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# Enigma of the “U<sub>8</sub>Si<sub>8</sub>O” phase

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Using U<sub>3</sub>Si<sub>2</sub> as a nuclear fuel for research and test reactors requires a thorough knowledge of the U-Si phase diagram to predict reliably the in-use fuel behavior. U<sub>3</sub>Si<sub>2</sub> seems to be a perfectly stoichiometric compound. Industrial manufacturers use slightly over-stoichiometric in silicon compositions, to avoid the presence of residual  $\alpha$ -U or U<sub>3</sub>Si that affect the irradiation behavior of this fuel. Consequently, a secondary phase with atomic ratio close to 50%U-50%Si is present in the industrial fuel plates. From the binary phase diagram [1], the thermodynamically stable compound is U<sub>34</sub>Si<sub>34.5</sub>. Nevertheless, our investigations on both laboratory and industrial samples by electron backscattered diffraction (EBSD) revealed the presence of the tetragonal U<sub>34</sub>Si<sub>34.5</sub> as well as the orthorhombic “U<sub>8</sub>Si<sub>8</sub>O” phases in arc-melted samples [2]. The latter was first described as USi [3], but later studies suggested that its stability was achieved only by adding some oxygen in the structure [4]. The presence of light elements (mainly carbon) in metallic uranium may justify the formation of this pseudo-binary phase during the synthesis of Si-rich U<sub>3</sub>Si<sub>2</sub>. So why does this “U<sub>8</sub>Si<sub>8</sub>X” (X = C, O) phase formation depend on the cooling rate after melting? Why does it decompose upon annealing?

To answer these important questions and to understand the phase equilibria in the U-Si system, experimental and theoretical studies were undertaken. On the one hand, phonon calculations predict instability of the binary “USi” at 0 K, but its stabilization if a small amount of C is added. On the other hand, the quantity of “USi” increases with Si-content in rapidly-cooled arc-melted samples, from 41 to 46 at.% Si, and then drops giving way to U<sub>34</sub>Si<sub>34.5</sub>, the latter being the only visible phase (aside U<sub>3</sub>Si<sub>2</sub>) from 48 at.% Si. It should be noted that U<sub>5</sub>Si<sub>4</sub> was not been observed, except when C was intentionally added (U<sub>20</sub>Si<sub>16</sub>C<sub>3</sub>).

A summary of results obtained from various samples we synthesized will be presented, including ab-initio phonon calculations, X-ray diffraction and EBSD analyses, and scanning and transmission electron microscopies. In particular, the hypothetical stabilization origin (kinetic vs. thermodynamic phases, epitaxy...) of the “USi” phase will be discussed, highlighting the open questions that remain around that topic.

## References

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