coherent scattering of thermal neutrons on D_2O, to assess the neutronics of a CANDU reactor using a uranium nitride fuel. The calculation was made for an infinite lattice of such assemblies, thus neglecting effects of leakage.

Figure 1 shows thecalculated evolution of k-infinity in a 37 rod CANDU bundle fuelled with natural uranium nitridehaving a density of 14.0 g/cm^3 = 98% TD, as compared with natural UO_2 (density of 10.6 g/cm^3 = 97% TD). It was assumed that the nitrogen is 99% enriched in nitrogen-15. Whereas standard residence time for UO_2 is 210 days, this simulation indicates that the service life of UN is approaching 340 days, equivalent to a gain in residence time of 60%!

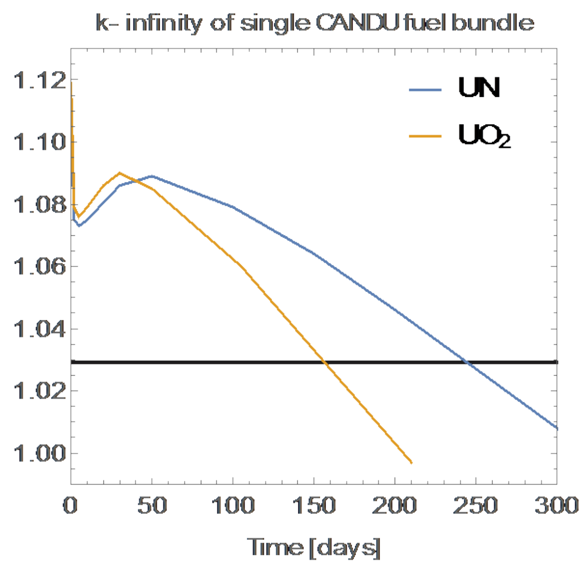

Figure 1: Evolution of reactivity in UO_2 and UN CANDU fuel bundles up to a burn-up of 7150 MWd/kg.

Such an improvement in performance means that the number of fuel bundles that have to be fabricated for a CANDU reactor is reduced by 37. Note that there is no savings in the amount of uranium used, since every fuel bundle contains 40% more uranium. It is remarkable that the extention of the residence time is considerably higher than the increase in uranium density, but a similar phenomenon is observed when studying slightly enriched UO_2 fuel, where reactivity increases considerably more than the relative increase in enrichment. Another issue to be noted is that k-infinity for a fresh fuel UN is very close to the the value found for UO_2. The reason for this is a reduction in cross sections for fission (and capture) resulting from the slightly harder neutron spectrum in the UN bundle (see Table 1). Hence, there is no need to adjust existing reactivity control systems.

Nuclide	$\sigma_f(UO_2)$	$\sigma_c\ (UO_2)$	$\sigma_f(UN)$	$\sigma_c\ (UN)$
U-235	131.3	24.0	102.8	19.2
U-238	0.1	1.0	0.1	0.8
Pu-239	298.5	158.6	232.4	123.7
Pu-240	0.4	152.1	0.4	121.2

Table 1: Spectrum averaged cross sections, in units of barn, for fission and capture in CANDU reactors with UO_2 and UN fuel.

2. Safety

A novel nuclear fuel must not only provide an economical incentive to motivate its introduction, is should also feature improved safety characteristics. This is of particular concern in CANDU reactors, where the positive power coefficient of the fuel leads to the reliance on active shut-down mechanisms in case of power excursions. In this context, the major advantage of introducing uranium nitride fuels is the very high thermal conductivity of UN. Figure 2 compares the conductivity of UN with that of UO_2. The resulting difference in temperature between the centre-line and the surface of the fuel pellet is seven times smaller for UN, improving the margin to failure of the fuel by 1300 K.

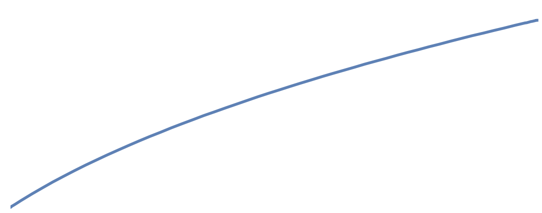

Figure 2: Thermal conductivity of UN and UO_2.

A safety concern that may be raised when introducing UN fuels is the potential production of hydrogen during water ingress in a damaged fuel rod according to:

$$3UN + 8H_2O \rightarrow U_3O_8 + 3NH_3 + \frac{7}{2}H_2$$

KTH has carried out tests were UN pellets were exposed to overheated steam at T = 425°C, showing that the rate of the above process can be reduced by fabricating pellets with zero open porosity [5]. Thus, the surface area available for attack by steam is minimised, and pellet integrity is improved.

It can be noted that UN is very difficult to fabricate without open porosity if conventional sintering methods are used. Applying spark plasma sintering [6], zero open porosity is achieved when total porosity of the pellet is less than 2.5%. Whereas such porosities would not permit use of the fuel in an LWR, where burn-up targets are exceeding 4%, they fuel may well be used in CANDU reactors, where the maximum fuel burn-up is less than 2%.

3. Spark plasma sintering

Figure 3 shows the density of UN achieved when manufacturing pellets using spark plasma sintering, applying different temperatures and pressures [6]. By sintering at 1600°C during three minutes, densities above 98% are achieved when the applied pressure exceeds 90 MPa. It can be noted that using conventional pressureless sintering, a sintering temperature of 2500°C is required to achieve such densities for UN [7].

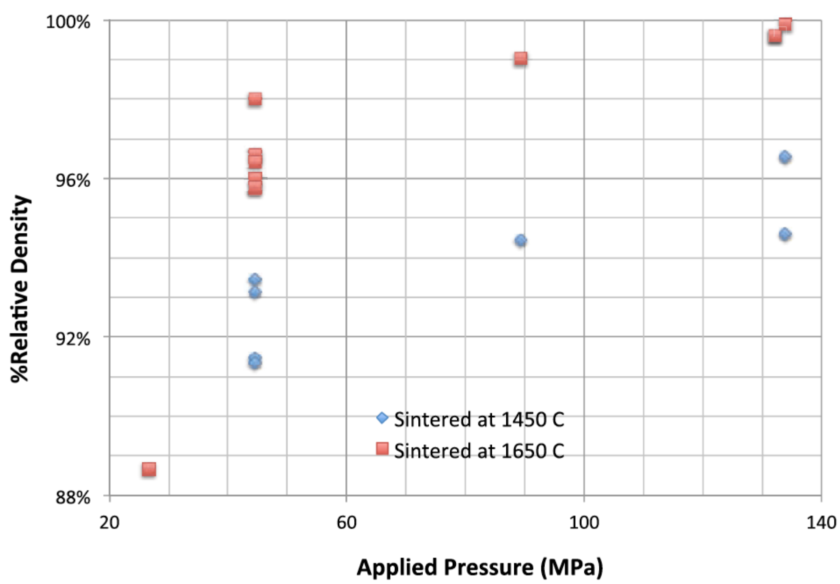

Figure 3: Sintering map for UN using SPS, as a function of applied pressure, temperature, and attained %TD.

Figure 4 displays the fraction of open porosity in UN pellets sintered with SPS as function of their total porosity. As already discusssed, full pore closure is achieved when the total porosity is less than 2.5%.

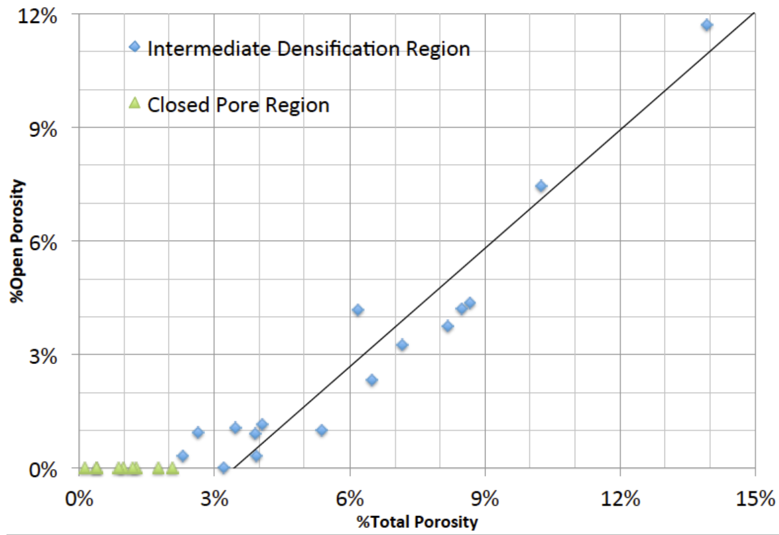

Figure 4: Relationship between open porosity and total porosity for UN pellets sintered with SPS.

Wheras the spark plasma sintering facility used in the fabrication process of KTH allows to sinter only about 10 pellets per day, commercial providers today offer SPS machines with a capacity that is two order of magnitudes higher. Considering that a CANDU fuel bundle contains about 1300 pellets and that the use of UN fuel would reduce the number of bundle insertions in a CANDU reactor to about ten per day, 1-2 high performance SPS machines may be sufficient for accomplishing manufacture of the fuel for this reactor.

4. Conclusions

Spark plasma sintering has been shown to provide a means to fabrivate UN fuel pellets with properties suitable for us in CANDU reactors. The application of UN fuel in CANDU reactors could permit to reduce the number of fuel bundles necessary to fabricate by up to 37%. Savings in fuel and waste management procedures may, in addition to improved safety characteristics, consitute an incentive to introduce this fuel on a commercial basis.

5. References

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