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Determination of Ba, Cs, Mo, Zr and U in SIMFuel samples by ICP-AES and ICP-MS for the study of fission products behavior during a nuclear severe accident

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Context and purpose

The development of realistic models for fission products behavior during a nuclear severe accident requires experimental data on fission products speciation into the fuel. In this context, several batches of dense UO_2 samples containing fission products surrogates under different chemical forms have been prepared and sintered, to be further submitted to thermal treatments in order to characterize fission products speciation under controlled temperature and oxygen potential conditions.

To that end, a large number of as-fabricated samples from these experiments were analyzed by ICP-AES and ICP-MS after dissolution. A separation step by liquid chromatography on UTEVA resin was essential before the ICP-AES measurements to overcome the problems of spectral interferences and matrix effects caused by uranium.

This study emphasizes the complementarity of these two techniques in nuclear fuel characterization. The advantage of the ICP-AES analysis on simultaneous device is explained in details. The importance of the integrated collision reaction cell in ICP-MS to avoid many problems of polyatomic interferences for the quantification of Cs and Ba is highlighted.

Pressurized Water Reactor (PWR)

Fuel assembly

Nuclear fission reaction

Fission yields : most likely fission products produced

SEM micrography

SPS apparatus

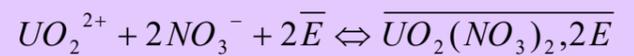
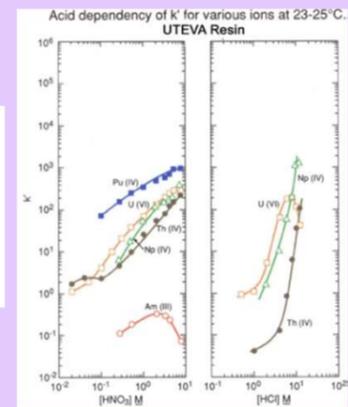
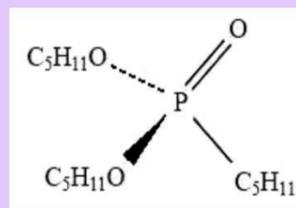
Development of a Spark Plasma Sintering (SPS) route to synthesize SIMFuel samples

$UO_2 + 0.9wt\% Cs_2MoO_4$

SPS is a Field Assisted Sintering Technique which uses an electrical field to facilitate sintering

Goal of the study = chemical analysis of Ba, Cs, Mo, Zr and U by ICP-AES and ICP-MS

Separation by liquid chromatography on UTEVA resin after dissolution of SIMFuel samples



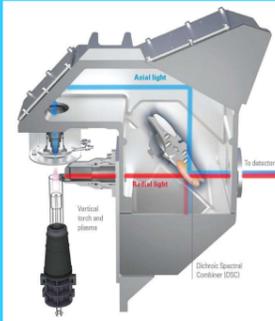
Remark : separation only for analysis by ICP-AES

Agilent Technologies 5100 SVDV apparatus



Seaspray nebulizer, cyclonic chamber, axial view, CCD detector

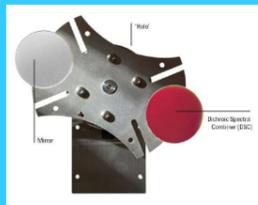
Four modes of operation in the 5100 SVDV configuration



Vertical torch



The mode selector component



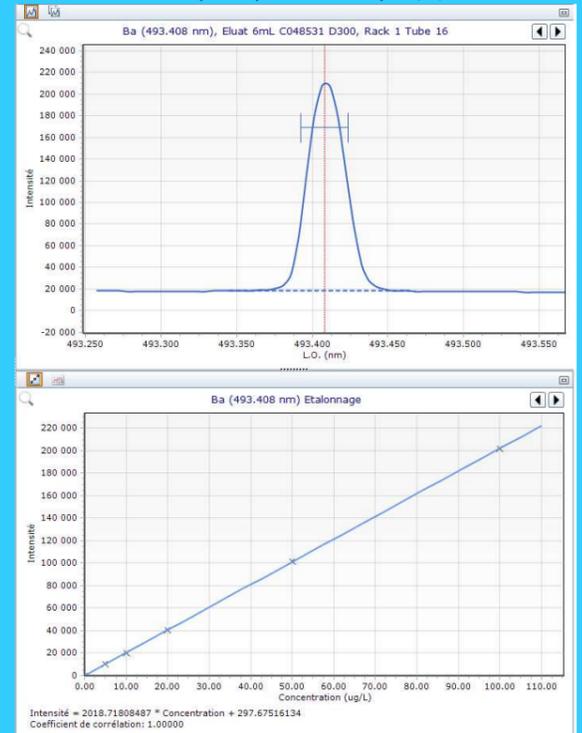
Analysis by ICP-AES



Operating conditions for quantitative analysis:

Conditions communes	
Risques:	10
Vitesse de pompe (l/min):	12
Débit de prélèvement (s):	60
Durée de stockage (s):	60
Activer le stockage intelligent:	<input type="checkbox"/>
Conditions de mesure:	
Durée de lecture (s):	10
Puissance RF (W):	1.20
Durée de stabilisation (s):	30
Mode de visualisation:	Anal
Hauteur de visée:	8
Utiliser plusieurs conditions:	<input type="checkbox"/>

Example of quantitative analysis (Ba)



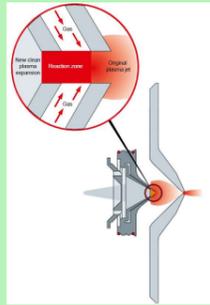
Analysis by ICP-MS

Analytik Jena PQMS Elite apparatus



Micro-concentric nebulizer, scott spray chamber cooled to 3°C with Peltier effect, DDEM detector

Integrated Collision Reaction Cell (iCRC)



Analysis with iCRC for the management of spectral interferences

Operating conditions :

Parameter	Value
Paramètres débits (l/min)	
Débit plasma	9.0
Débit auxiliaire	1.80
Gaz de gainage	0.00
Débit nébuliseur	0.98
Alignement Torche (mm)	
Prof. Echant.	5.0
Aides	
Puissance RF (W)	1.25
Vitesse Pompe (l/min)	15
Durée de stabilisation (s)	60
Octave Ionique (V)	-0.65
Optique Ionique (V)	
Première Lentille Extraction	-114
Deuxième Lentille Extraction	-102
Troisième Lentille Extraction	-0.65
Lentille opposée	-0.28
Lentille Miror Gauche	94
Lentille Miror Droit	62
Lentille Miror Bas	55
Lentille entrée	-1
Entrance Plate	-82
Base Flap	-4.4
Flow Rate	0.0
iCRC (l/min)	
Source Gaz slimmer	112
Débit Slimmer	120
Matrix	
Débit (l/min)	

Examples of spectral interferences :

- ^{133}Cs interfered by $^{98}Mo^{25}Cl$, $^{96}Zr^{37}Cl$, $^{96}Mo^{37}Cl$...
- ^{138}Ba interfered by $^{98}Mo^{40}Ar$
- ^{137}Ba interfered by $^{97}Mo^{40}Ar$, $^{100}Mo^{37}Cl$

Results

Examples of final results in mg/g

Sample number	C048531	C048532	C067011	C067012	C067013
Ba	6,930 ± 0,693				
Cs	0,109 ± 0,011	1,196 ± 0,120	2,125 ± 0,213	1,579 ± 0,158	1,510 ± 0,151
Mo	2,479 ± 0,248	1,144 ± 0,114	0,348 ± 0,035	0,122 ± 0,012	0,154 ± 0,015
Zr	7,476 ± 0,748				
U	824,6 ± 16,5	860,1 ± 17,2	848,5 ± 17,0	858,2 ± 17,2	816,7 ± 16,3

Conclusions

The results of chemical quantitative analysis obtained by ICP-AES and ICP-MS are very important and they allowed acquiring valuable information for the continuation of the study. Indeed, the dispersion of the additives in the UO_2 matrix after sintering is not homogeneous in the pellets. The initial mixing of the different powders has not been effective. This is demonstrated by the chemical characterizations performed on several pellets from the same initial batch which showed that the final amount of additives varied a lot from a sample to another. Moreover, important Cs and Mo release took place during the sintering.