



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

Julien Havette, Xaviere Iltis, Herve Palancher, Doris Drouan, Olivier Fiquet, Etienne Castelier, Mathieu Pasturel

► To cite this version:

Julien Havette, Xaviere Iltis, Herve Palancher, Doris Drouan, Olivier Fiquet, et al.. From arc-melted ingot to MTR fuel plate: a SEM/EBSD microstructural study of U₃Si₂. JdA 2021 - 50èmes Journées des Actinides + 13th school on the physics and chemistry of actinides, Mar 2021, virtual event, France. cea-03697840v1

HAL Id: cea-03697840

<https://cea.hal.science/cea-03697840v1>

Submitted on 20 Jun 2022 (v1), last revised 17 Jun 2022 (v2)

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



DE LA RECHERCHE À L'INDUSTRIE

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

Julien Havette^{1,2}, X. Iltis¹, H. Palancher¹, D. Drouan¹,
O. Fiquet¹, E. Castelier¹, M. Pasturel²

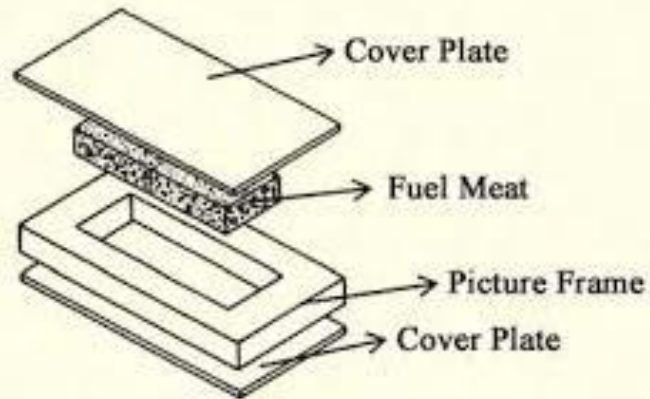
¹ CEA, DES, IRESNE, DEC, Cadarache F-13108 Saint-Paul-Lez-Durance, France

² Univ. Rennes 1, CNRS, Institut des Sciences Chimiques de Rennes –
UMR6226, F-35000 Rennes

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

IRESNE | DEC | SA3E | LCPC

Institut de recherche sur les systèmes nucléaires pour la production d'énergie bas carbone



Introduction

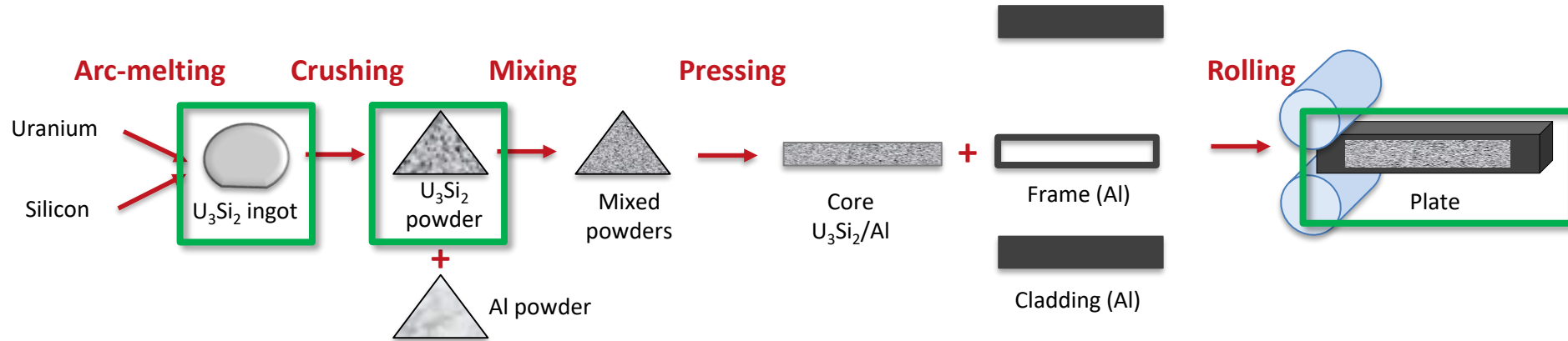
IRESNE | DEC | SA3E | LCPC

Institut de recherche sur les systèmes nucléaires pour la production d'énergie bas carbone

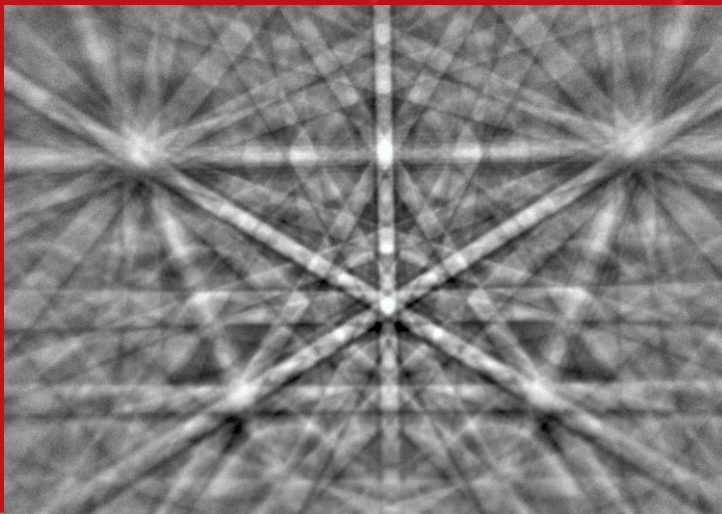
- Conversion of Research Reactors to Low Enriched Uranium (<20% ^{235}U).
- Development of new nuclear fuels with higher uranium density, for high performance MTRs.
- U_3Si_2 is one of the main candidates.

<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.



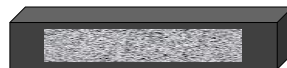
Experimental methods

IRESNE | DEC | SA3E | LCPC

Institut de recherche sur les systèmes nucléaires pour la production d'énergie bas carbone

- **$\text{U}_3\text{Si}_2/\text{Al}$ fuel plate**

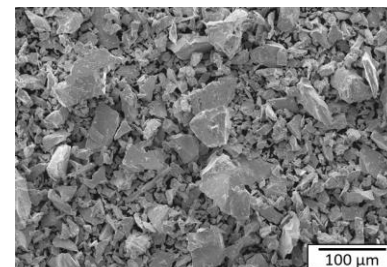
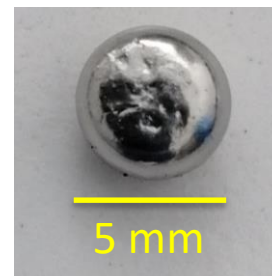
- Framatome-CERCA production
- Fabrication in the frame of the CEA SHARE irradiation test (Leenaers *et al.*, 2008)



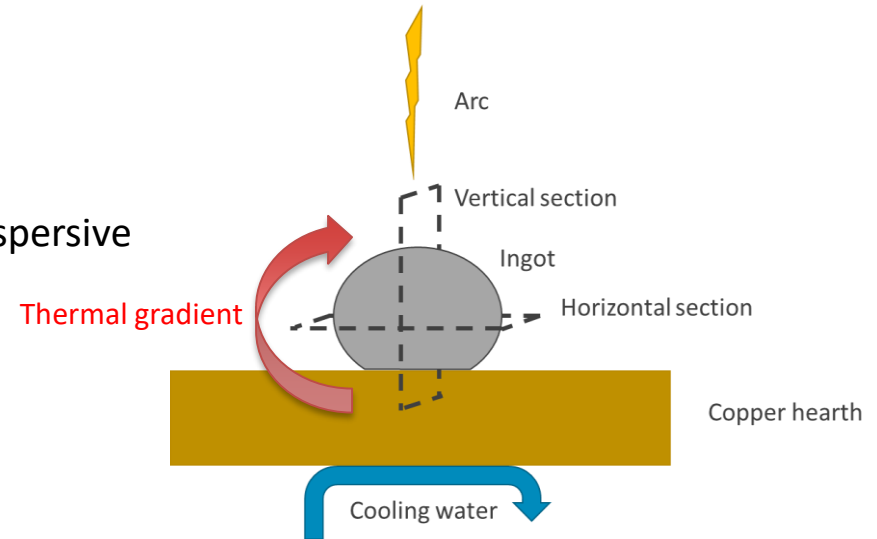
- **Arc-melted ingots and crushed ingots**

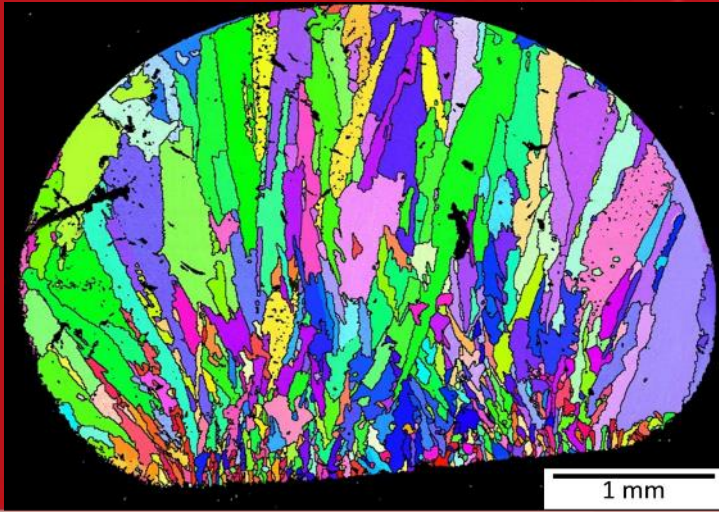
- Laboratory (CEA-Cadarache) production

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)

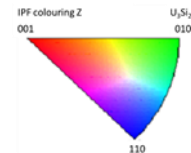
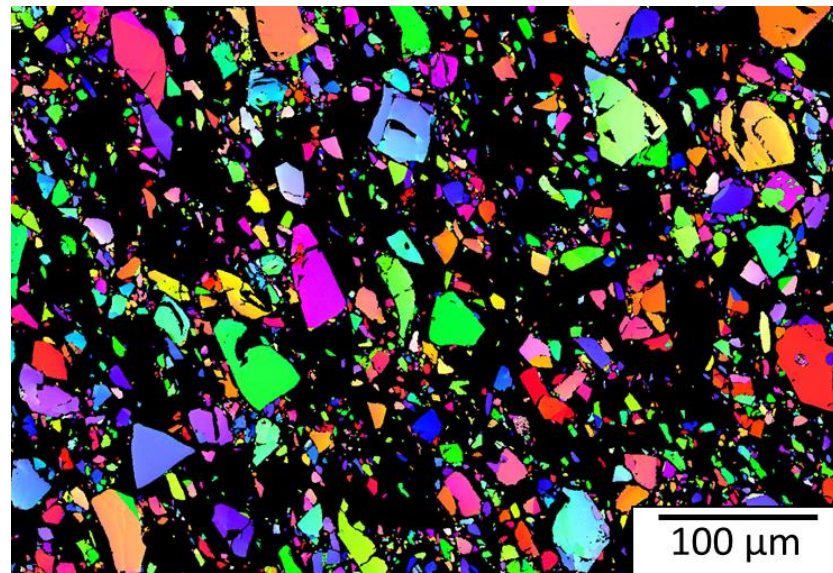
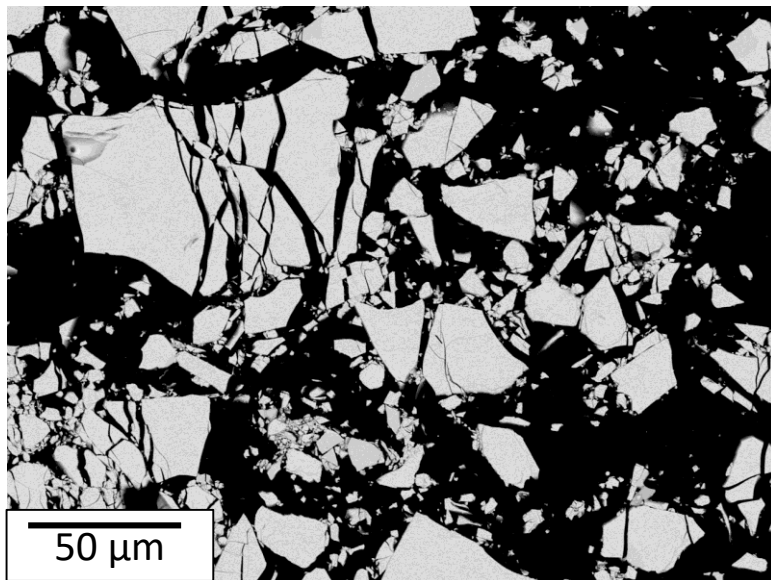




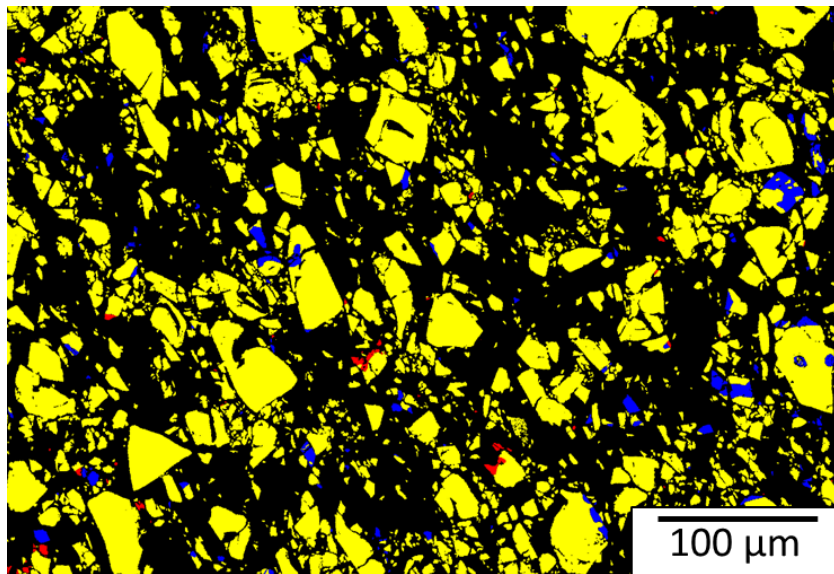
Results and discussion

IRESNE | DEC | SA3E | LCPC

Institut de recherche sur les systèmes nucléaires pour la production d'énergie bas carbone

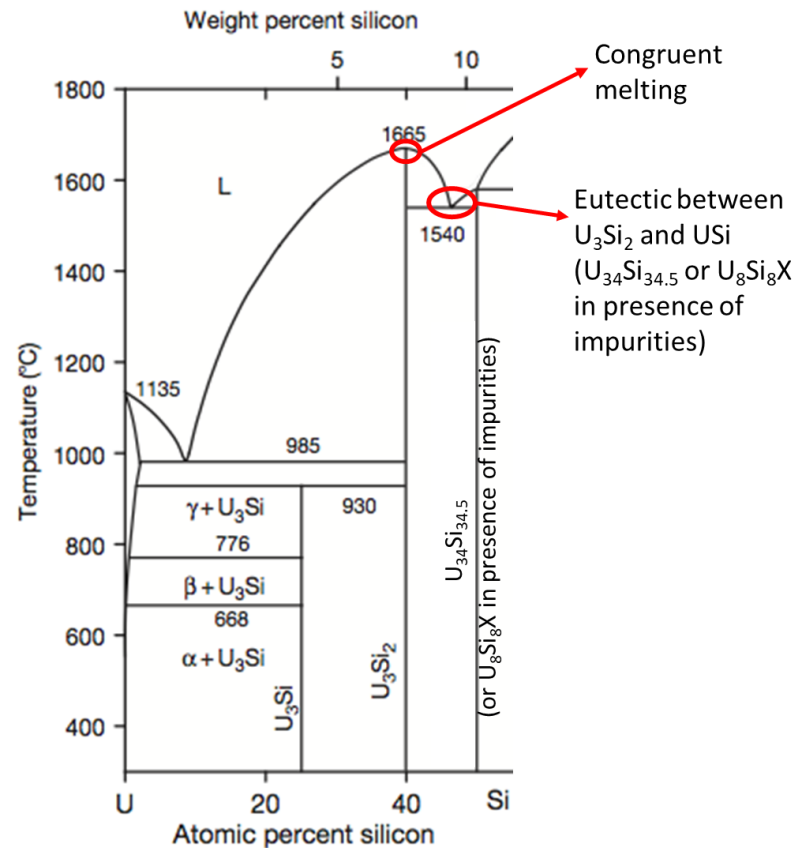


- Important fragmentation of particles.
- Single crystalline particles.



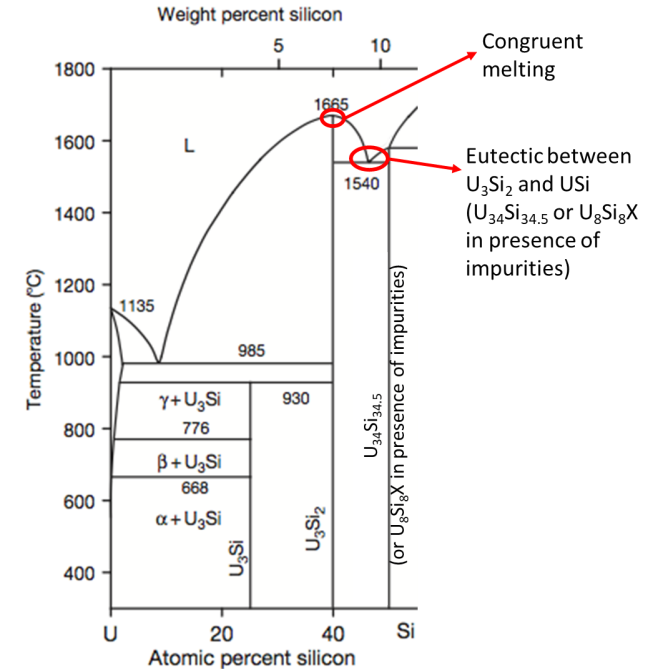
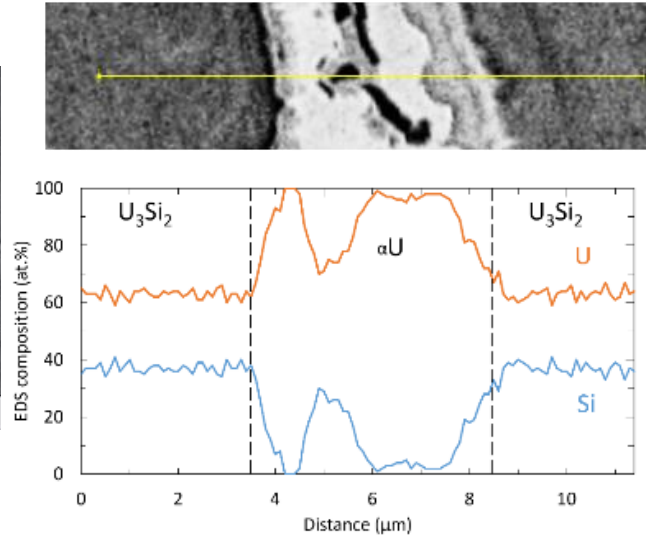
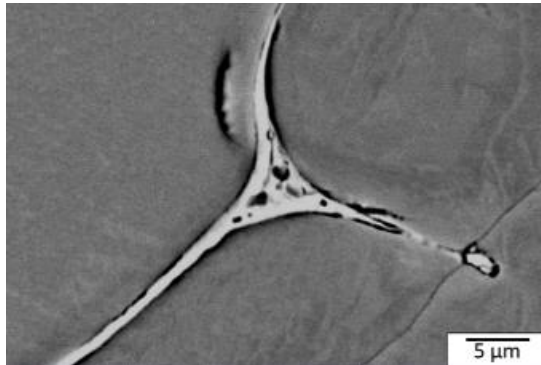
Map coloured in phase highlights the presence of Si rich secondary phases :

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

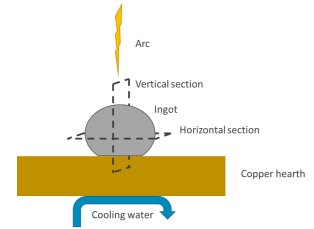
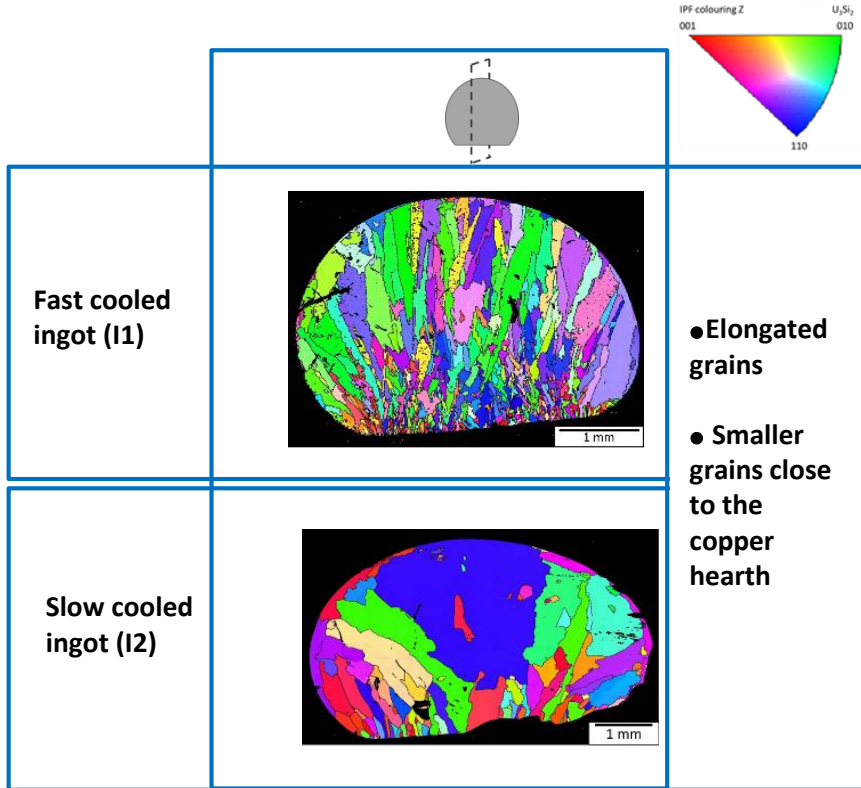


SEM characterizations

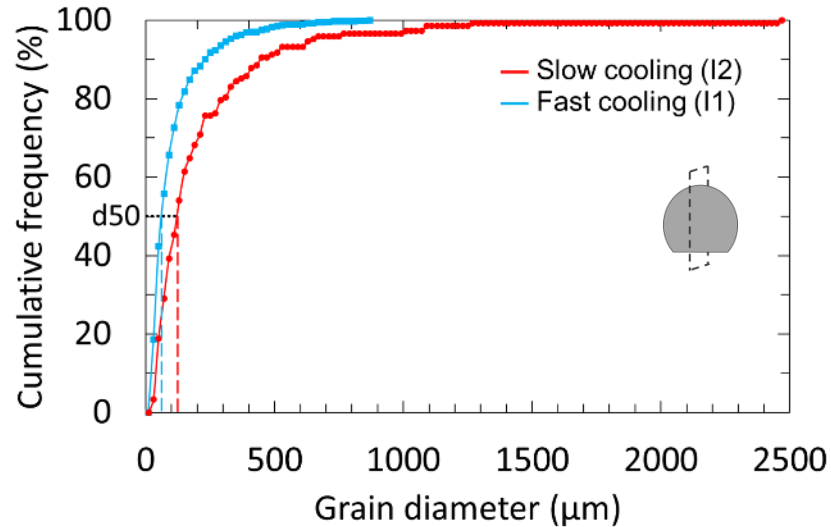
- α U at grain boundaries.



Electron Backscattered Diffraction

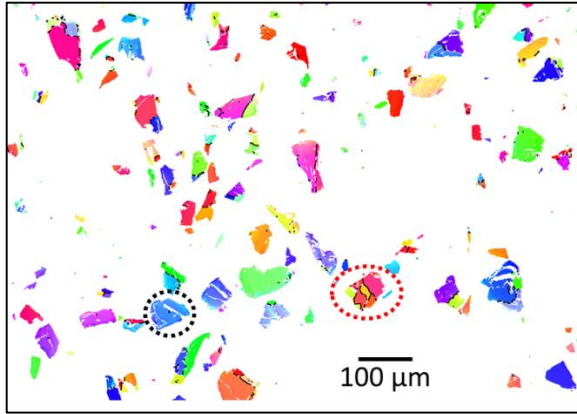


Grains size analysis

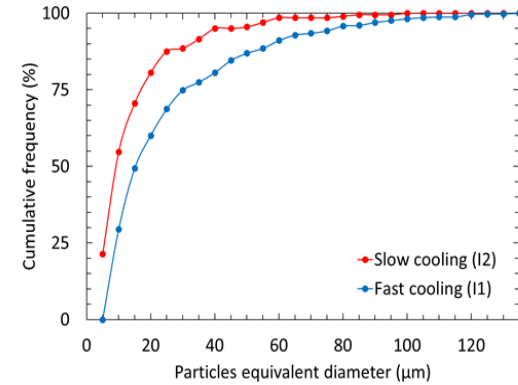


Slow cooling favours grain growth.

I1: fast cooled ingot



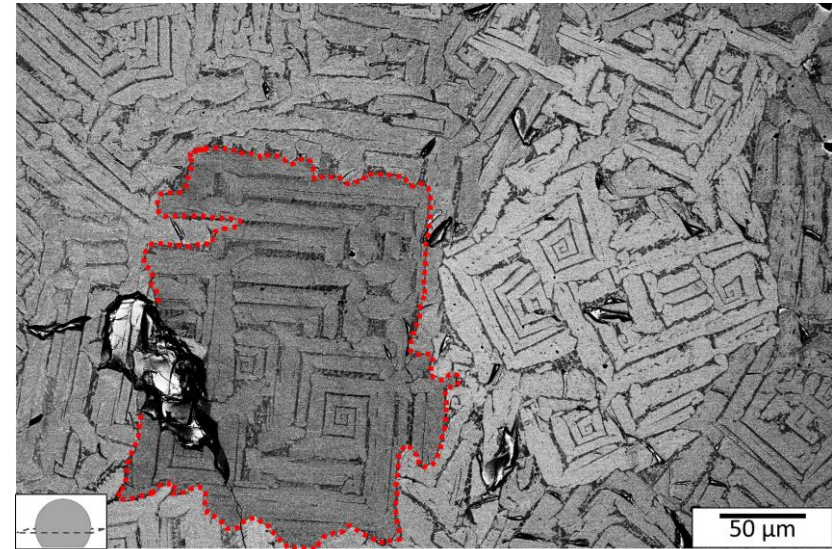
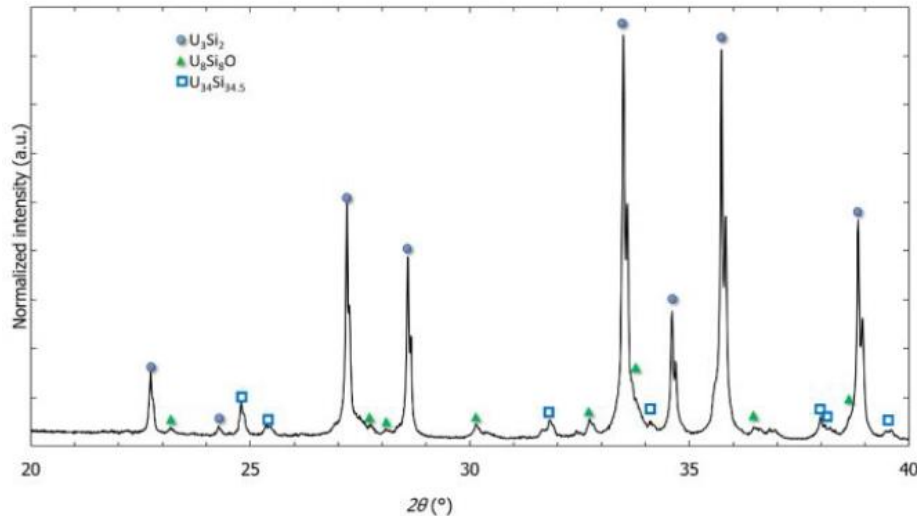
I2: slow cooled ingot



- 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?

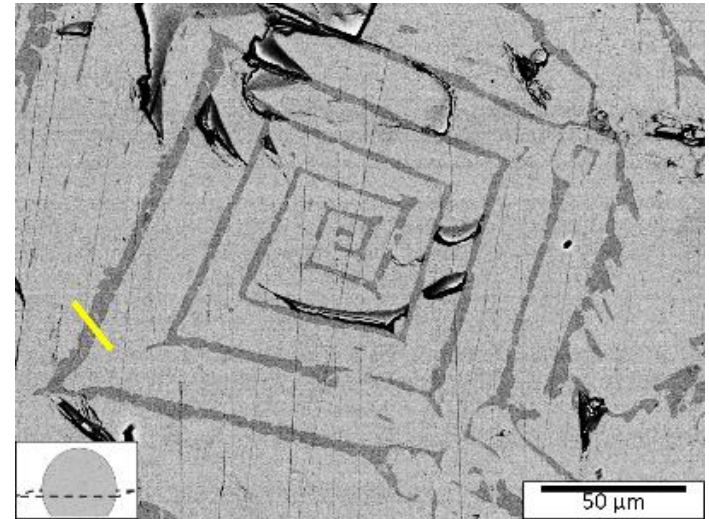
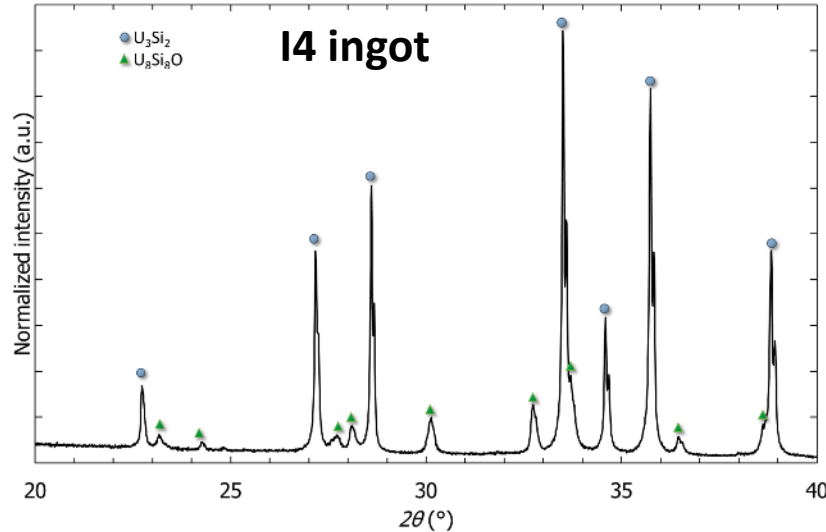
- Secondary phases indexed :
 - $\text{U}_8\text{Si}_8\text{X}$
 - $\text{U}_{34}\text{Si}_{34.5}$ (Le Bihan *et al.*, 1996)
- Solidification in spiral shape

I3 ingot

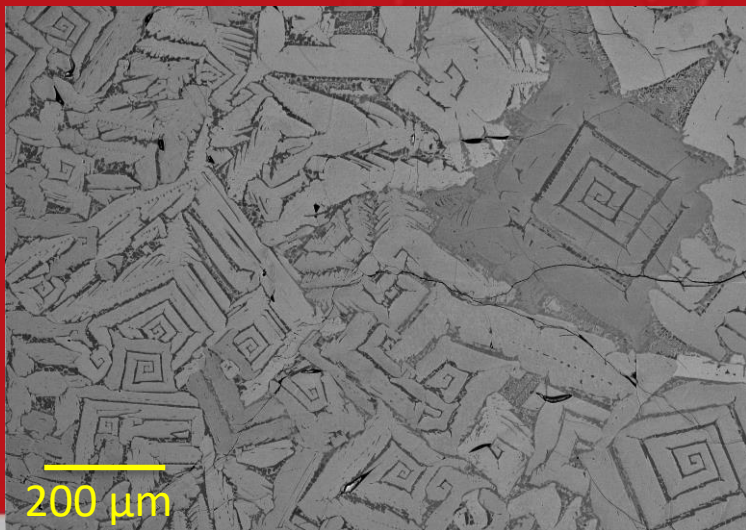


2%at. Silicon excess

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



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



Conclusion

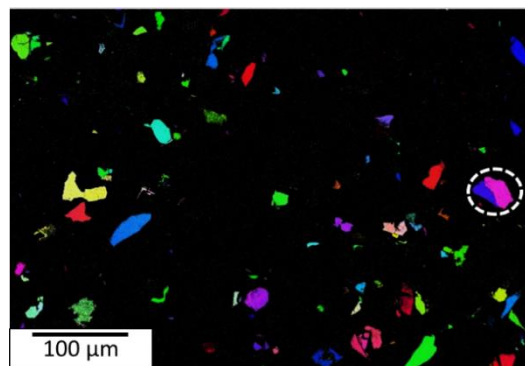
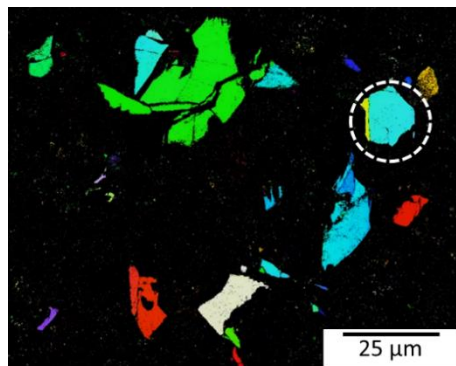
IRESNE | DEC | SA3E | LCPC

Institut de recherche sur les systèmes nucléaires pour la production d'énergie bas carbone

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.**

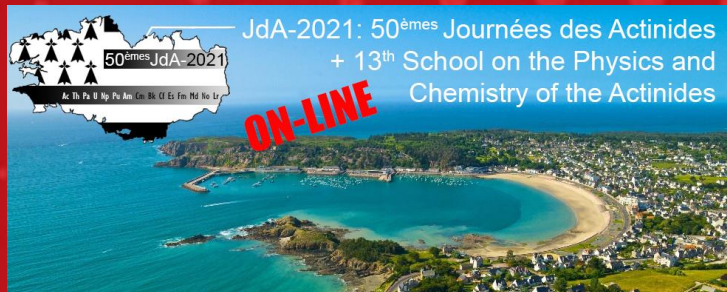


First EBSD examination performed on U_3Si_2 → new insights in characterization

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

DE LA RECHERCHE À L'INDUSTRIE

IRESNE | DEC | SA3E | LCPC

Institut de recherche sur les systèmes nucléaires pour la production d'énergie bas carbone

Commissariat à l'énergie atomique et aux énergies alternatives - www.cea.fr



DE LA RECHERCHE À L'INDUSTRIE

Appendix

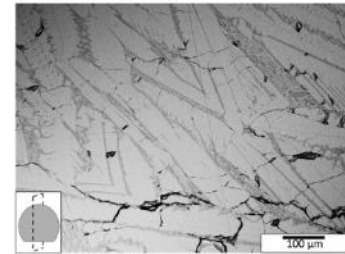
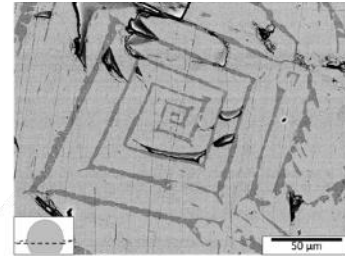
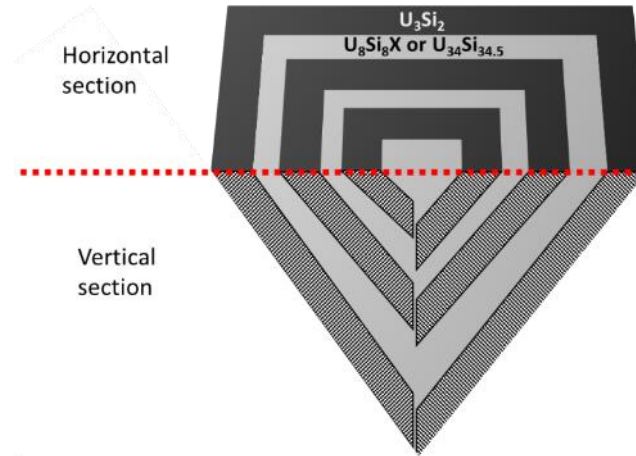
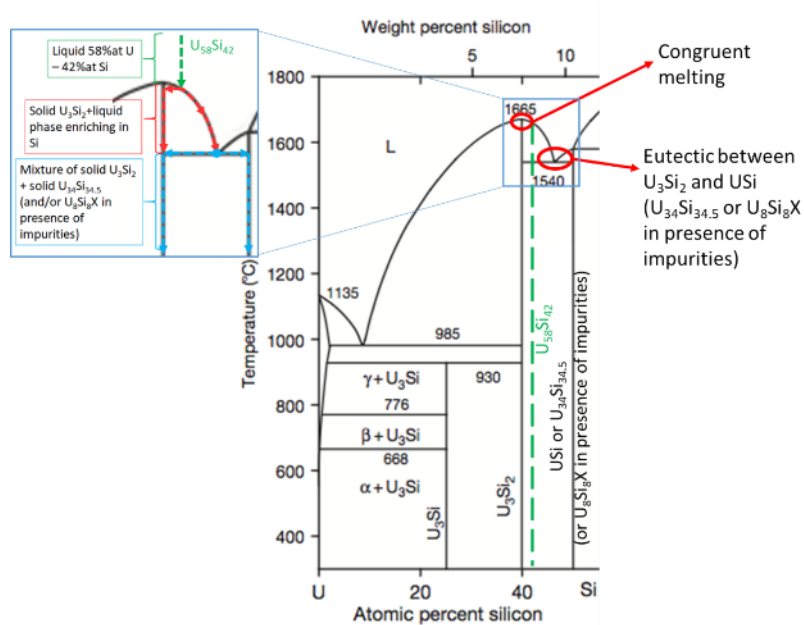
IRESNE | DEC | SA3E | LCPC

Institut de recherche sur les systèmes nucléaires pour la production d'énergie bas carbone

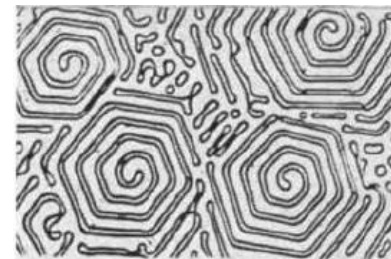
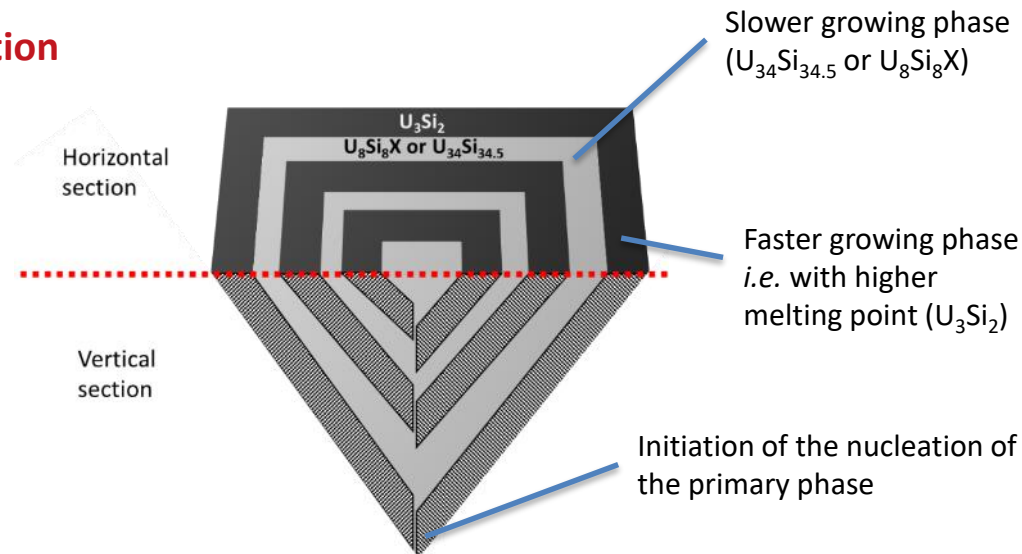
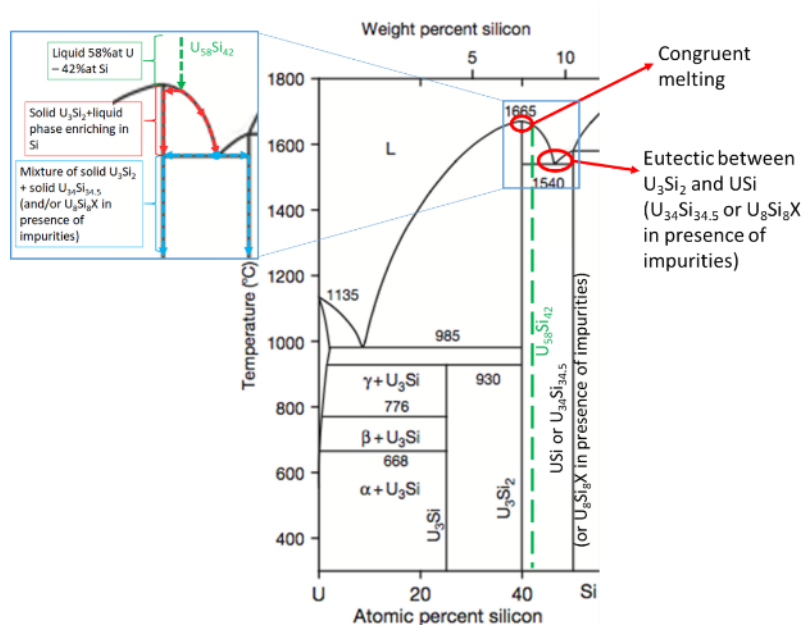
Table 1. Main impurities (in mg.kg⁻¹) in the two uranium batches used for the synthesis of U₃Si₂ by arc-melting.

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	-

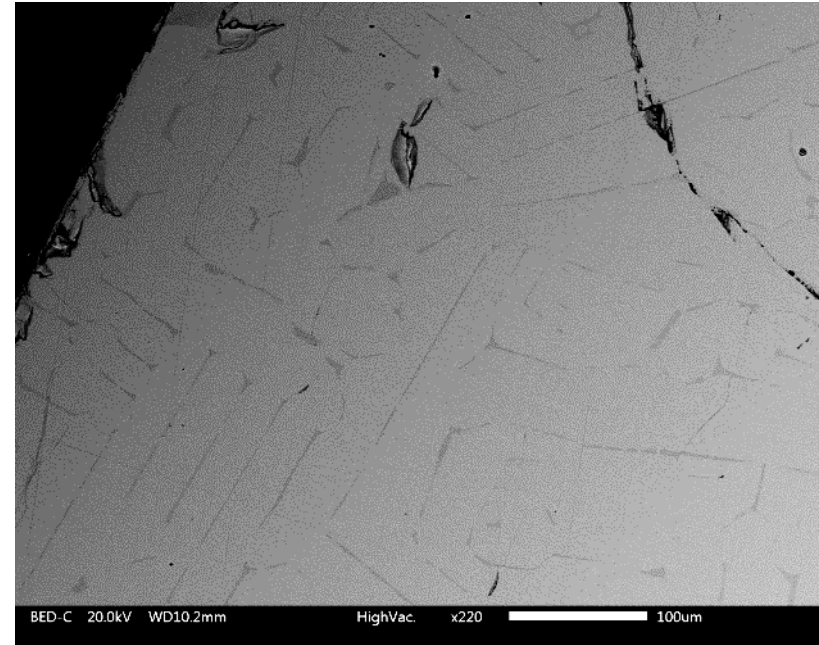
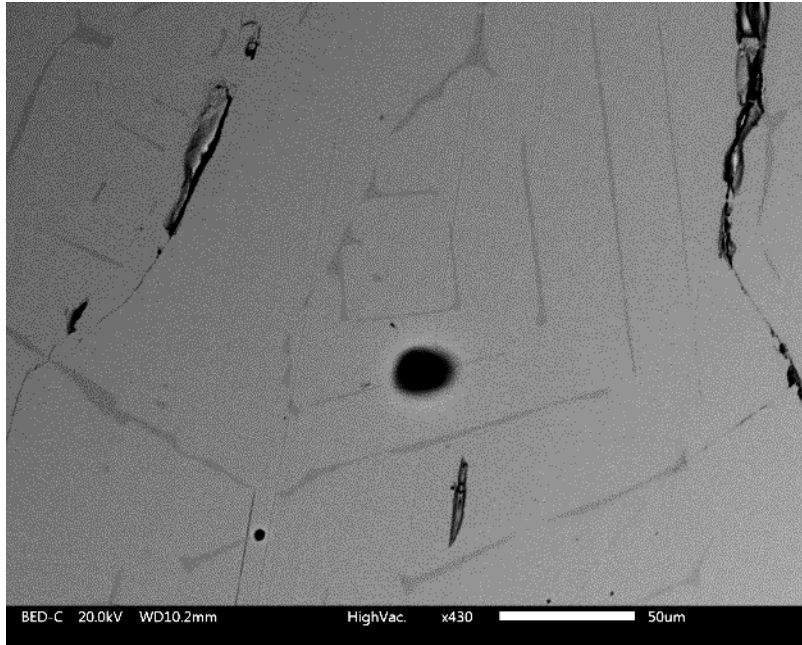
Mechanism of spiral microstructure formation



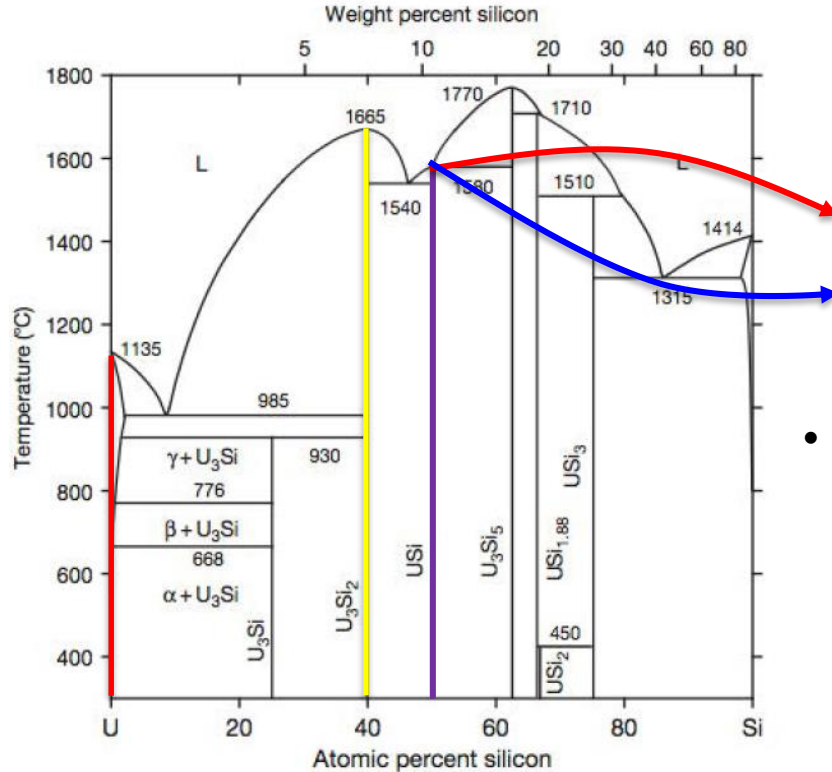
Mechanism of spiral microstructure formation



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