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Oxidation Protection of Uranium Nitride Fuel Using Liquid Phase Sintering

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The advantages (high thermal conductivity, very high melting point, and high density) of nitride fuel have long been recognized. The sodium cooled BR-10 reactor in Russia operated for 18 years on uranium nitride fuel (UN was used as the driver fuel for two core loads). However, the potential advantages (large power up-grade, increased cycle lengths, possible high burn-ups) as a Light Water Reactor (LWR) fuel are offset by uranium nitride's extremely low oxidation resistance (UN powders oxidize in air and UN pellets decompose in hot water). Innovative research is proposed to solve this problem and thereby provide an accident tolerant LWR fuel that would resist water leaks and high temperature steam oxidation/spalling during an accident.

It is proposed that we investigate two methods to increase the oxidation resistance of UN: (1) Addition of USi_x (e.g. U_3Si_2) to UN nitride powder, followed by liquid phase sintering, and (2) "alloying" UN nitride with compounds (followed by densification via Spark Plasma Sintering) that will greatly increase oxidation resistance.

<u>Liquid Phase Sintering Using molten USi_x</u>

Liquid phase sintering requires a molten second phase the wets and spreads on powder surfaces within a compact. Wetting requires a slight solubility of the powder in the liquid phase. Densification proceeds by particle rearrangement followed by solution-reprecipitation[1]. If there is sufficient liquid phase present, a liquid network is formed that surrounds every grain of UN. This approach has been demonstrated by liquid phase sintering of UO_2 using a corrosion-resistant alumino-silicate[2,3]. The resulting composite structure (See Figure 1) proved to be extremely resistant to steam oxidation and chemical corrosion[4] whereas pure sintered UO_2 swelled and cracked when exposed to steam.

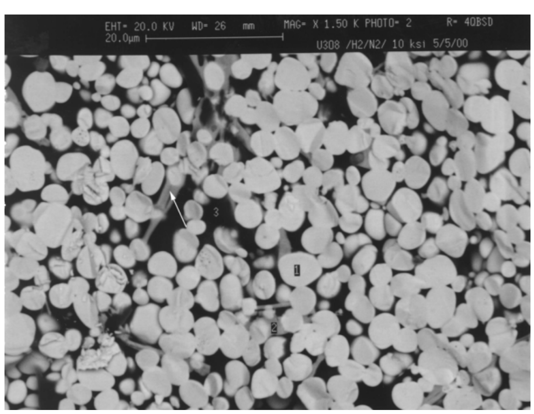

Figure 1. Backscatter SEM micrograph of Liquid-Phase sintered UO_2 grains (light colored area) surrounded by corrosion resistant alumino-silicate second phase (dark colored area).

The compounds selected[5] for study as a liquid phase formers for UN are U_3Si_2 (has been studied and used as nuclear fuel), USi, and U_3Si_5. These compounds are high melting (1665-1770 ^oC), can contain U^{235} enrichment, have good thermal conductivity, and will form high purity SiO_2 when exposed to air or steam at the surface of the fuel pellet.

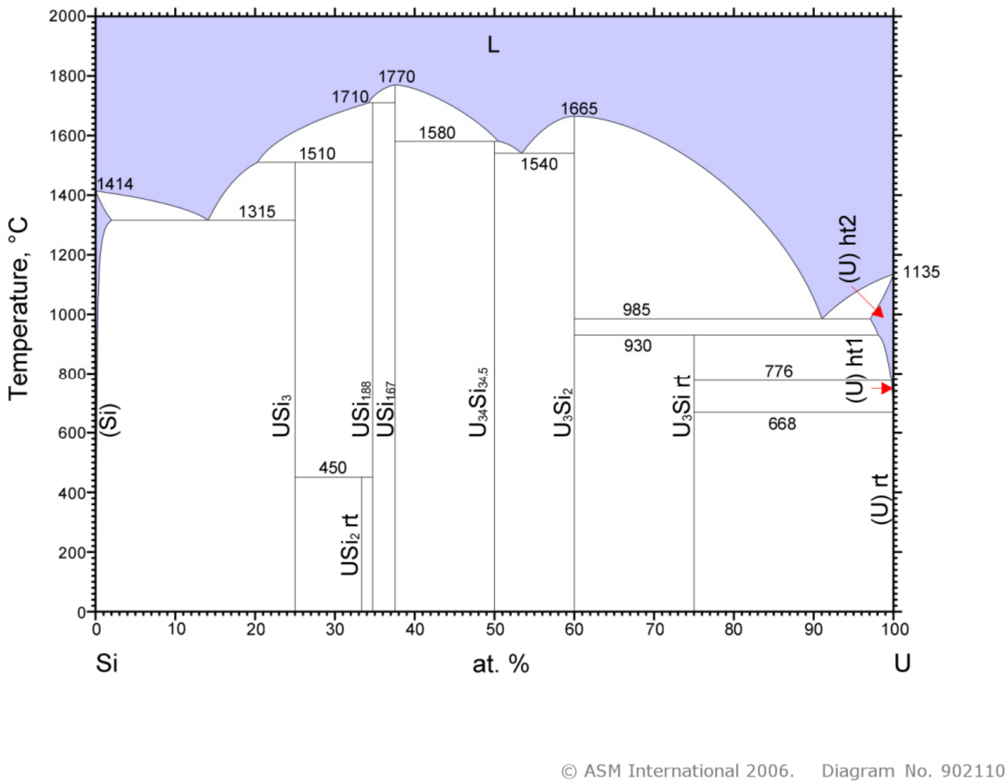

Figure 1. U-Si phase diagram showing melting points of various refractory phases.

<u>Alloying to provide for Oxidation Resistance</u>

Alloying of UN with ZrN and producing 94% dense pellets has been demonstrated[6] in Sweden using Spark Plasma Sintering (SPS) of alloyed powders. A pellet of SPS-fabricated UN is shown in Figure 3. Pellets of pure ZrN demonstrate a much higher but not complete resistance to oxidation – this is presumed to be due to a formation of a zirconia surface layer. Research in Sweden is ongoing to determine the oxidation resistance of UN-ZrN pellets.

Figure 3. UN pellets (left) fabricated by Spark Plasma Sintering (right) at KTH in Sweden.

We propose to investigate alloying of UN with CrN and AlN and subsequently densify using SPS. Chromium nitride (CrN) is known to be extremely resistant to corrosion. Steam or air oxidation of the CrN content will form protective Cr_2O_3 surface films analogous to those formed by steam oxidation of Cr-alloyed stainless steels[7,8]. It is thought that the addition of small amounts of AlN in the pellet will aid adhesion and oxidation resistance of the protective film[9] at very high temperatures. Also, adhesion could be improved by the presence of SiO_2 that could be provided by adding some USix (as noted above).

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