

# CORROSION OF TRIURANIUM DISILICIDE IN VAPOR UNDER SIMULATED PWR CONDITIONS

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## 1. Introduction

Triuranium disilicide is a promising candidate for an accident tolerant fuel (ATF). Compared to the traditionally used uranium dioxide, it has a high uranium capacity and thermal conductivity, which increases with increasing temperature. However, as reported [1-2], the stability of triuranium disilicide in water vapor is very low. It is believed that due to the formation of the hydride phase, internal stresses arise, leading to sample pulverization [3].

The purpose of this work was to experimentally simulate the interaction of triuranium disilicide with water vapor under conditions close to reactor conditions and to observe the evolution of sample oxidation. The reaction kinetics was also evaluated, since zirconium, which acts as an element of the fuel cell shell, is an effective hydrogen absorber and its presence in the system can affect the kinetics of hydrogenation of triuranium disilicide.

## 2. Experiment details

To determine the corrosion resistance of triuranium disilicide in water, autoclave tests were carried out at various temperatures (350–600°C) and holding times (5–1440 min). The tests were carried out in a steel autoclave in a muffle furnace. The temperature field inside the furnace was controlled by a thermocouple. Samples and distilled water were loaded into the autoclave and sealed in an argon box. Some of the samples were pre-polished to compare the structure before and after corrosion. A tablet of triuranium disilicide was placed in an E110 zirconium alloy tube acting as a sheath material. The volume fraction of distilled water was selected so that at the test temperature the vapor pressure was 160 atm.

## 3. Results and discussion

Tablets from triuranium disilicide showed resistance to oxidation and fragmentation at a temperature of 350°C up to 24 hours of exposure. After 350°C, the samples quickly pulverize; at 400°C, the exposure time without pulverization does not exceed 1 hour, and at 450°C, 10 minutes. At a higher temperature, pulverization took place almost instantly, most likely, even in the process of heating the autoclave to the holding temperature.

Using SEM and EPMA, the structures before and after corrosion were compared. It was shown that the amount of oxygen in the surface layer under such conditions changes from 4 wt.% to 25 wt.%, silicon from 7.2 wt.% to 3.6 wt.%, uranium from 88.8 wt.% to 68.6 wt%. The growth of the second phase is observed, the inclusions of which were previously uniformly distributed over the area. A typical example of microstructure comparison is shown in Figure 1, the corresponding EPMA spectra are shown in Table 1. No exposure time dependence was found,

the presumably cleaned surface of triuranium disilicide quickly reacts with water vapor to saturation.

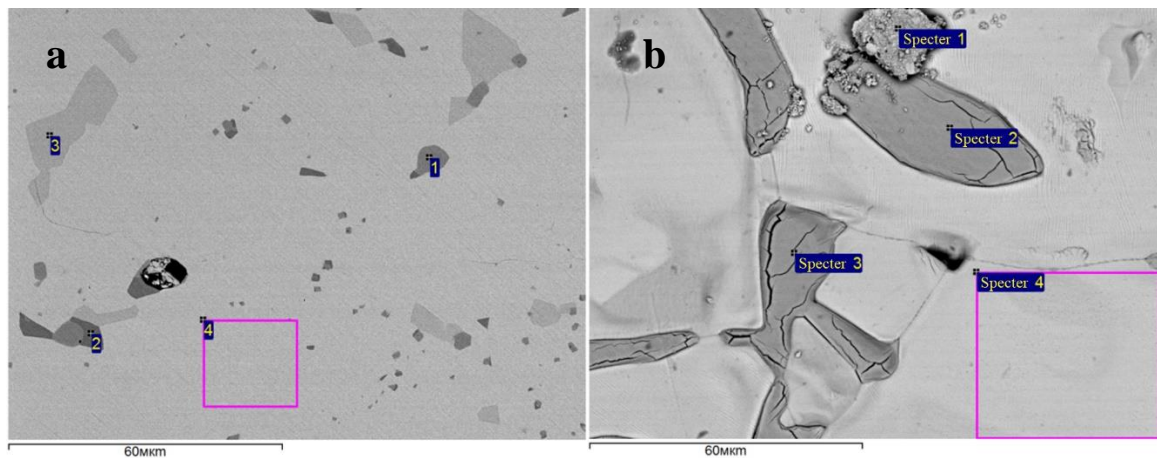


FIG.1 – a) before and b) after corrosion at  $T=350^{\circ}\text{C}$ ,  $t = 8$  hours

TABLE 1. EPMA FOR FIG.1

Specter	O, w. %	Si, w. %	U, w. %
1(a)	4,0	7,6	88,9
2(a)	3,7	7,5	88,8
3(a)	4,0	8,2	87,8
4(a)	3,5	7,2	89,3
1(b)	25,2	4,2	70,6
2(b)	25,9	3,9	70,2
3(b)	24,9	3,8	71,3
4(b)	21,3	4,2	74,5

The progression of tablet oxidation can be observed in Figure 2, which shows the change in the concentration of the main elements on the surface of the tablet during the oxidation process. Several stages can be distinguished. At stage I, a film of silicon oxides is formed on the surface of the sample, which is rapidly destroyed already at a temperature of  $350^{\circ}\text{C}$ . Being quite thin, this oxide film gives a strong color effect due to interference. At stage II, the silicon oxide film is destroyed and replaced by a film of uranium oxides in the order  $\text{UO}_2 - \text{U}_3\text{O}_8 - \text{UO}_3$  in accordance with the phase analysis data for samples with a long exposure time. Intergranular boundaries become visible, with an increase in exposure time they become clearer and destruction begins to occur along them. At stage III, almost no silicon remains in the surface layer of the tablet, and it consists entirely of uranium oxides. Sample pulverization starts. At stage 4, the sample is no longer a tablet, but a powder. As the temperature rises, the length of the first three stages is greatly reduced.

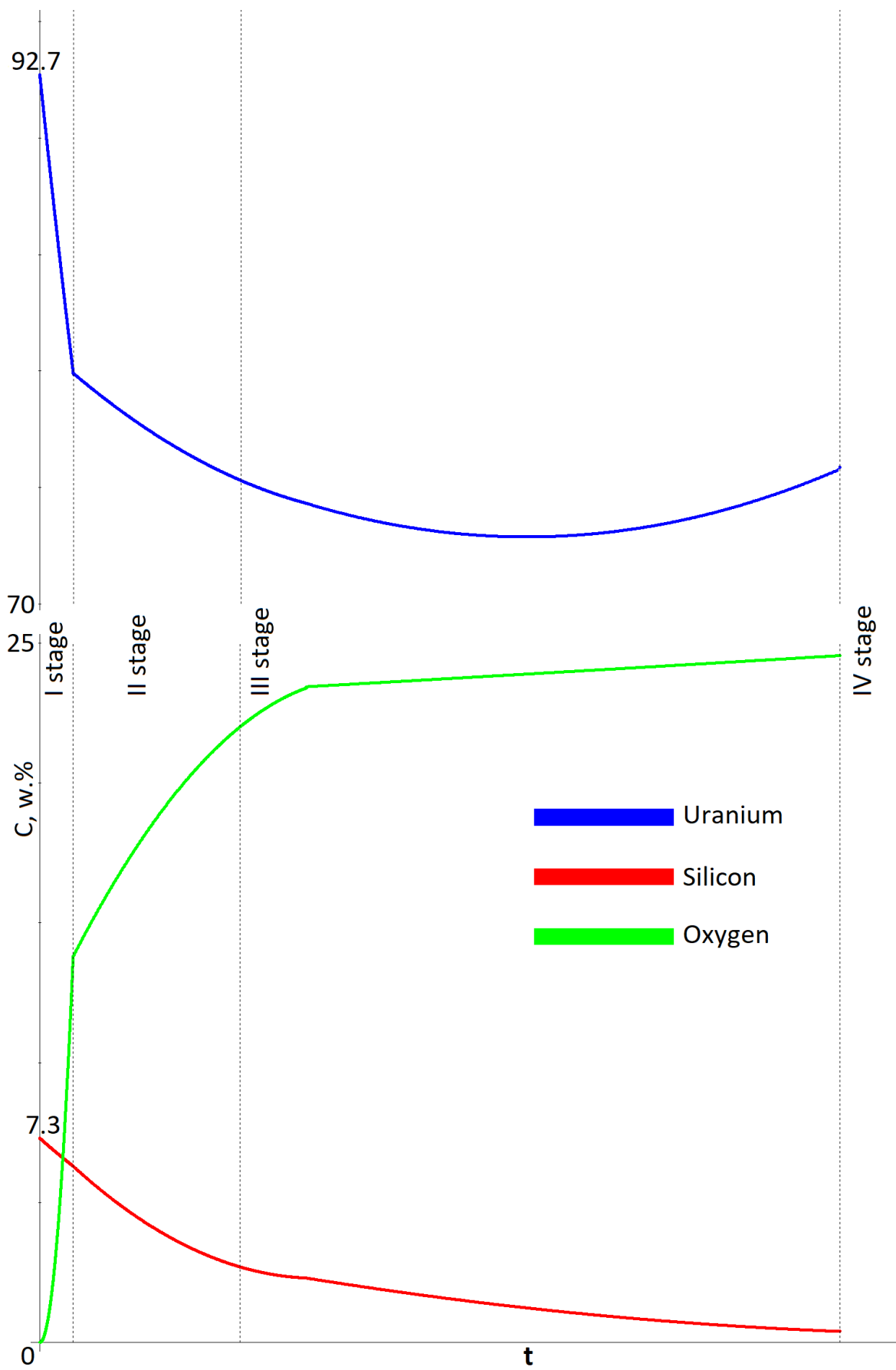


FIG.2 – Change in the concentration of the main elements

#### 4. Conclusion

At a temperature of 350°C under PWR conditions, triuranium disilicide samples retain structural stability and shape up to 24 hours of exposure. Oxidation of triuranium disilicide occurs with the formation of a silicon dioxide film at an early stage, which protects the material from further oxidation, when the temperature increases to 400°C or sufficient exposure time, the film is destroyed and replaced by a film of uranium oxides in the order  $\text{UO}_2$  -  $\text{U}_3\text{O}_8$  -  $\text{UO}_3$ . With a further increase in temperature, triuranium disilicide is fragmented and ground to the state of a powder, mainly consisting of uranium oxides due to the active oxidation of the enlarged surface. The destruction of triuranium disilicide in the course of such fragmentation proceeds along the grain boundaries.

#### REFERENCES

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- [3] Mašková S., Miliyanchuk K., Havela L., Hydrogen absorption in  $\text{U}_3\text{Si}_2$  and its impact on electronic properties, *Journal of Nuclear Materials* 487 (2017): 418-423.