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

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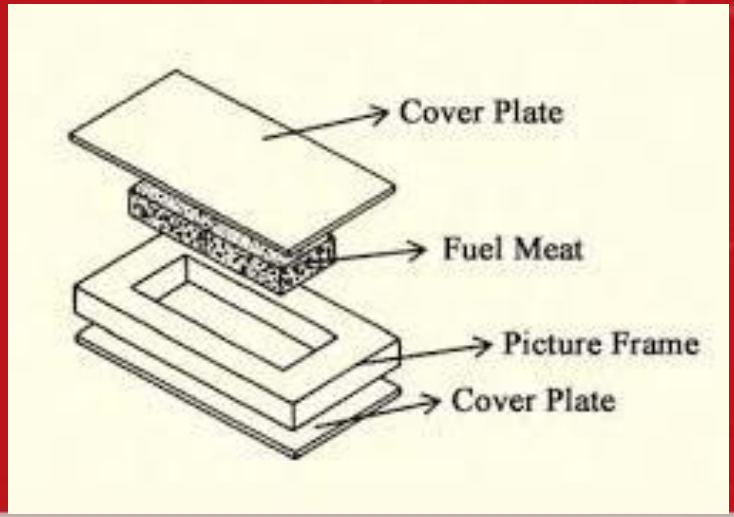

Introduction

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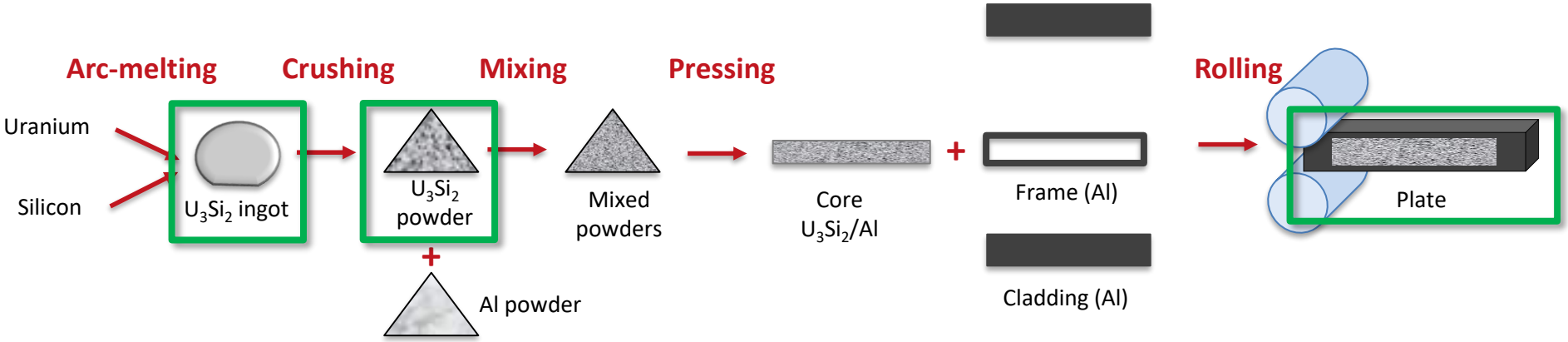
Context

- 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.

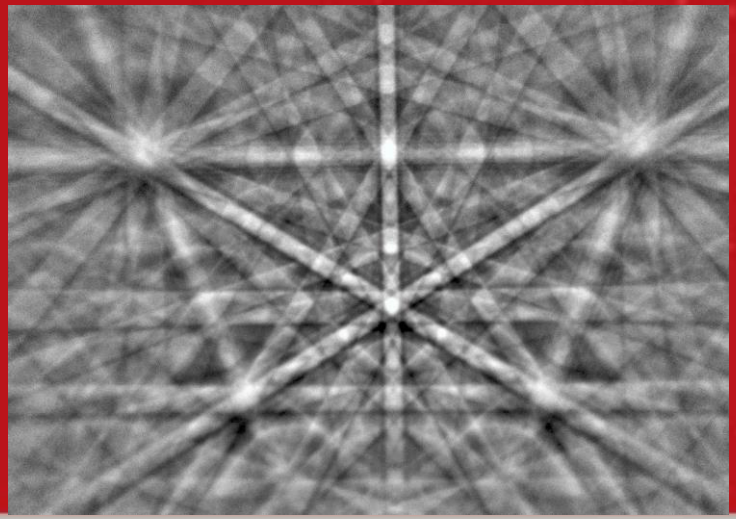
Fuel	*Melting point (°C)*	*Physical density ($g\,cm^{-3}$)*	*Uranium loading ($g\,cm^{-3}$)*
U	1133	19.1	19.1
U–7Mo	1145	18.4	17.1
U–10Mo	1150	18.2	16.4
U_6Mn	726	17.8	17.1
U_6Fe	815	17.7	17.0
U_3Si	930	15.6	15.0
U_3Si_2	1665	12.2	11.3
USi	1580	10.96	9.8
UAl_2	1590	8.1	6.6
UAl_3	1350	6.8	5.0
UAl_4	731	6.1	4.2
$U_{0.9}Al_4$	641	5.7	3.7
UAl_x	NA	6.4	4.5

Objectives of this study

- Material Testing Reactors (MTRs) → mainly nuclear fuel plates.
- U_3Si_2/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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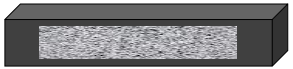
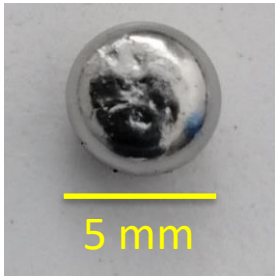
Samples

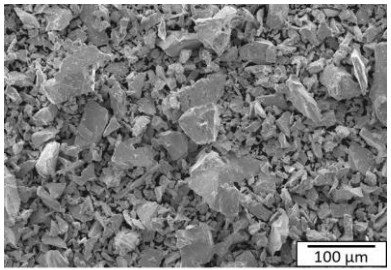
- **U_3Si_2/Al fuel plate**
  - Framatome-CERCA production
  - Fabrication in the frame of the CEA SHARE irradiation test (Leenaers *et al.*, 2008)



- **Arc-melted ingots and crushed ingots**
  - Laboratory (CEA-Cadarache) production

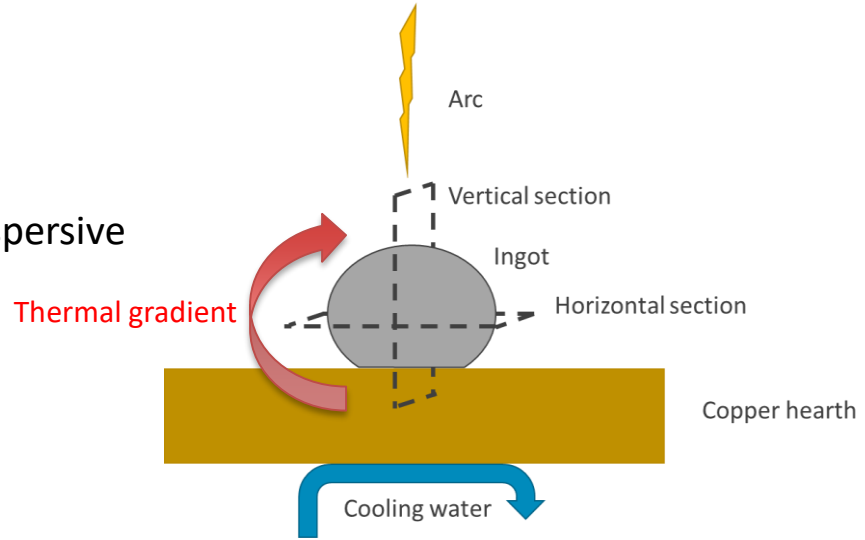
Label	Cooling	U/Si atomic ratio	Weight (g)
I1	“Very fast”	60/40	0.5
I2	“Slow”	60/40	1
I3	“Fast”	60/40 “low purity U”	1
I4	“Fast”	58/42	1

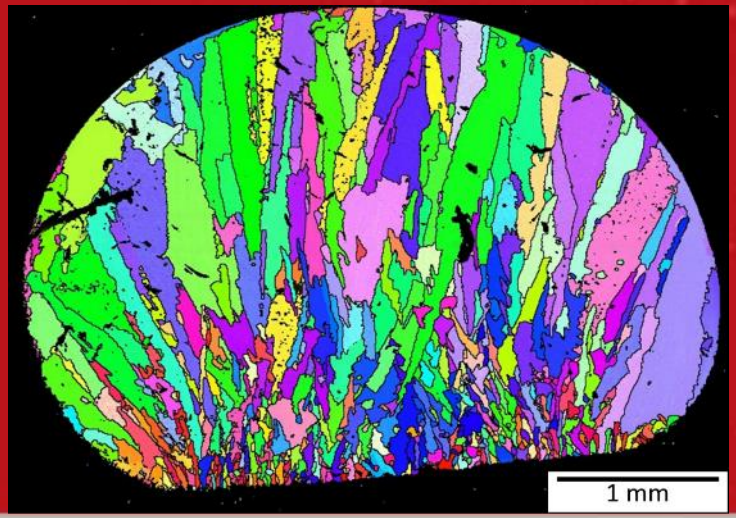




Characterization techniques

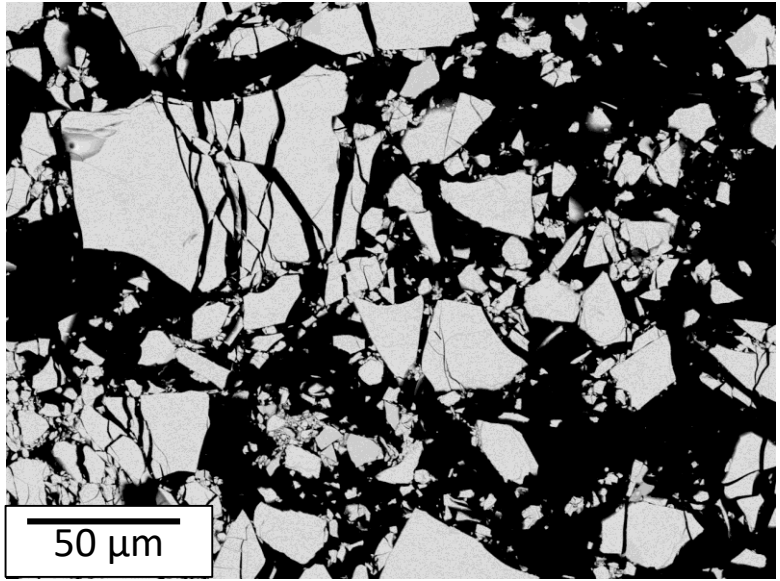
- X-Ray Diffraction (XRD)
- Scanning Electron Microscopy (SEM) + Energy Dispersive Spectroscopy (EDS)
- Electron BackScattered Diffraction (EBSD)

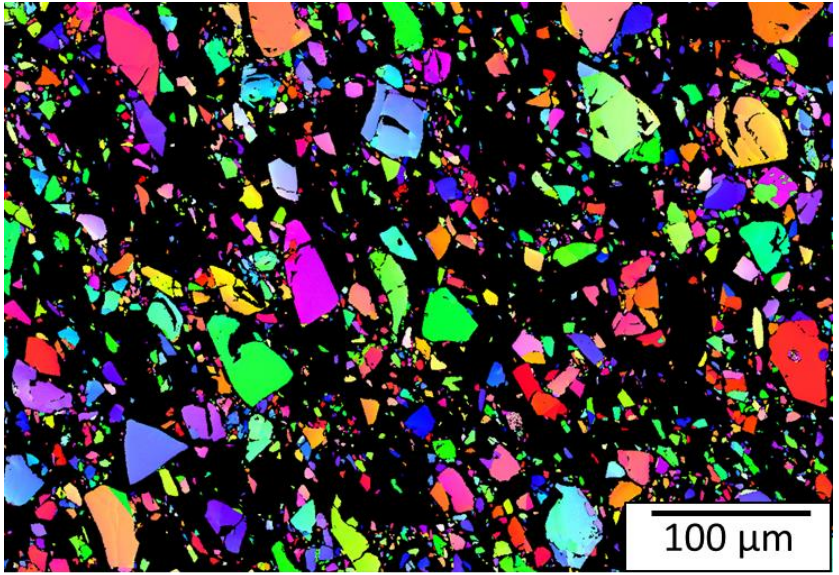


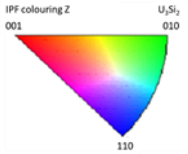


Results and discussion

MTRs fuel plate

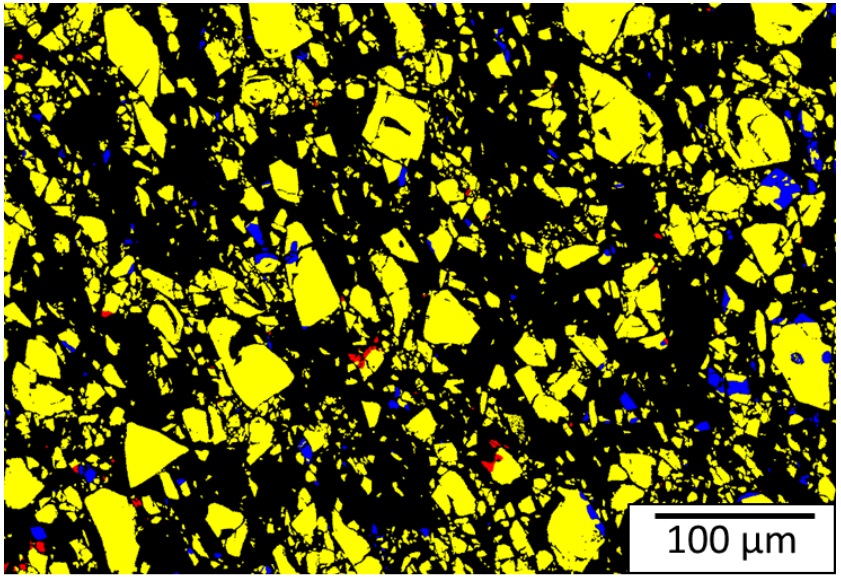




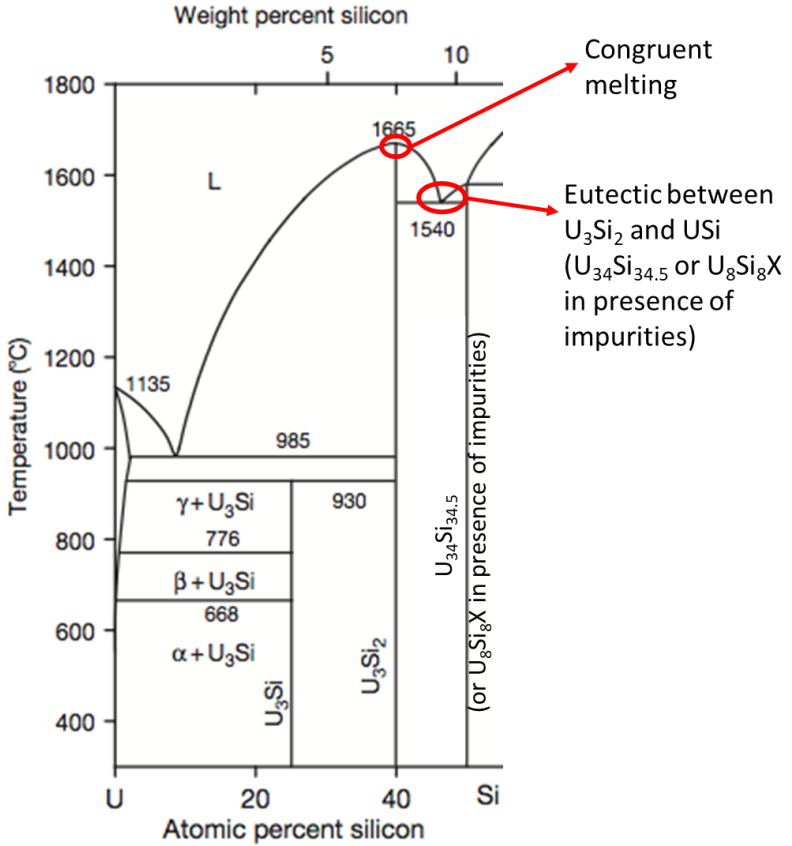


- Important fragmentation of particles.
- Single crystalline particles.

MTRs fuel plate



$U_{34}Si_{34.5}$
U_8Si_8X
U_3Si_2

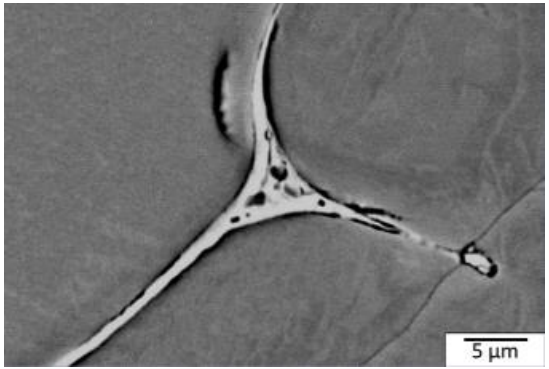


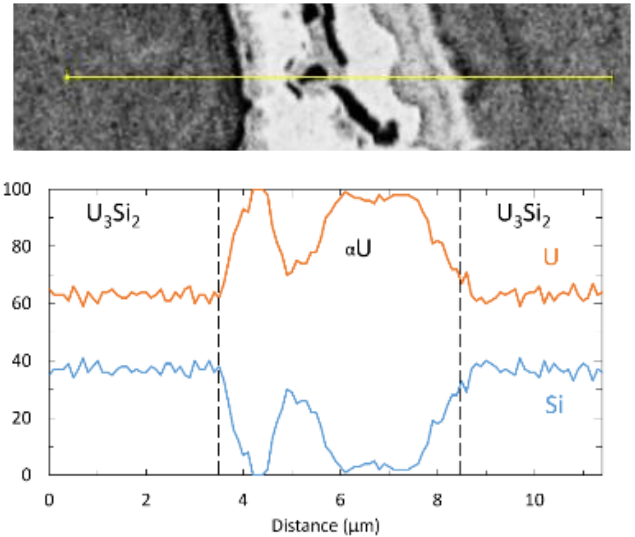
Map coloured in phase highlights the presence of Si rich secondary phases :

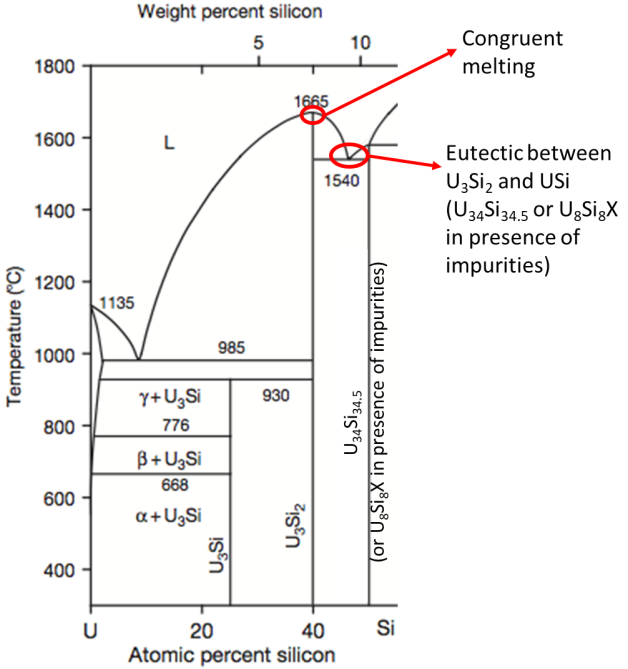
- $U_{34}Si_{34.5}$ (3.5%)
- U_8Si_8X (0.5%)

SEM characterizations

- αU at grain boundaries.

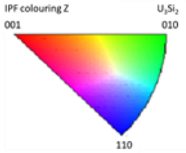


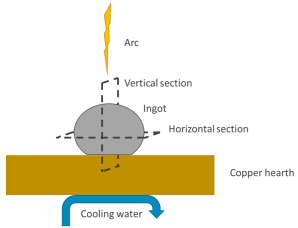


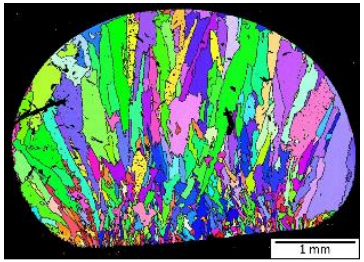
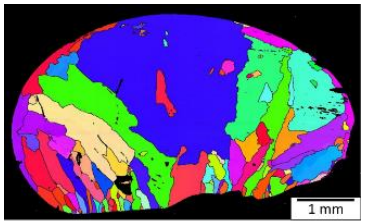


“High purity” stoichiometric ingots - Cooling rate effect

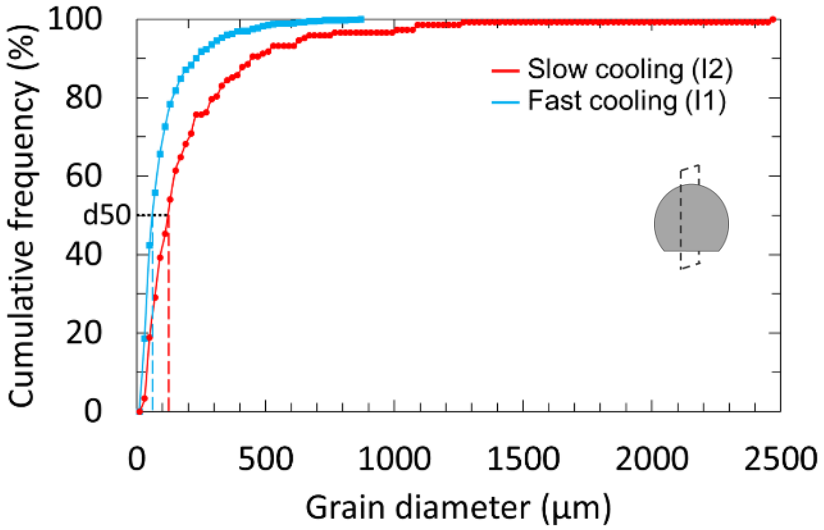
Electron Backscattered Diffraction





Fast cooled ingot (I1)		●Elongated grains ● Smaller grains close to the copper hearth
Slow cooled ingot (I2)		

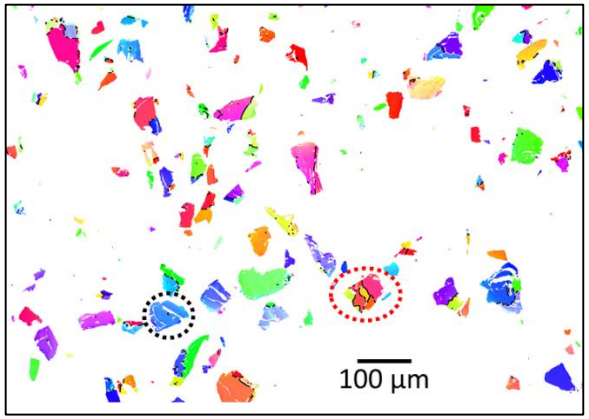
Grains size analysis



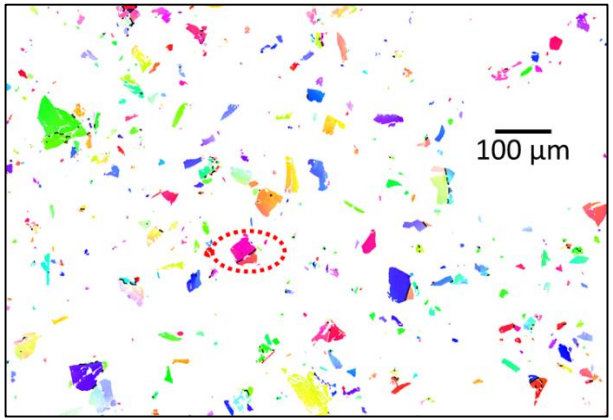
Slow cooling favours grain growth.

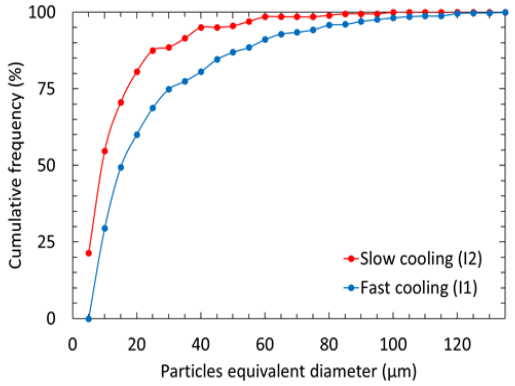
Impact on the crushed ingot

I1: fast cooled ingot



I2: slow cooled ingot





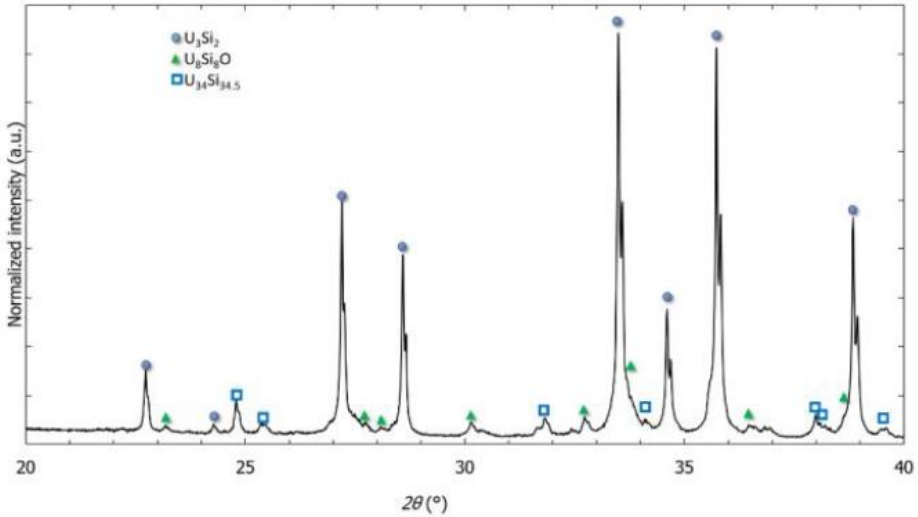
- Mainly single crystalline particles.
- Grain boundaries in particles → intragranular fragmentation (as evidenced by Metzger *et al.*, 2017).
- **Influence of fast cooling:**
  - **- U_3Si_2 grains are smaller in the ingot**
  - **- U_3Si_2 particles are larger after a given crushing sequence → influence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

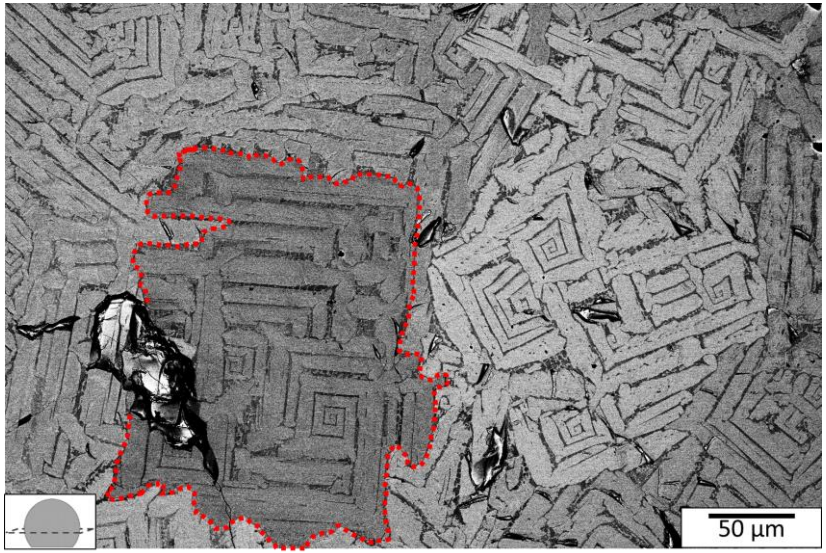
“Low purity” stoichiometric ingot

- Secondary phases indexed :
  - U_8Si_8X
  - $U_{34}Si_{34.5}$ (Le Bihan *et al.*, 1996)

- Solidification in spiral shape

I3 ingot

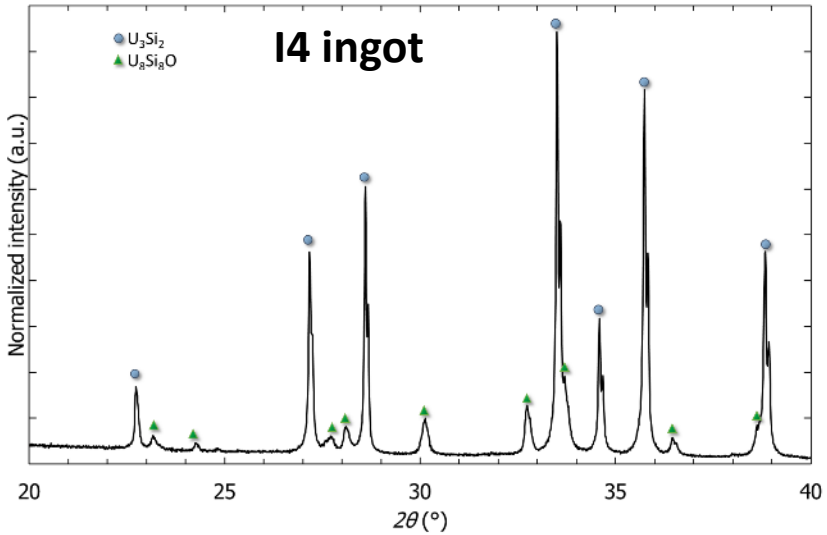


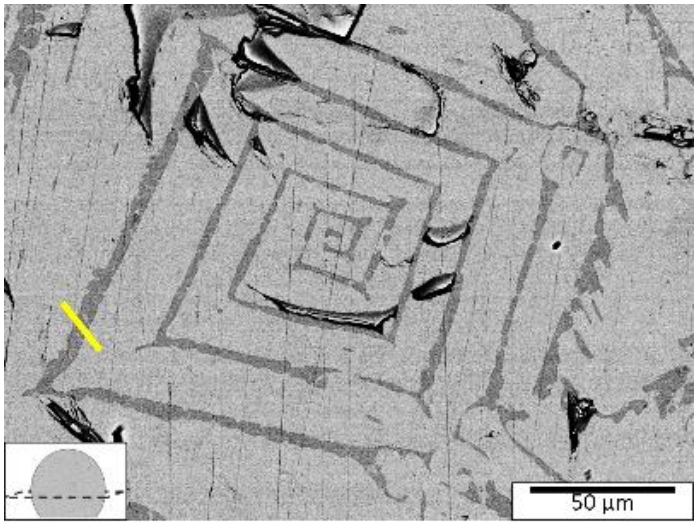


“High purity” non stoichiometric ingot

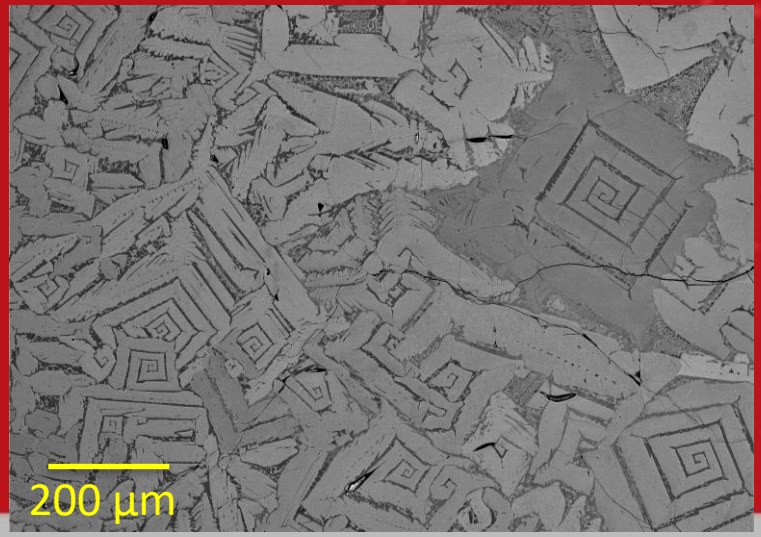
2%at. Silicon excess

- Secondary phase indexed as U_8Si_8X.
- Solidification in spiral shape.





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



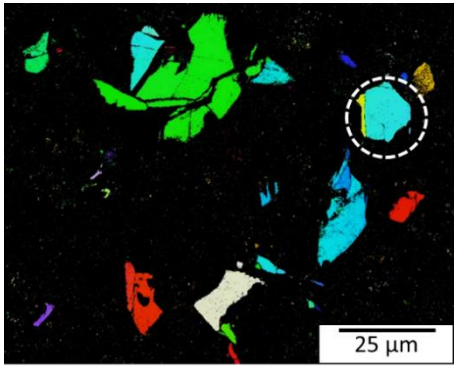
Conclusion

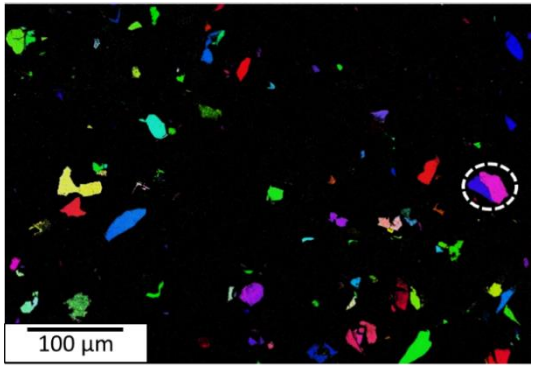
Conclusion

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.**





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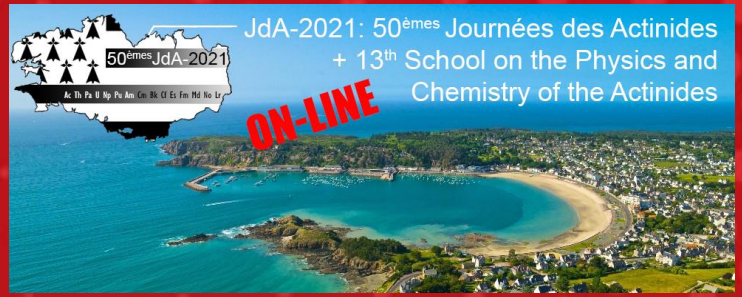
  → **Same fragmentation mechanism observed after U_3Si_2 hydrogenation.**

- **two different Si rich secondary phases**.
  → U_8Si_8X supposingly 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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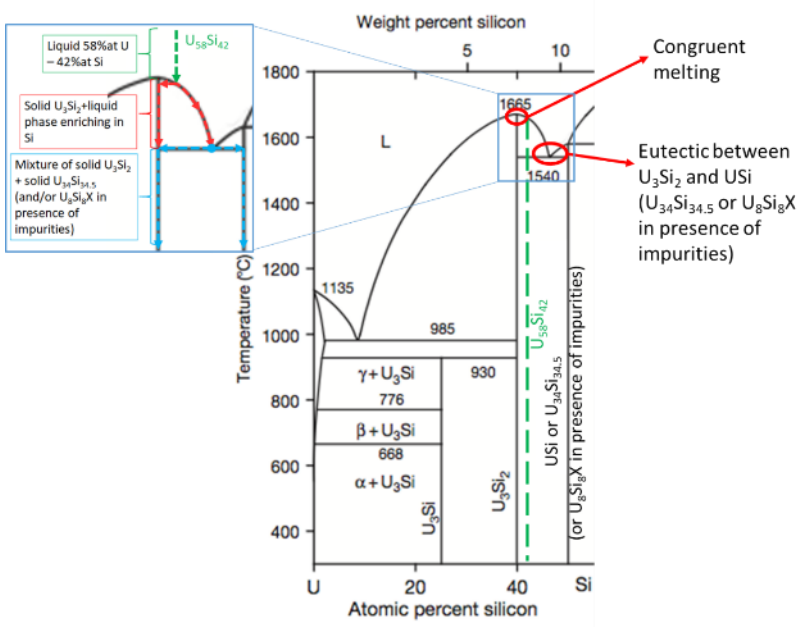
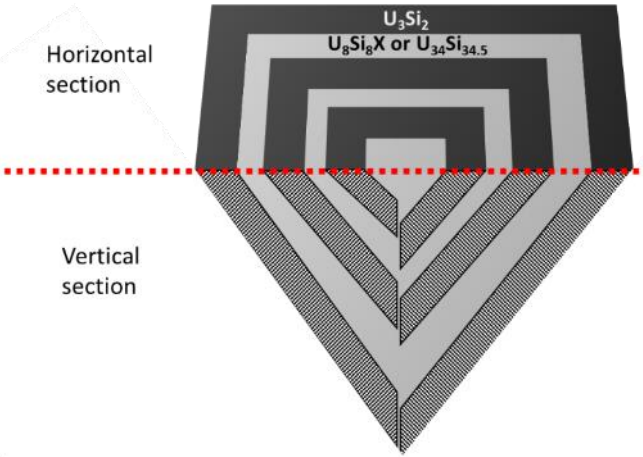
Uranium batch purity

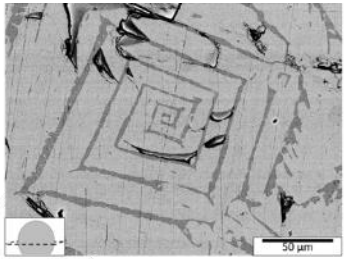
Table 1. Main impurities (in $mg.kg^{-1}$) in the two uranium batches used for the synthesis of U_3Si_2 by arc-melting.

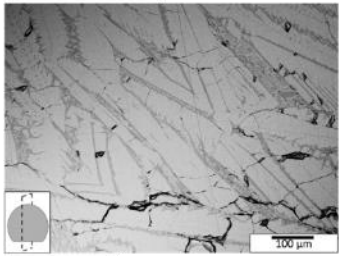
U batches	Elements														
	Ag	Al	B	C	Co	Cr	Cu	Fe	Mn	Mo	O	Ni	Pb	Si	Sn
"High purity"	<0.5	15	<0.2	300	≤1	3.5	2	25	3.5	<1	300	7	≤1	10	2
"Low purity"	-	90	-	300	-	12	15	85	5	-	1500	40	-	37	-

Mechanism of spiral microstructure formation

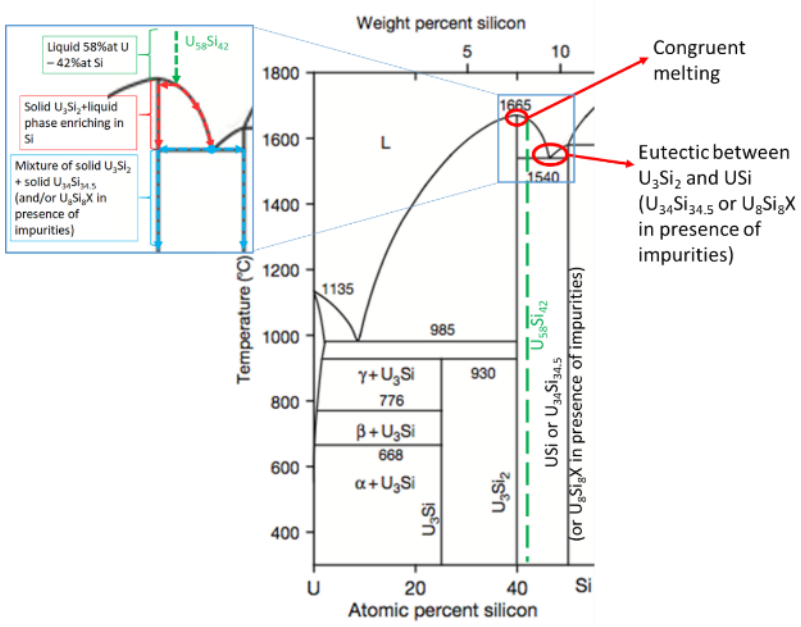


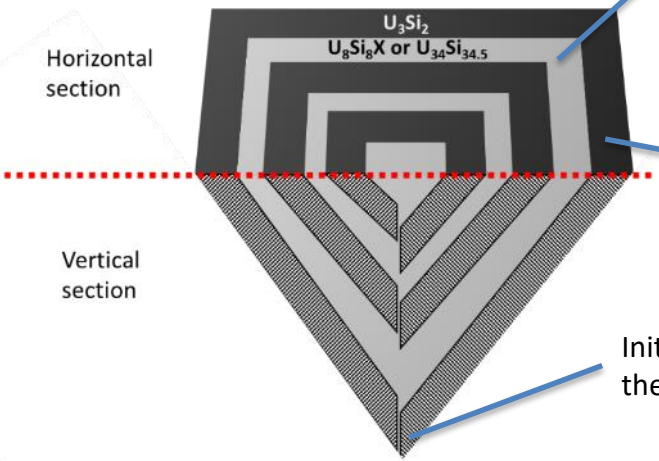






Mechanism of spiral microstructure formation

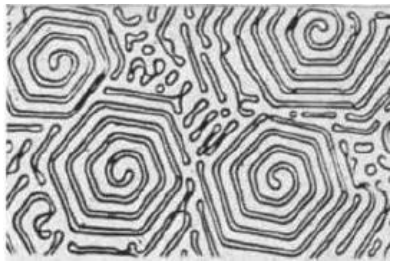




Slower growing phase ($U_{34}Si_{34.5}$ or U_8Si_8X)

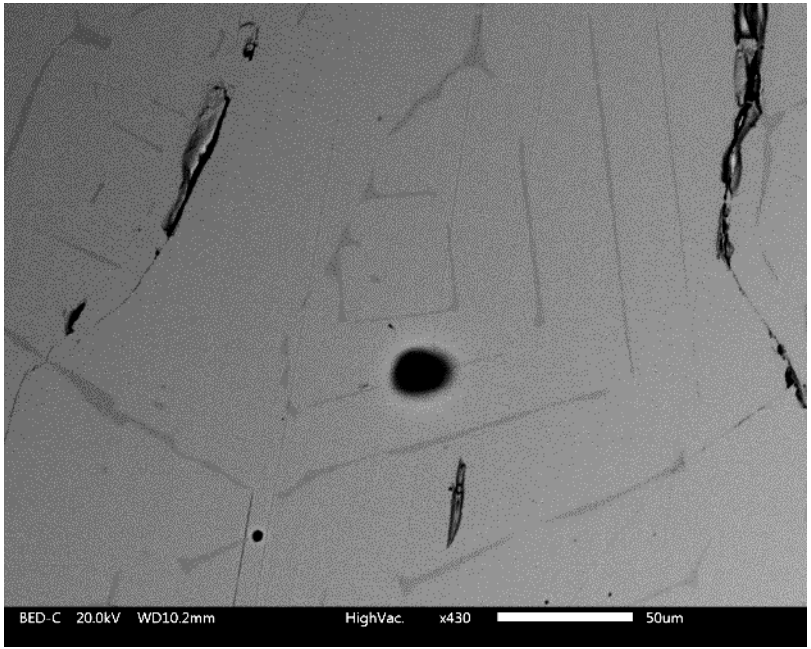
Faster growing phase *i.e.* with higher melting point (U_3Si_2)

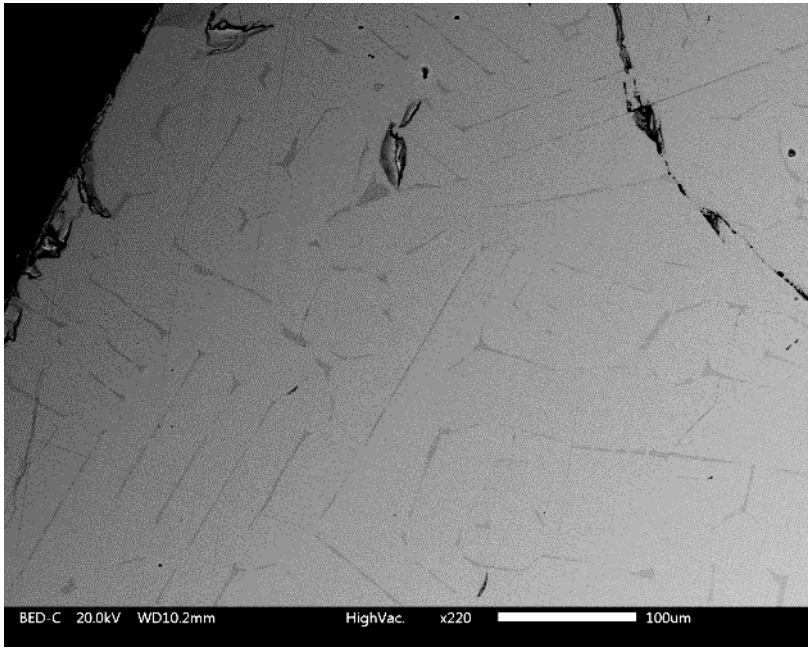
Initiation of the nucleation of the primary phase



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

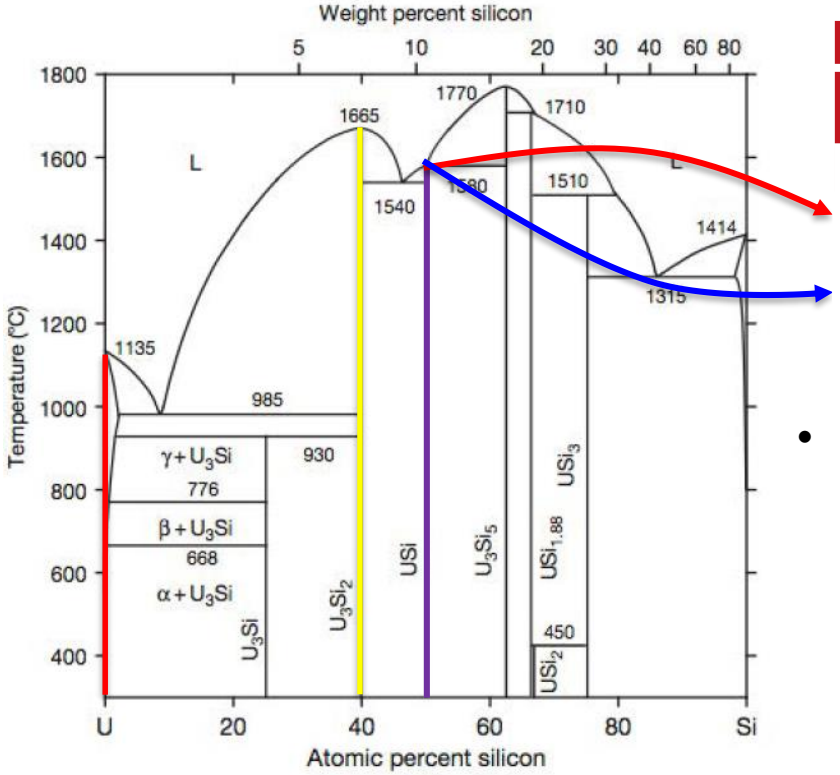
1%at. Si excess





U-Si binary diagram

- Several compounds in the binary diagram:



U-Si binary phase diagram (Massalski *et al.*, 1990)

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