



From arc-melted ingot to MTR fuel plate: a SEM/EBSD microstructural study of U₃Si₂

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From arc-melted ingot to MTR fuel plate: a SEM/EBSD microstructural study of U_3Si_2

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O. Fiquet¹, E. Castelier¹, M. Pasturel²

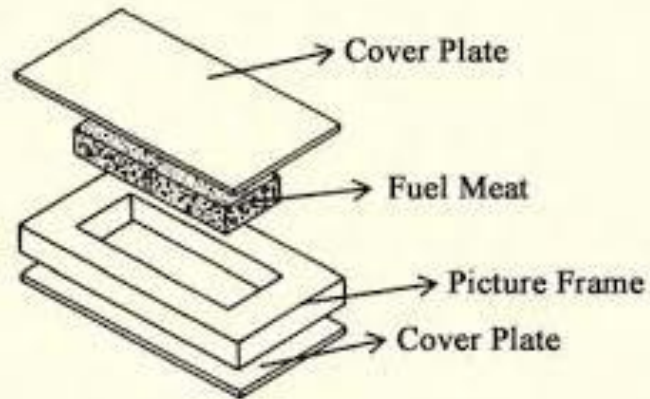
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50^{èmes} Journées des Actinides – 24/03/2021

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Introduction

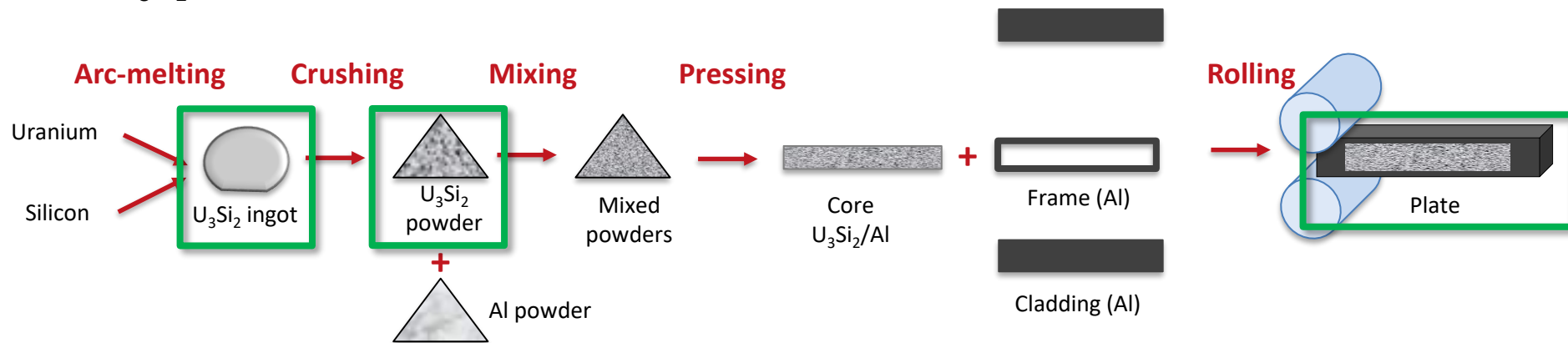
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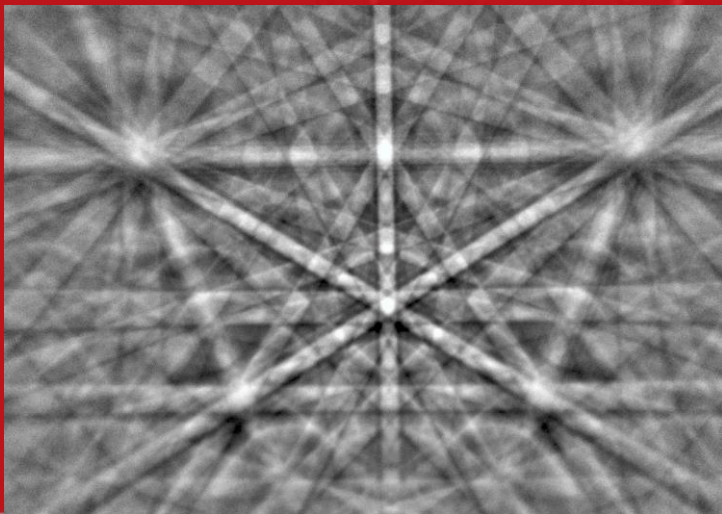
- Conversion of Research Reactors to Low Enriched Uranium (<20% ^{235}U).
- Development of new nuclear fuels with higher uranium density, for high performance MTRs.
- U_3Si_2 is one of the main candidates.

<i>Fuel</i>	<i>Melting point (°C)</i>	<i>Physical density (g cm⁻³)</i>	<i>Uranium loading (g cm⁻³)</i>
U	1133	19.1	19.1
U-7Mo	1145	18.4	17.1
U-10Mo	1150	18.2	16.4
U ₆ Mn	726	17.8	17.1
U ₆ Fe	815	17.7	17.0
U ₃ Si	930	15.6	15.0
U₃Si₂	1665	12.2	11.3
USi	1580	10.96	9.8
UAl ₂	1590	8.1	6.6
UAl ₃	1350	6.8	5.0
UAl ₄	731	6.1	4.2
U _{0.9} Al ₄	641	5.7	3.7
UAl _x	NA	6.4	4.5

- Material Testing Reactors (MTRs) → mainly nuclear fuel plates.
- $\text{U}_3\text{Si}_2/\text{Al}$ fuel plate manufacturing process



- Microstructural characteristics before irradiation important to understand the in-pile behaviour.**
- EBSD is an efficient technique.
- U_3Si_2 microstructure reinvestigated after the 3 main steps of the manufacturing process.



Experimental methods

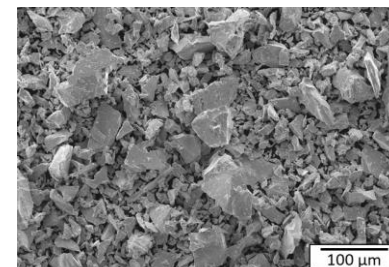
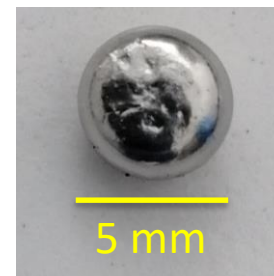
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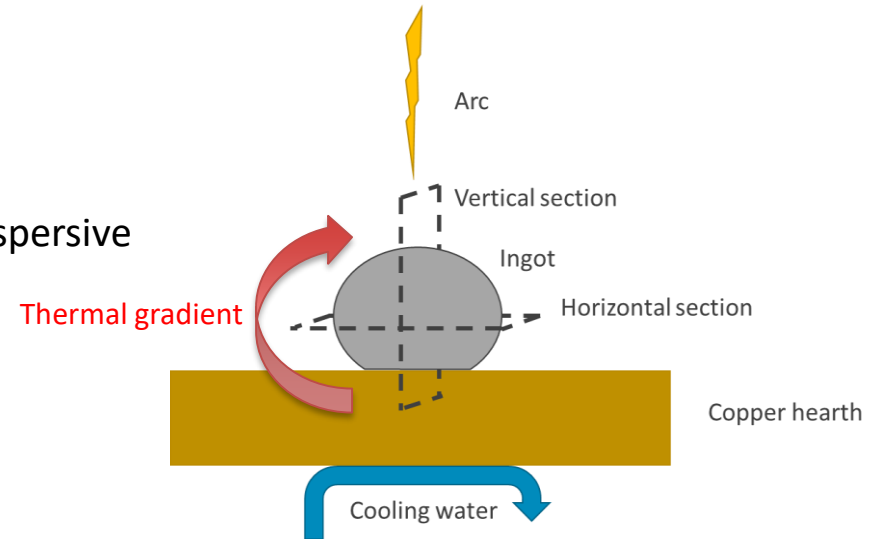
- **$\text{U}_3\text{Si}_2/\text{Al}$ fuel plate**
 - Framatome-CERCA production
 - Fabrication in the frame of the CEA SHARE irradiation test (Leenaers *et al.*, 2008)
- **Arc-melted ingots and crushed ingots**
 - Laboratory (CEA-Cadarache) production

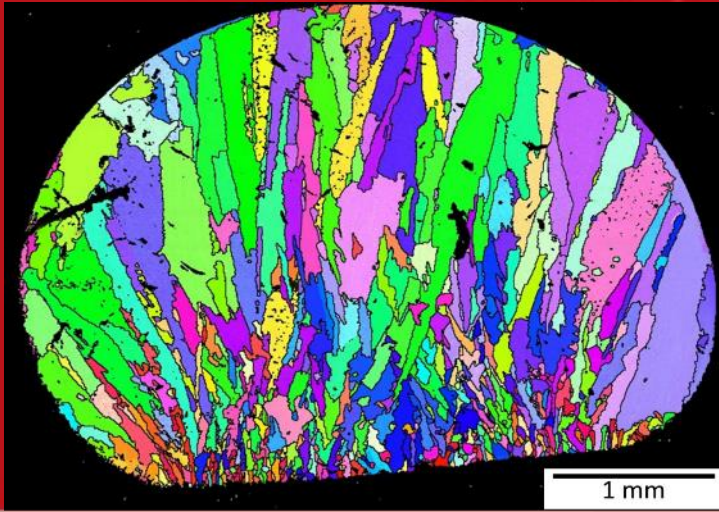


Label	Cooling	U/Si atomic ratio	Weight (g)
I1	"Very fast"	60/40	0.5
I2	"Slow"	60/40	1
I3	"Fast"	60/40 "low purity U"	1
I4	"Fast"	58/42	1



- X-Ray Diffraction (XRD)
- Scanning Electron Microscopy (SEM) + Energy Dispersive Spectroscopy (EDS)
- Electron BackScattered Diffraction (EBSD)

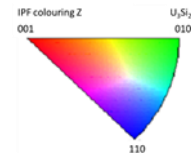
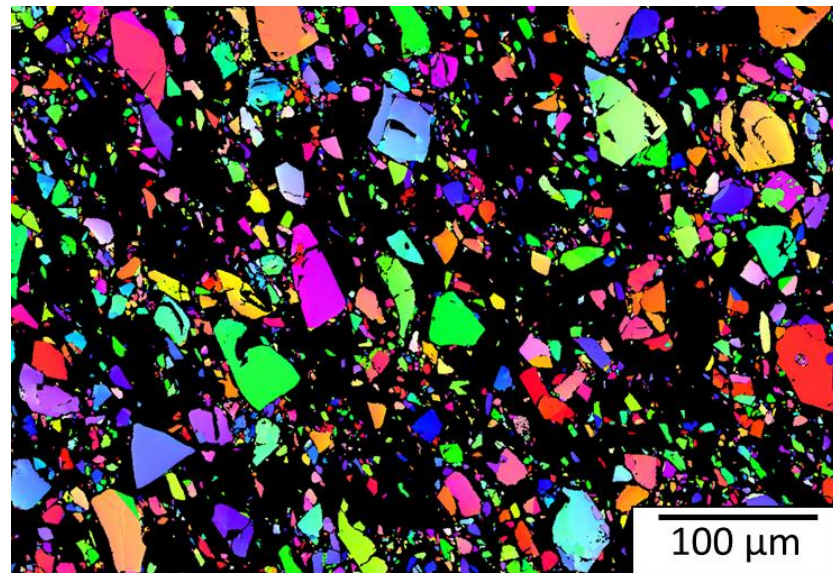
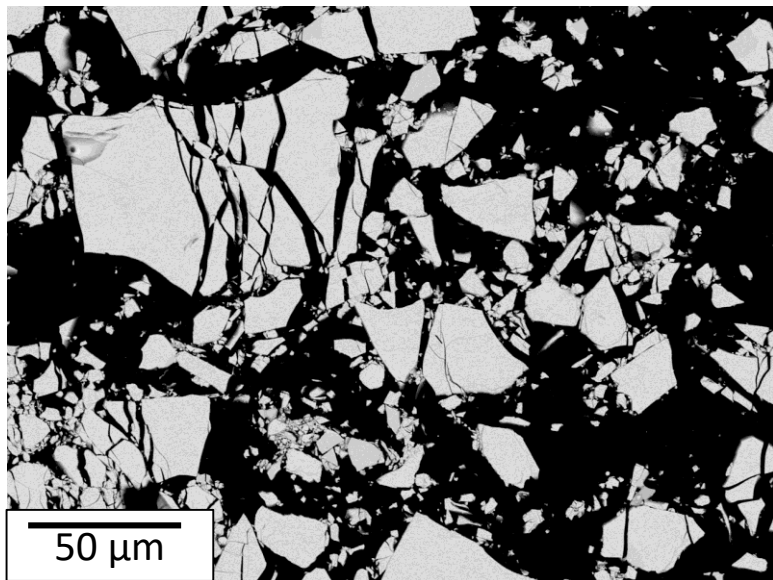




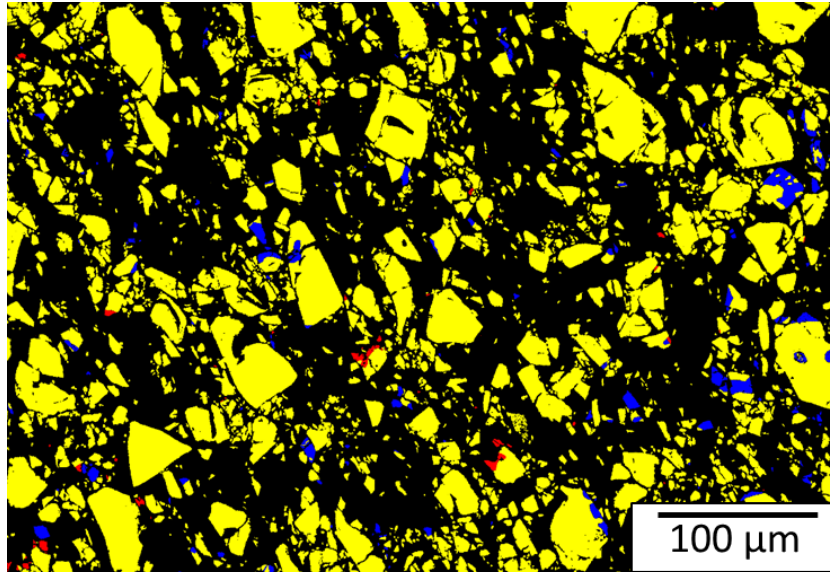
Results and discussion

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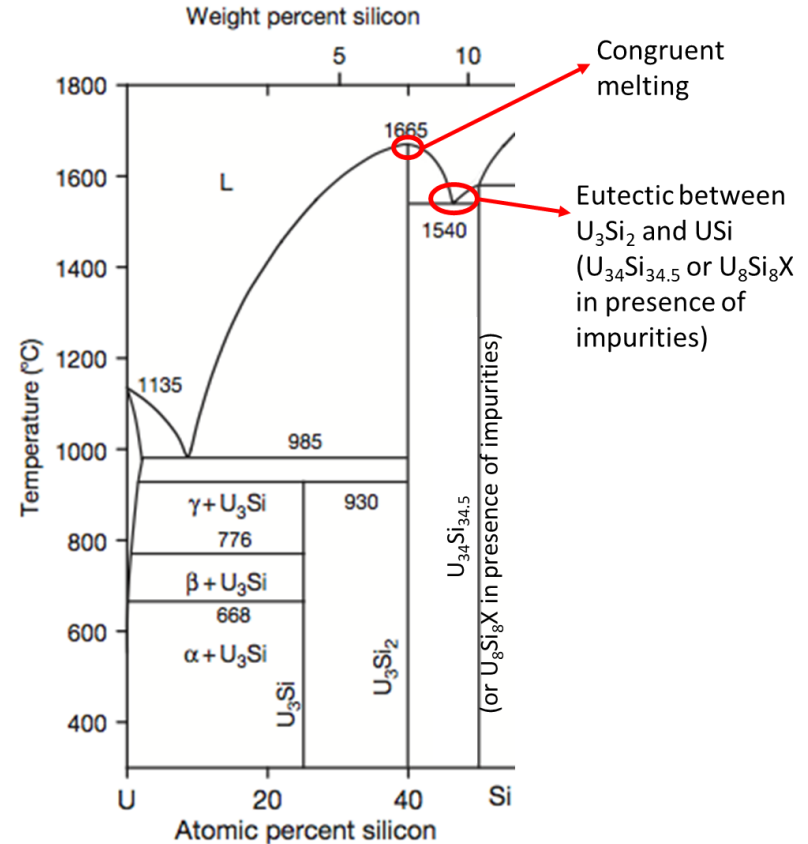


- Important fragmentation of particles.
- Single crystalline particles.



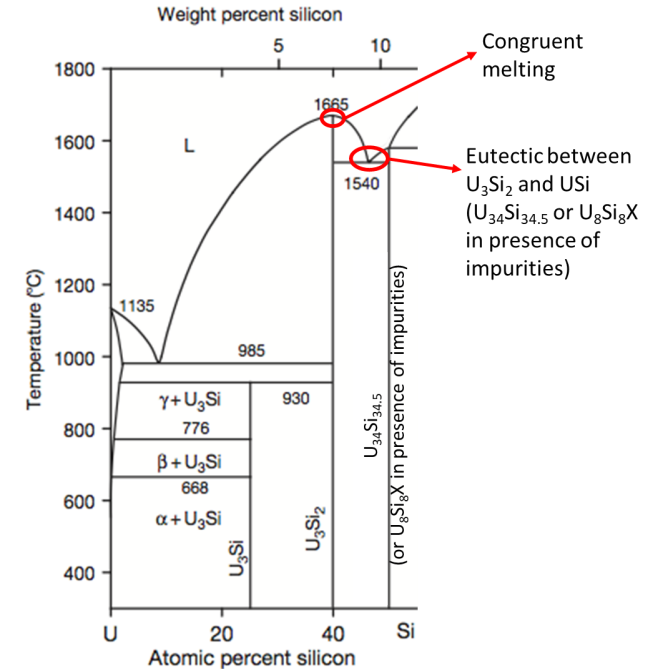
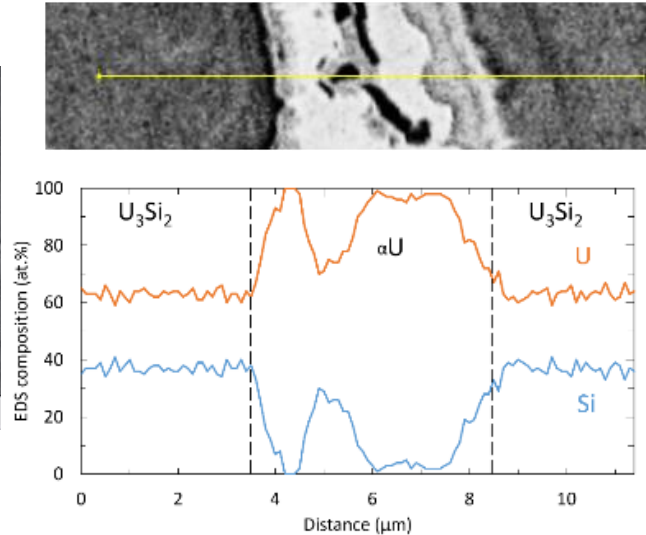
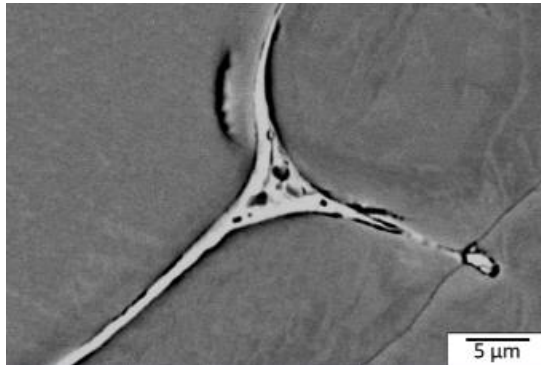
Map coloured in phase highlights the presence of Si rich secondary phases :

- $U_{34}Si_{34.5}$ (3.5%)
- U_8Si_8X (0.5%)

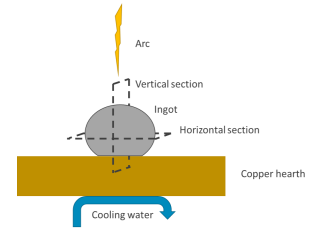
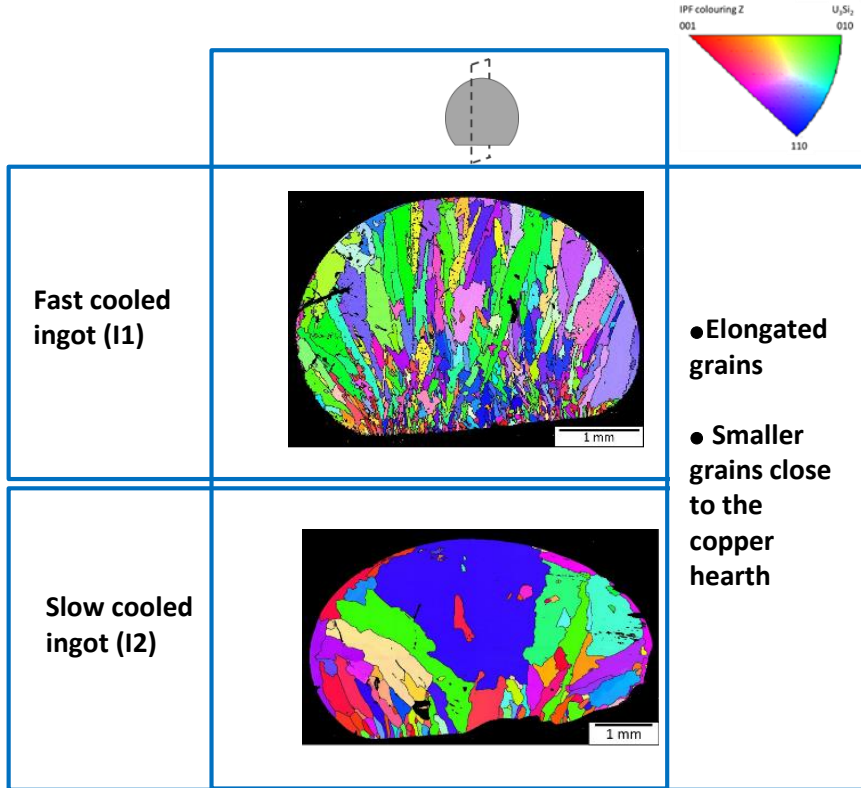


SEM characterizations

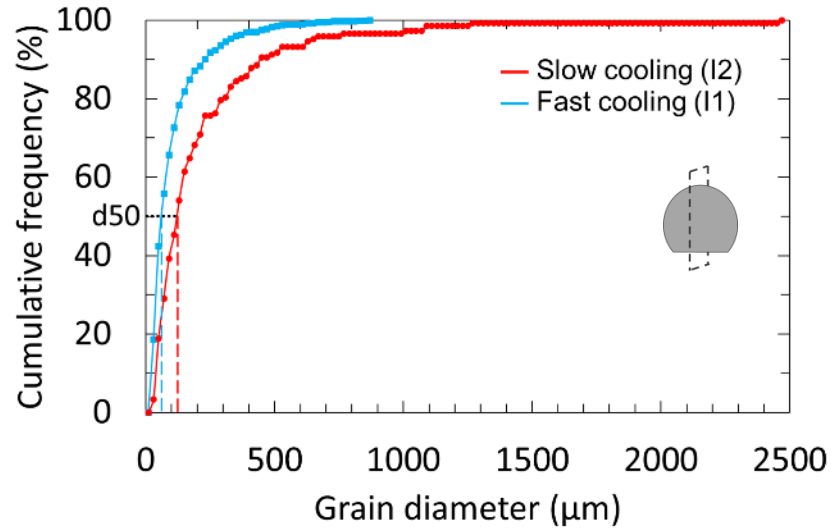
- α U at grain boundaries.



Electron Backscattered Diffraction

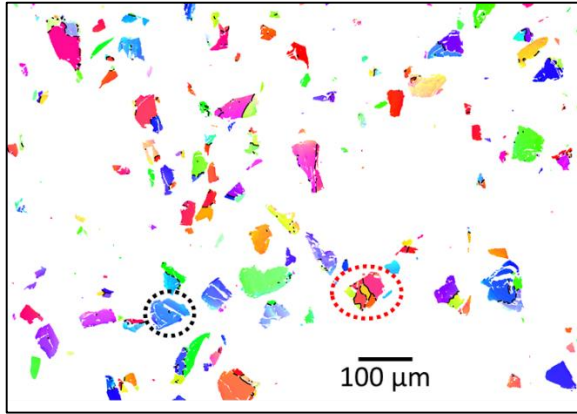


Grains size analysis

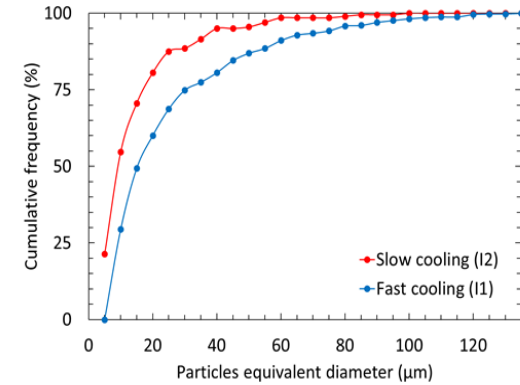


Slow cooling favours grain growth.

I1: fast cooled ingot



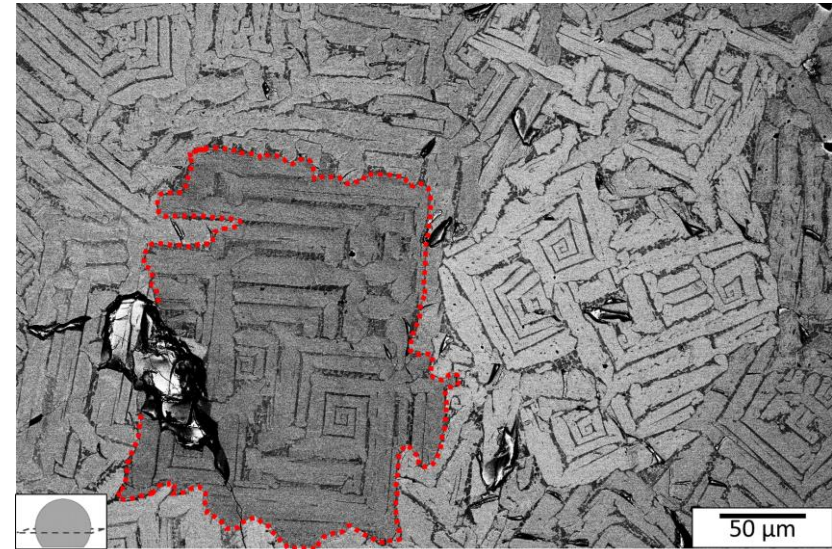
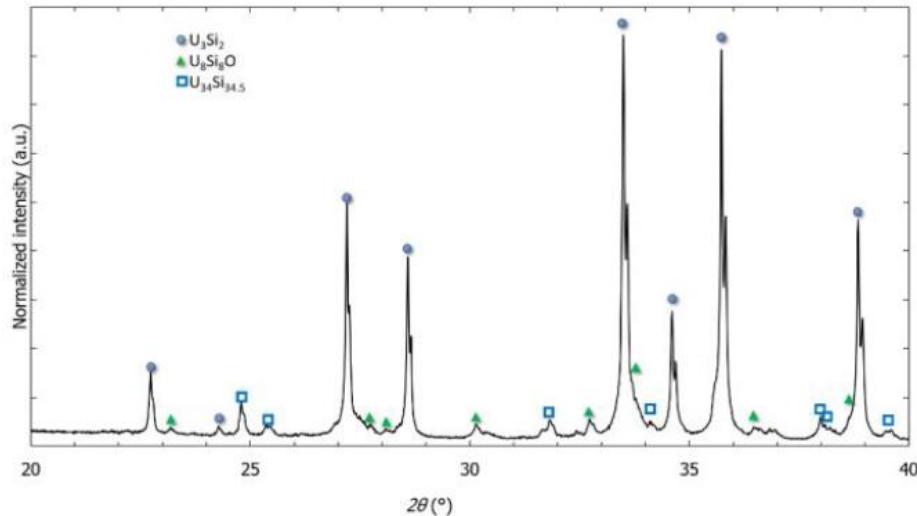
I2: slow cooled ingot



- Mainly single crystalline particles.
- Grain boundaries in particles → intragranular fragmentation (as evidenced by Metzger *et al.*, 2017).
- **Influence of fast cooling:**
 - U_3Si_2 grains are smaller in the ingot
 - U_3Si_2 particles are larger after a given crushing sequence → influence of the higher amount of grain boundaries on the mechanical resistance?

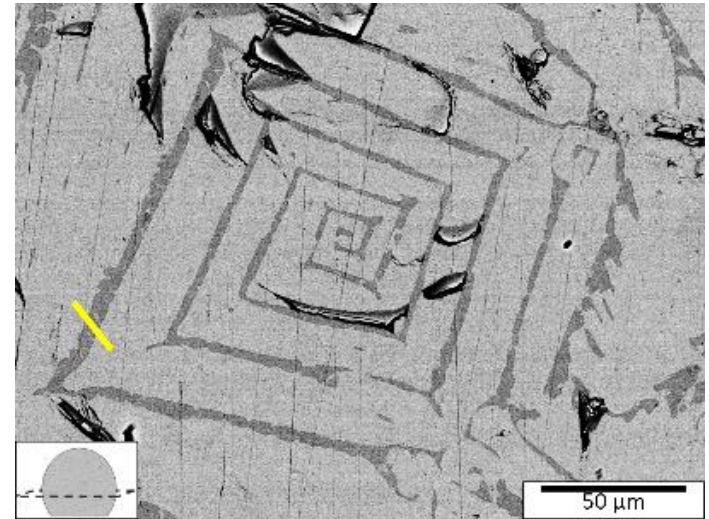
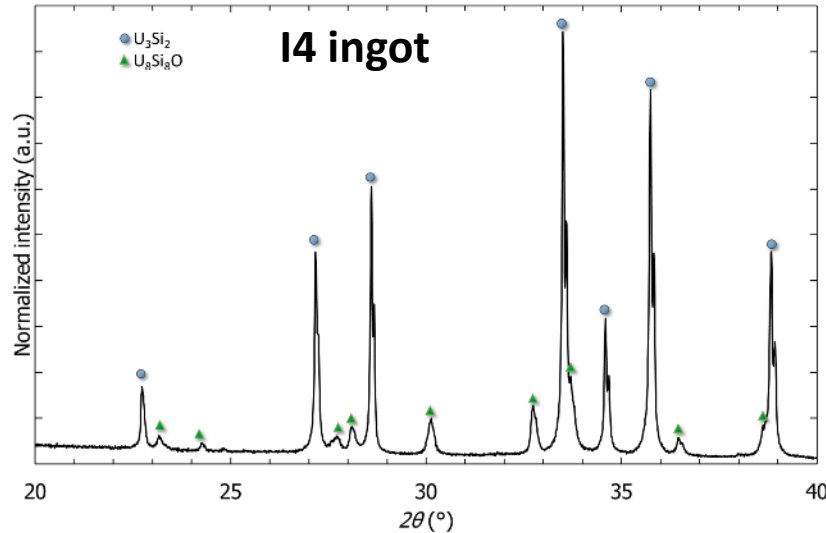
- Secondary phases indexed :
 - $\text{U}_8\text{Si}_8\text{X}$
 - $\text{U}_{34}\text{Si}_{34.5}$ (Le Bihan *et al.*, 1996)
- Solidification in spiral shape

I3 ingot

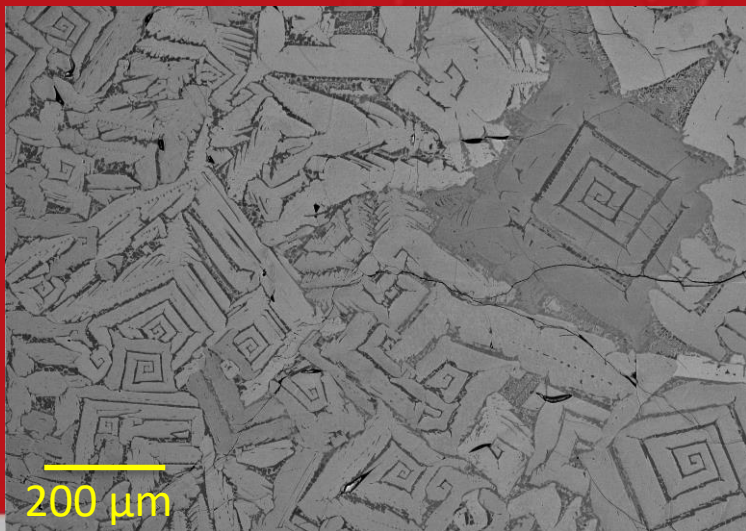


2%at. Silicon excess

- Secondary phase indexed as $\text{U}_8\text{Si}_8\text{X}$.
- Solidification in spiral shape.



→ Low purity uranium leads to an overestimation of uranium and deviation to stoichiometry



Conclusion

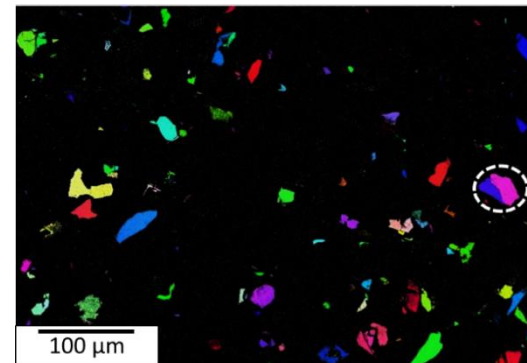
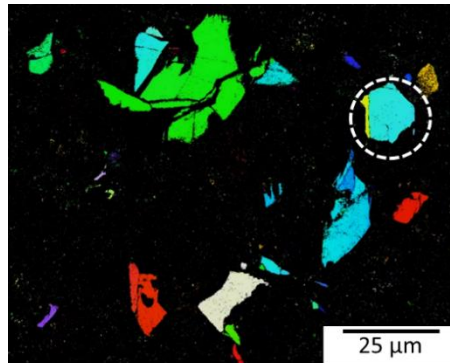
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First EBSD examination performed on U_3Si_2 → new insights in characterization

Study of as-fabricated $\text{U}_3\text{Si}_2/\text{Al}$ fuel plate and demonstration of the presence of:

- **large grains and single crystalline particles (slow cooling rate on industrial ingots),**
 - Laboratory production highlights that fast cooling rate ↘ grain size and ↗ mechanical resistance because of the intragranular fragmentation?
 - **Same fragmentation mechanism observed after U_3Si_2 hydrogenation.**



First EBSD examination performed on U_3Si_2 → new insights in characterization

Study of as-fabricated U_3Si_2 /Al fuel plate and demonstration of the presence of:

- **large grains and single crystalline particles (slow cooling rate on industrial ingots),**
 - Laboratory production highlights that fast cooling rate ↘ grain size and ↗ mechanical resistance because of the intragranular fragmentation?
 - **Same fragmentation mechanism observed after U_3Si_2 hydrogenation.**
- **two different Si rich secondary phases.**
 - U_8Si_8X supposedly stabilised by impurity (Laugier *et al.*, 1971) observed with high purity materials.
 - **Thermodynamic calculations underway to understand its formation condition.**

For further reading: Havette *et al.*, From arc-melted ingot to MTR fuel plate: A SEM/EBSD microstructural study of U_3Si_2 , in *Journal of Nuclear Materials*, vol. 537, 2020



Thank you for your attention

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Appendix

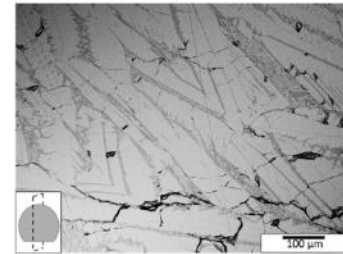
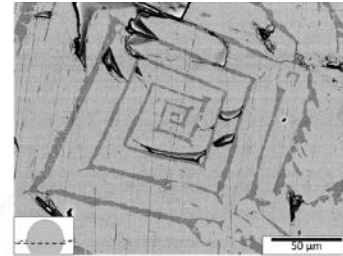
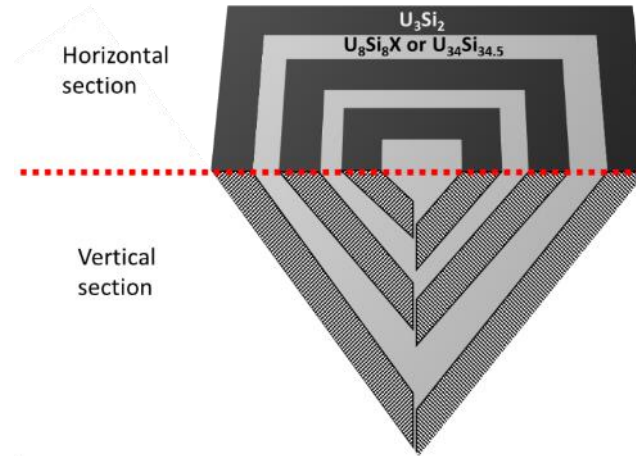
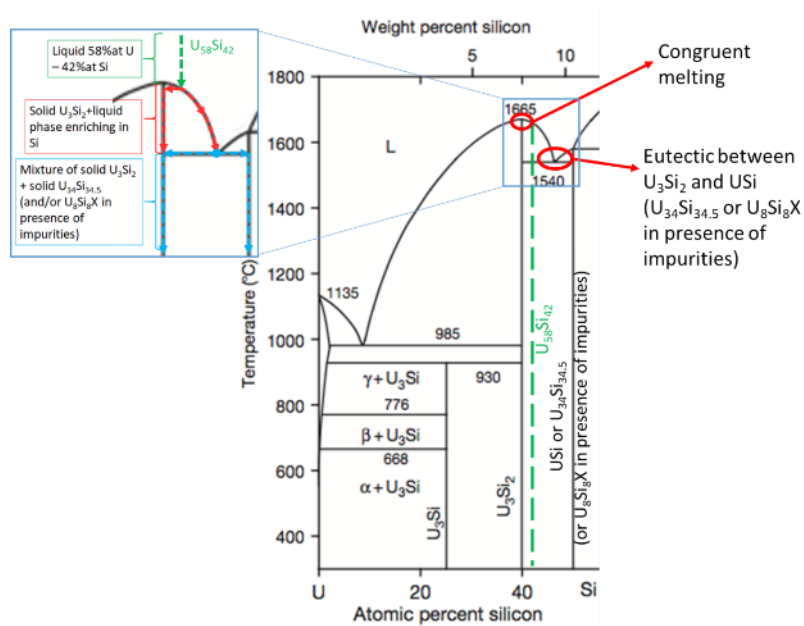
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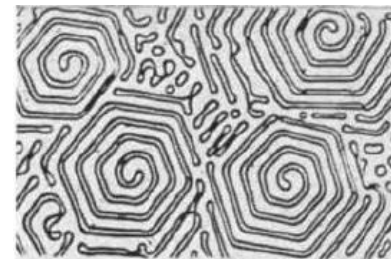
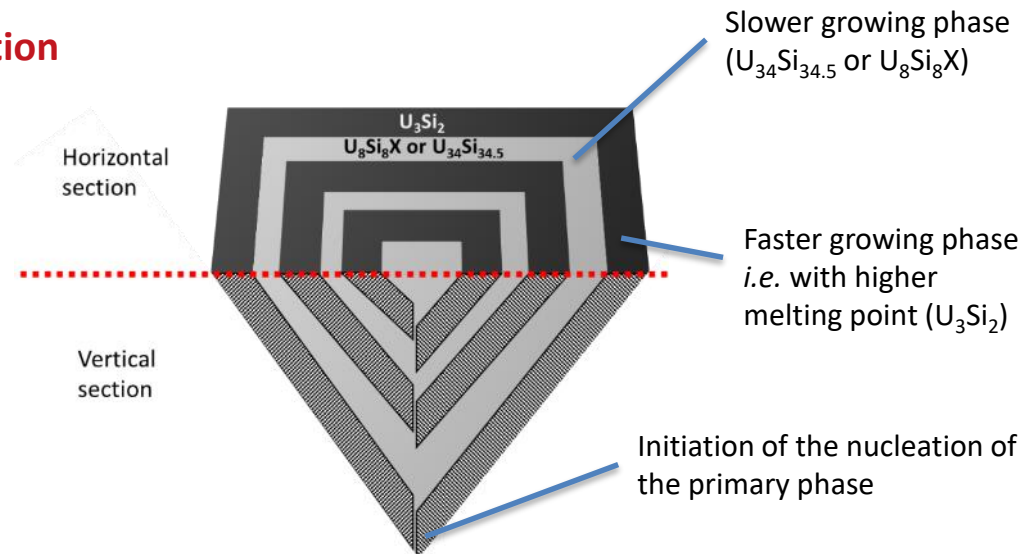
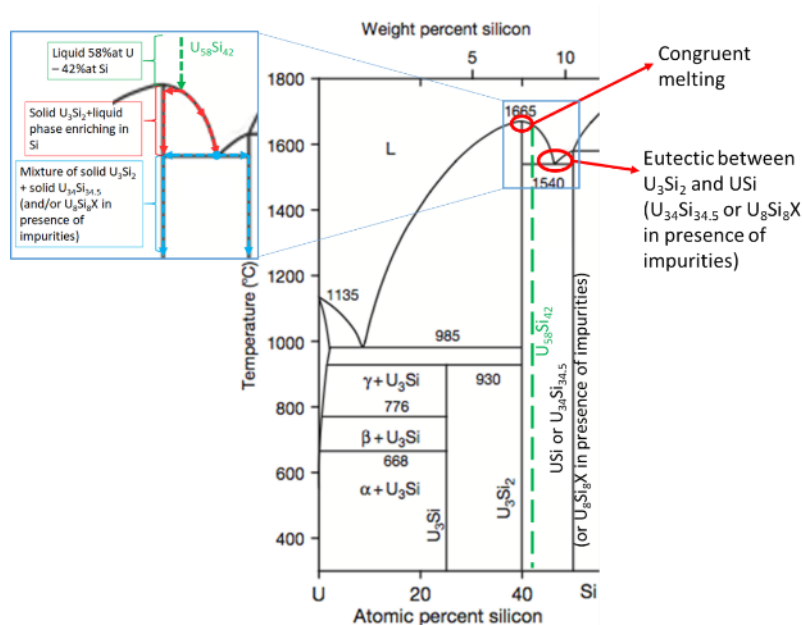
Table 1. Main impurities (in mg.kg⁻¹) in the two uranium batches used for the synthesis of U₃Si₂ by arc-melting.

U batches	Elements														
	Ag	Al	B	C	Co	Cr	Cu	Fe	Mn	Mo	O	Ni	Pb	Si	Sn
“High purity”	<0.5	15	<0.2	300	≤1	3.5	2	25	3.5	<1	300	7	≤1	10	2
“Low purity”	-	90	-	300	-	12	15	85	5	-	1500	40	-	37	-

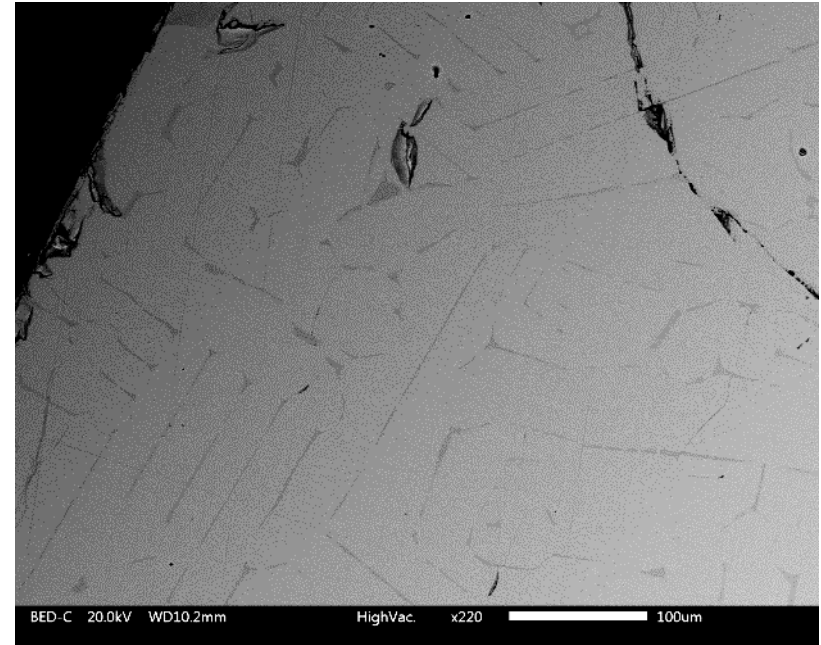
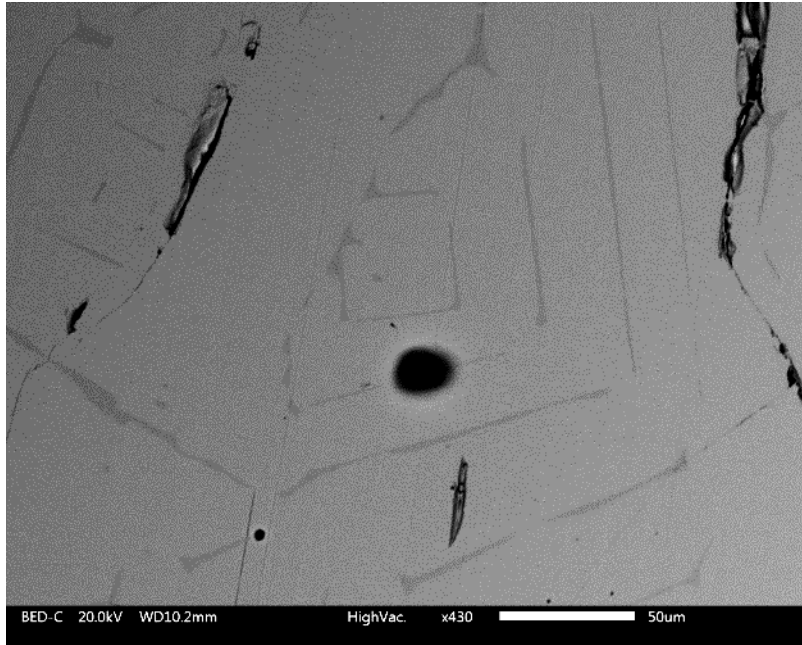
Mechanism of spiral microstructure formation



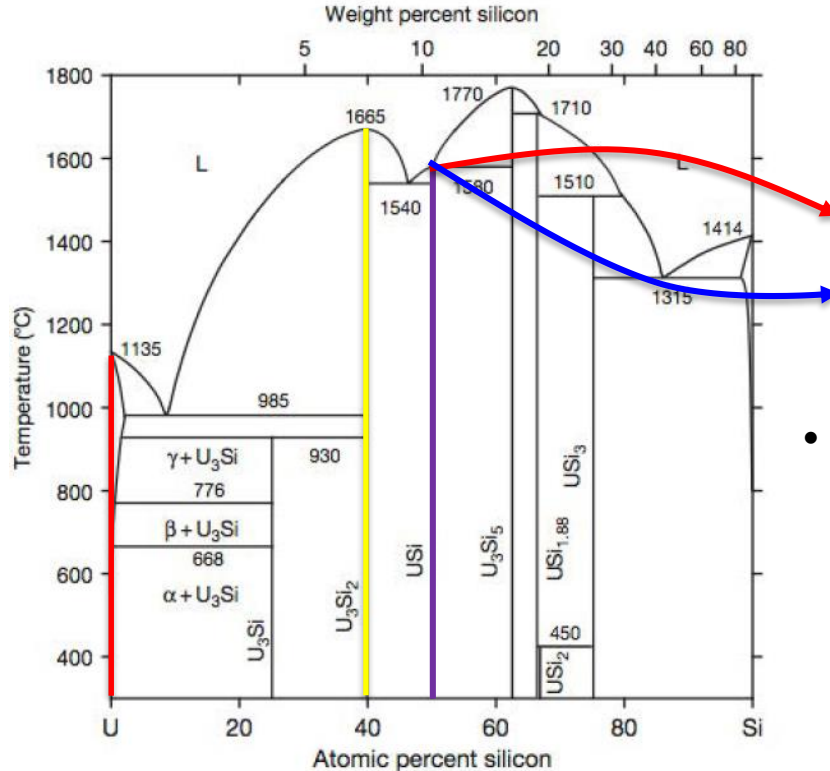
Mechanism of spiral microstructure formation



Already observed in Zn-Zn+3%Mg eutectic (hexagonal) [9].



- Several compounds in the binary diagram:



Phase	Space group	a (Å)	b (Å)	c (Å)	Reference
U_3Si_2	127 (P4/mbm)	7.33	7.33	3.90	[4]

- U_3Si_2 is a line compound