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**Characterization of Si - coated UMo Fuel Particles  
Before and After Interaction Annealing**

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**ABSTRACT**

In order to study the Si-coating layer formation and its effect as an interaction barrier, cylindrical compacts based on Si-coated atomized UMo fuel particles and dispersed in an aluminium matrix, were manufactured. The starting microstructure (as - coated) and after interaction annealing were characterized through both, optical and SEM with EDS analyses. After Si-coating treatment by pack cementation, carried out in vacuum for 3 hours at 1000°C, a silicon coating area was observed consisting of a thin layer (about 10 µm of thickness), comprised by 35% at. of Si and 65% at. of U plus oxygen. Regarding the coating treatment carried out under inert atmosphere (argon), diffusion of silicon into the UMo particles up to 22% at. was detected, besides 24% at. of uranium and 53% at. of oxygen at the surface of UMo particles. Nevertheless, no interaction layer was observed for coated UMo particles treated in Argon. After an interaction annealing, conducted in vacuum by 7 hours at 500°C, the Si-coated UMo particles, surrounded by aluminium, revealed a non-continuous interaction layer containing Al, U, Si and Mo. For the case of UMo particles coated in argon, the aluminium content was very low (4% at.) at the center of a very small particle. This beneficial effect might be caused by the presence of 1.7% at. of silicon in the surface and 0.7% at. in the center of UMo particle, and most likely forming a solid solution. This paper presents and discusses in detail the first results obtained for silicon coating of UMo particles, aiming to control the formation and composition of the deleterious fuel/matrix interaction layer observed in UMo dispersion type fuel.

## 1. Introduction

The attempts to control the interaction between the fuel phase and the aluminum matrix in dispersed nuclear fuel based on UMo type alloys have been only partially successful. The silicon addition to the aluminium matrix may not be the definitive solution, because some critical questions regarding the maximum amount of Si addition are still open [1]. The addition of silicon in the aluminium matrix has shown a reduced layer growth [2], and while showing its effectiveness in delaying the interaction, it may not prevent the formation of uranium aluminides type compounds, which are formed spontaneously and progressively until most of the UMo particles have been consumed by the fuel/matrix interaction [3].

The latest developments reported in the open literature are regarding to the UMo coating particles with interaction barriers (layers) formed in situ by several coating methodologies [4], such as PVD, solid state reactions as pack cementation, solid gas reactions and the formation of zirconium and uranium nitride layers [5]. The first interaction test results show that the layers formed on the surface of the particles are barriers that, in principle, delay or prevent interdiffusion of elements between the fuel UMo and Al-Si matrix. Similarly these layers inhibit the formation of undesirable compounds, which comprises the fuel/matrix interaction layer [6].

In this paper, the first results for silicon – coating of UMo particles conducted at CCHEN are reported. The coating tests have been carried out using the pack cementation methodology, in which the main mechanism is solid state diffusion between large UMo particles and a bed of fine pure silicon powder, in a temperature driven process.

## 2. Experimental Set - up

The UMo alloy was prepared in a laboratory induction furnace whose heating coil is placed inside of a closed chamber, capable of operating in a vacuum or protective inert atmosphere. The uranium 7% wt Mo alloy was prepared by melting natural uranium and pure molybdenum (99,5 wt% Mo). The UMo alloy was poured and casted into an Al<sub>2</sub>O<sub>3</sub>-coated graphite die, in order to produce cylindrical UMo pins.

The UMo powder was prepared through the Rotating Electrode atomization Process – REP. The centrifugal atomization parameters were: 36000 RPM, 60 DC Amps, applied to pin by a tungsten electrode and inert atmosphere (argon flow 10 liters/min). After atomization, the UMo particles were collected from the atomizer container and then classified in a set of Tyler mesh sieves.

The silicon coating of UMo particles was conducted at 900 and 1000 °C, for 1 to 3 hours in vacuum or inert atmosphere (argon). A pure silicon powder bed (100% < 45 µm), was used for coating the UMo particles (100% > 150 µm), applying the methodology known as cementation pack.

The samples for the interaction test were prepared blending aluminium powder with Si-coated UMo particles, and then pressing the mix into a die of 8 mm diameter, using a load of 260 MPa.

Each sample was placed at the center of a die of 25 mm diameter, filled by aluminium powder and res compacted to 330 MPa, resulting in a unique specimen usable for interaction annealing

and follow up by Scanning Electron Microscopy – SEM equipped with EDS microanalyses. These specimens were prepared through metallographic techniques, grinded and polished after each thermal treatment. The interaction annealing test was carried out under vacuum for 7 hours at 500 °C.

### 3. Results

The UMo pins were prepared with 413,1 grams of uranium and 31,5 g of molybdenum, after melting and pouring, the surface of UMo pins were machined to improve its alignment and cleaning.

Starting from 3 pins of UMo alloy, with a total mass of 145.1 grams, the centrifugal atomization produced a batch of 121.33 grams of UMo powder, with 10.49 grams of particles larger than 150 µm (used for Si coating). The total mass of UMo powder with proper sizes for fuel fabrication was 110.84 grams, 91.35 wt% of total atomized UMo particles.

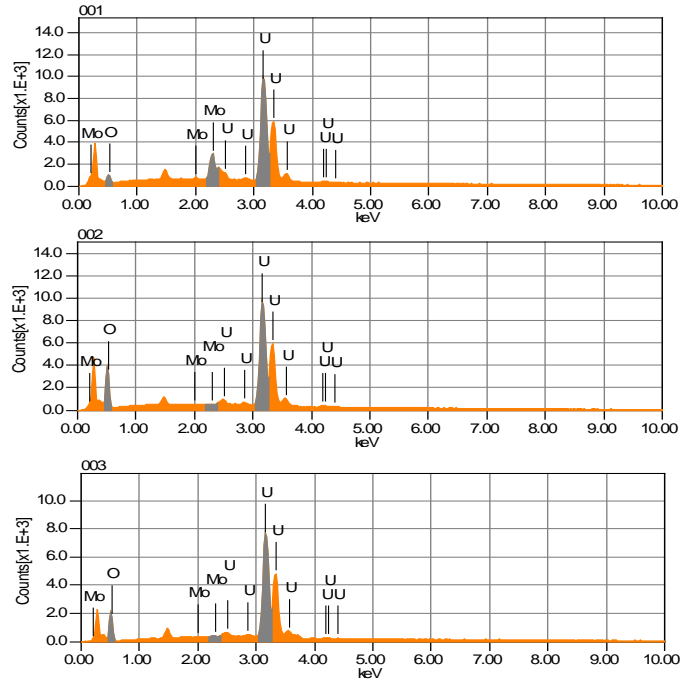
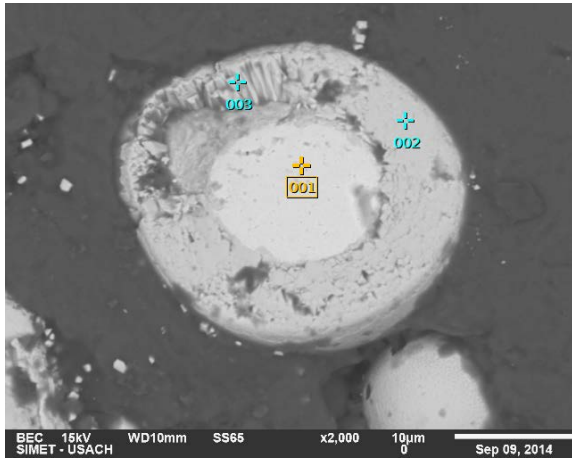
The size and distribution of UMo particles were analyzed with conventional Tyler Series sieving, and the size distribution is summarized in Table 1.

**Table 1.-** Granulometry of atomized UMo powder

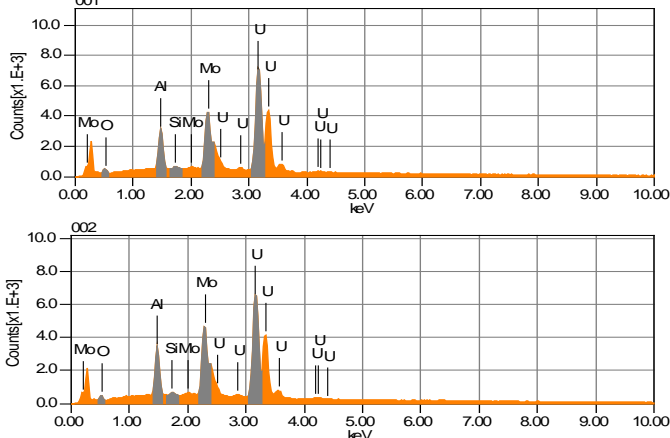
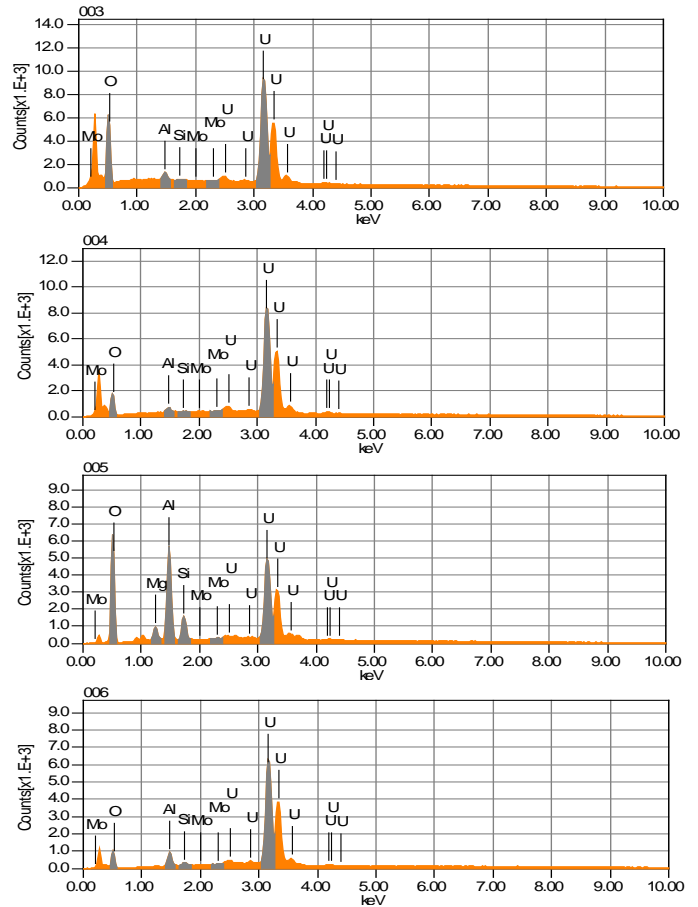
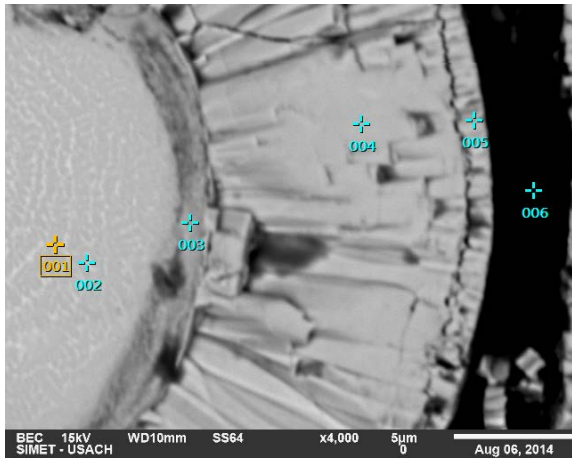
Tyler Sieve	Particle size (µm)	Material	
		(g)	(%)
# 100	> 150	10.49	8.61
# 100	< 150	44.96	91.35
# 170	< 90	22.30	54.30
# 230	< 63	16.27	35.92
# 325	< 45	27.31	22.51
Total		121.33	

The coating of UMo particles allowed adding important silicon content to the particles surface. When the Si coating treatment was conducted in vacuum, it was possible to observe a layer in some areas of the particle surface; meanwhile when the coating treatment was carried out in argon, the diffusion of silicon was not visible and its presence was confirmed only by means of EDS microanalysis.

The characterization of some atomized UMo particles without Si – coating allowed the detection of thick uranium oxide layers, most likely formed during atomization. These oxide layers showed columnar growth along the surface of the particles, regardless of its morphology. Figures 1 and 2 show UMo particles before and after the Si coating treatment, including EDS of the uranium oxide layer.

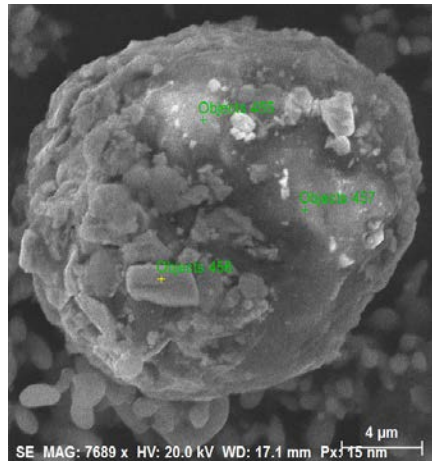


**Figure 1.** Cross-section micrograph of an uncoated UMo particle, before the annealing.

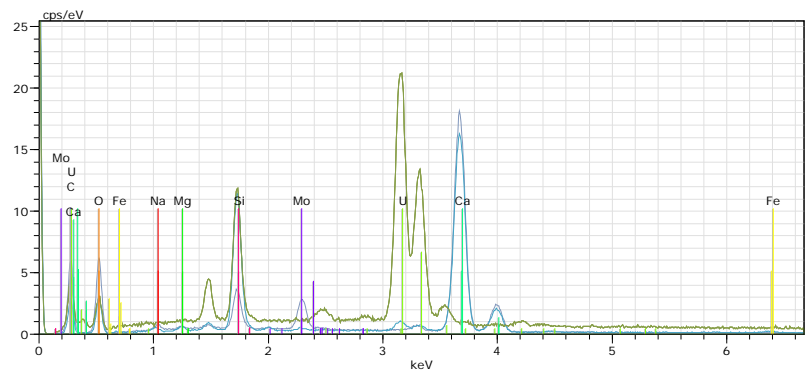


**Figure 2.** Cross-sectional micrograph of an uncoated UMo particle, after the annealing.

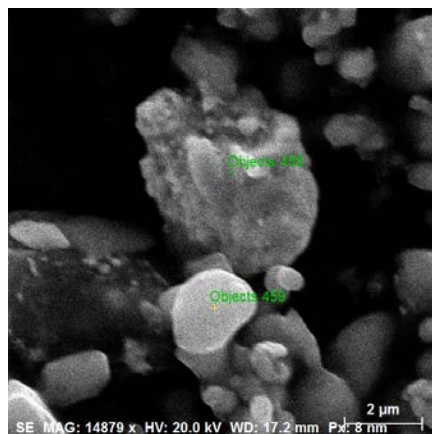
Figure 3 shows a Si coated UMo particle, including results of EDS microanalysis taken from selected points of its cross section surface. The silicon coating appears as a discontinuous layer with non-homogeneous thickness. Figure 4 shows compositional analyses of the surface for UMo and pure silicon particles



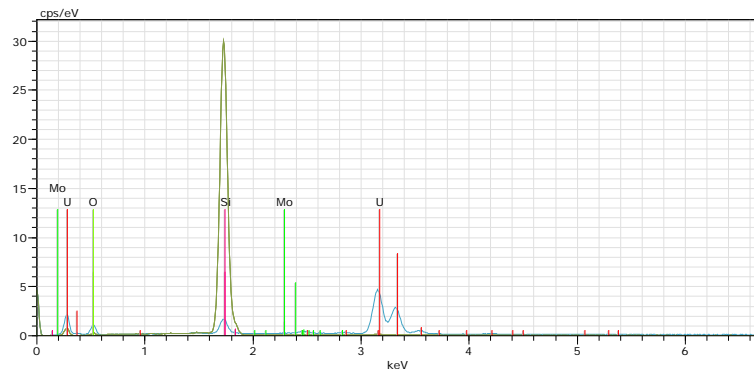
Point	Mass percent (%)				
	C	O	Si	Mo	U
455 UMo particle	16.72	26.92	11.17		3.73
456 UMo particle		7.04	8.84		84.11
457 oxide particle		49.18	3.02	5.70	1.38



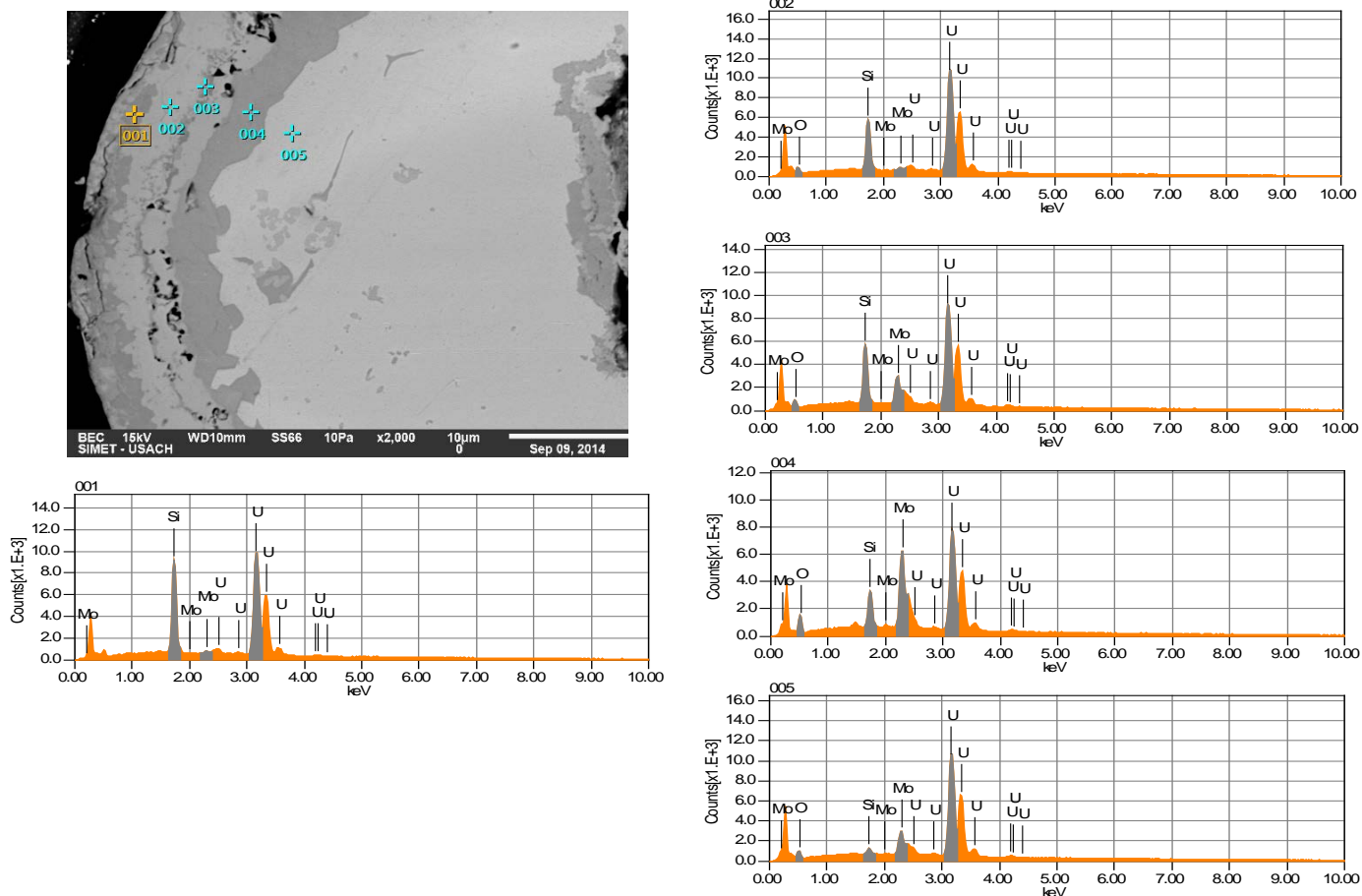
**Figure 3.** Si-coated UMo particle, SEM micrograph and EDS compositional microanalyses in selected points of its surface.



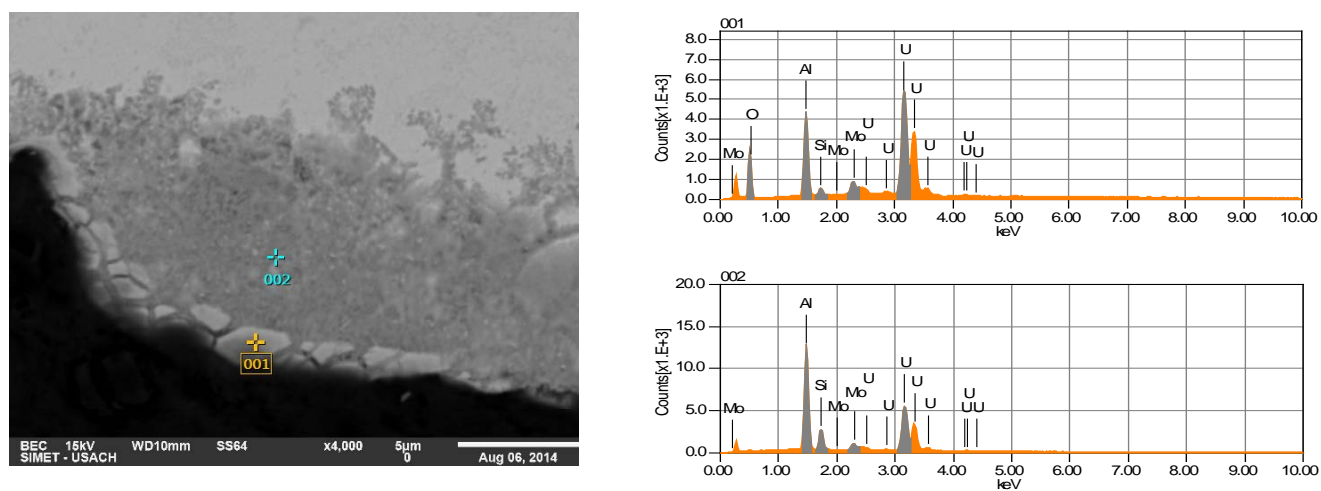
Point	Mass percent (%)		
	O	Si	U
458 UMo particle	12.04	5.50	82.46
459 Si particle	4.15	93.89	1.96



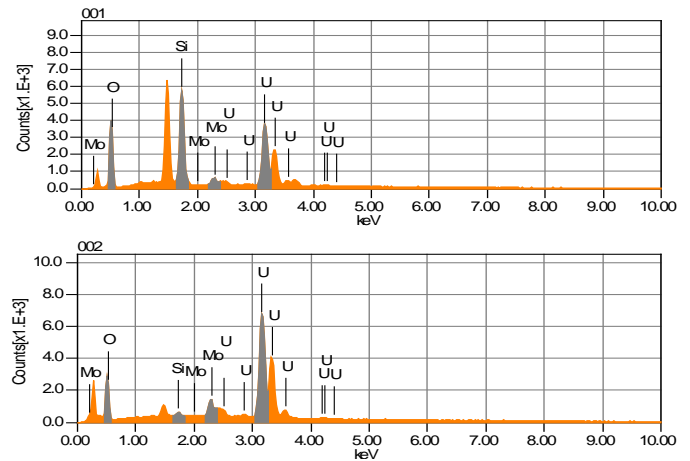
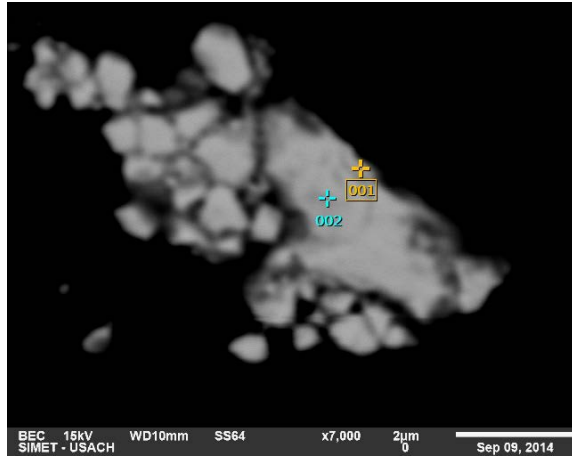
**Figure 4.** Si-coated UMo particle (gray), next to a pure Si particle (white), SEM micrograph and EDS compositional microanalyses at the surface of each particle.



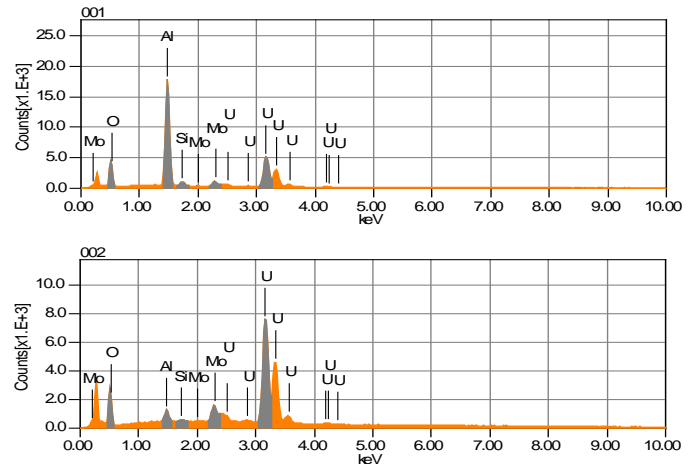
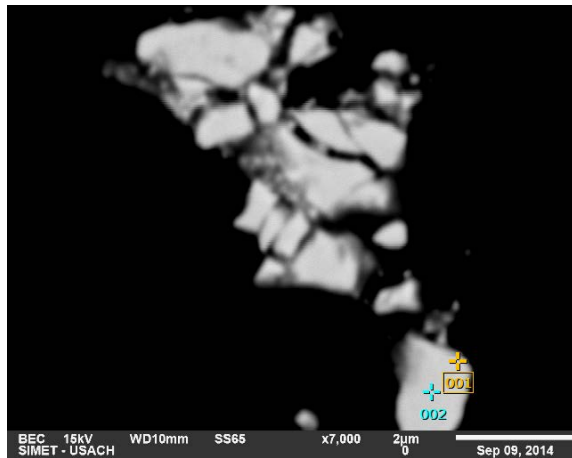
**Figure 5.** Cross-section micrograph of an UMo particle, Si coated in vacuum, before the interaction annealing. SEM micrograph and EDS microanalyses.



**Figure 6.** Cross-section micrograph of an UMo particle, Si - coated in vacuum, after the interaction annealing.



**Figure 7.** Cross-sectional micrograph of an UMo particle, Si - coated in argon, before the interaction annealing.



**Figure 8.** Cross-sectional micrograph of an UMo particle, Si - coated in argon, after the interaction annealing.

#### 4. Discussion

In Figure 1, EDS microanalyses detect small amounts of aluminium in the UMo particle but this content is most likely contamination from the aluminium powder used as filler, and carried during the polishing of the sample. Nevertheless, in figure 2, the aluminium content is non negligible (2.3 wt % and 2.7 wt % in points 1 and 2 respectively), in this case, the presence of this element is evidence of the fuel/matrix interaction.

Figure 5 reveals the coating layer produced by vacuum Si-coating. The Si content is higher at the surface and decreases towards the center of the particle. Microanalysis at point 4 reveals an increase in Mo content (12.58 wt %), higher than the content detected at point 5 (4.35 wt %), and even more than the nominal Mo content (7.0 wt %). The interaction layer in point 4 is, likely, constituted by an U-Si mixed oxide.

The sample of Figure 6 is an UMo particle Si - coated in vacuum, after interaction annealing (500°C/7 hours). The aluminum content in point 2 (13.01 wt %) is higher than the same element at point 1 (5.01 wt %). Except in Figure 8, corresponding to UMo particle Si- coated in Ar, where the Al content in the center of a fine particle was very low (0.76 wt%).

The silicon content at the surface of the UMo particle coated in argon, point 1 in Figure 7, is around 10 wt %, nevertheless the silicon is most likely present as a solid solution and not as a visible layer.

An alternative to improve the homogeneity in the thicknesses of the silicon coating layer could be to use a blending of silicon and UMo powder at high temperature (900-1100 °C), into a capsule sealed under vacuum, instead of using a passive Si-powder bed.

## **5. Conclusions**

The Si-coating of UMo particles is an effective alternative for reducing the fuel/matrix interaction; nevertheless, the results can be improved in terms of obtaining coating layers with more homogeneous thicknesses.

The silicon coating carried out in vacuum reveals the formation of Si-rich layers at the surface of the UMo particles. However, these layers do not avoid the interaction, and the EDS analysis detects a high content of aluminium into the particles.

In the case of silicon coating carried out in argon atmosphere, the silicon diffuses into the UMo particles and is revealed by EDS analysis, but no Si-layer is observed. The silicon presence constitutes an effective barrier for IL formation and fuel/matrix interaction. This effect was evidenced by the very low aluminium content at the center of these Si-coated UMo particles, as detected by EDS analysis.

Based on the results reported in this paper, the immediate following activity will be to prepare a new coating system for UMo-Si powder blending using a rotating mixer placed inside a furnace in order to improve the homogeneity in the thickness of the silicon coating layer.

## **8. Acknowledgements**

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## 9. References

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