Behavior of UO₂ and FISSIUM in Sodium Vapor Atmosphere at Temperatures up to 2800° C

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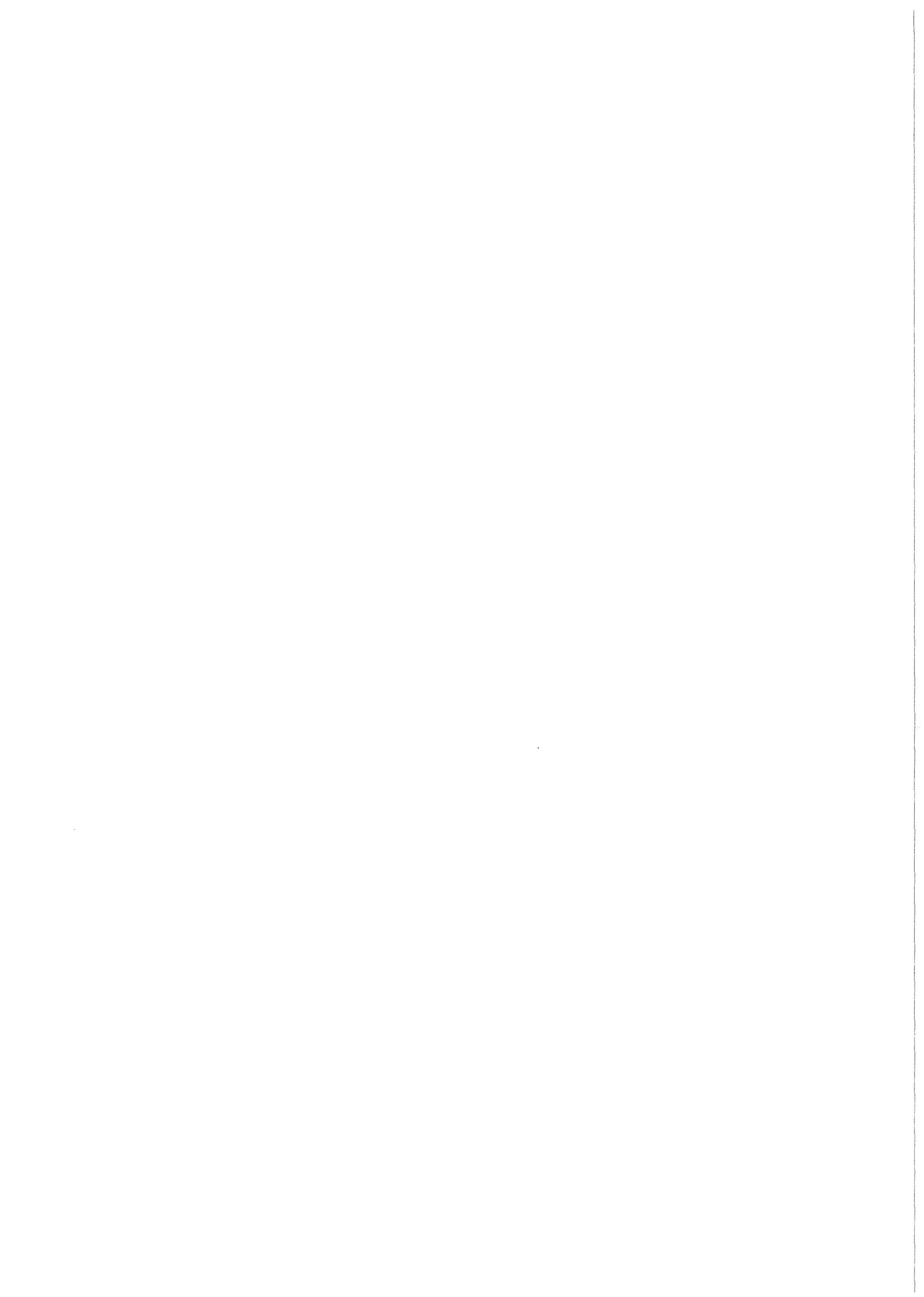
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BEHAVIOR OF UO_2 AND FISSIUM IN SODIUM VAPOR ATMOSPHERE

AT TEMPERATURES UP TO 2800 °C [*]

Abstract

In case of a HCDA a rubble bed of fuel debris may form under a sodium pool and reach high temperatures. An experimental technique was developed to study the behavior of fuel and fission products in out-of-pile tests in a sodium vapor atmosphere.

Evaporation rates of UO_2 were measured up to 2800 °C. The evaporation was found to be a complex process, depending on temperature and the 'active' surface.

Evaporation restructures the surface of the samples, however no new 'active' surface is formed.
UO_2 forms sometimes well shaped crystals and curious erosion products. The efficiency of the used condenser/filter lines was higher than 99.99 % . In case of a HCDA all the evaporated substances will condense in the sodium pool.

Thermal reduction of the UO_2 reduces the oxygen potential of the system. The final composition at 2500 °C was found to be $UO_{1.95}$.

The only influence of the sodium vapor was found for the diffusion of UO_2 into the thoria of the crucible. Compared with experiments in an atmosphere of pure argon, the diffusion rate was reduced.

VERHALTEN VON UO_2 UND FISSIUM IN EINER NATRIUMDAMPF-ATMOSPHÄRE BEI TEMPERATUREN BIS ZU 2800 °C [*]

Zusammenfassung

Im Falle eines HCDA kann sich eine Schüttung von Brennstoffstücken unter einem Natriumpool bilden, in der hohe Temperaturen vorkommen können. Es wurde eine Versuchstechnik entwickelt, um das Verhalten von Brennstoff und FISSIUM in einer Natriumdampf-Atmosphäre out-of-pile untersuchen zu können.

Die Verdampfungsraten von UO_2 wurden bis zu einer Temperatur von 2800 °C bestimmt. Die Verdampfung ist ein komplexer Prozeß, der von der Temperatur und der 'aktiven' Oberfläche der Probe abhängt.

Bei der Verdampfung wird die Oberfläche der Proben sehr stark restrukturiert, es wird jedoch keine neue 'aktive' Oberfläche gebildet.
Bei hohen Verdampfungsraten kondensiert das UO_2 in Form von gut ausgebildeten Kristallen, durch Erosion entstehen daraus oft kuriose Gebilde. Der Wirkungsgrad der gesamten Abscheidestrecke war höher als 99.99 % . Für den Fall eines HCDA bedeutet das, daß die verdampften Stoffe im Natriumpool vollständig absorbiert werden.

Das Sauerstoffpotential des Gesamtsystems wird durch thermische Reduktion verringert. Die Endzusammensetzung des Oxides betrug bei 2500 °C $UO_{1.95}$.

Nur für die Diffusion von UO_2 in Thoriumdioxid wurde ein Einfluß von Natriumdampf beobachtet. Die Diffusionsgeschwindigkeit war geringer als bei Versuchen in reinem Argon.

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Content

1. Introduction

One accident scenario for a liquid metal fast breeder reactor (LMFBR) assumes that the reactor core has suffered catastrophic disassembly with the rubble of reactor fuel and cladding covered with liquid metal coolant and some structural or core-catcher material (1). An important, yet unanswered question, is then how much heat must be extracted from the rubble (2).

The produced heat is the decay heat of the radioactive nuclides, mainly from the fission products. The temperatures in the rubble bed depend strongly on the total amount of these radioisotopes, and also from their distribution between the different liquid and solid phases. At least parts of the bed will dryout and reach high temperatures. Crusts will isolate this part of the bed against the underlying structures, and a sodium vapor layer will be between the rubble bed and the sodium pool.

An out-of-pile experimental program is under way at KFK to study the behavior of radionuclides and fuel under such conditions (3). The behavior of UO_2-fuel will be described in this report.

2. Experimental facility

The facility SUSI is used for the experiments (4). It may be considered to consist mainly of a stainless steel vessel with a volume of 1100 liters, with a water cooled induction coil in the center part. Auxiliary systems are for evacuating to a pressure of 10^{-3} mbar, inert-gas supply and to deliver liquid sodium at a rate of 0.5 to 10 grams per minute into the vessel. This

sodium supply system consists of a hydraulic press, squeezing
solid sodium into a melting crucible. The transport gas for the
molten sodium is Ar-6.0.

All of the experimental arrangement is build inside of the
vessel. Fig.1 shows a photography of the arrangement. The atmosphere
in the vessel was always Ar-6.0.

3. Experimental arrangement

3.1 High temperature part

Figure 2 shows the high temperature part of the arrangement.
The lower part of a 30 cm long tungsten receptor is in the
induction coil. This is the furnace to heat the samples.
The samples are kept in thoria crucibles, which are in a
protective tungsten cup.

The sodium is evaporated by heating to 1000 to 1100 °C and blown
to the sample through a tantalum tube. In some tests Ar-6.0 is
used instead of sodium, also pre-heated in the evaporator.
For easier retrievability of condensed substances a thin walled
tungsten cylinder is placed inside of the receptor.

In most of the experiments the receptor is used without any
thermal shielding. Radiation losses limit the possible
temperature to about 2600 °C. For higher temperatures
granulated thoria isolation in an alumina beaker is used.

3.2 Temperature measurement

The temperature measurement is done by optical pyrometers from
outside of the vessel at the receptor (fig.2).
Two-wave-length-pyrometers were used, measuring at 440 and 653 nm,

or at 850 and 1000 nm . The pyrometers got calibrated by the
PTB-Braunschweig with tungsten lamps. Because in the experiments
also a clean tungsten surface is seen in pure argon,
the values are considered to be reliable. The temperature
at the position of the sample of course is somewhat lower. It
is obtained from special calibration experiments, by observing
a W-sample from the top.

The temperature difference between the receptor wall
and the sample position was not constant. For e.g. 2500 °C
is was between zero and 120 °C. It was independent on a preheated
gas stream through the Ta-tube. Probably temperatures from the top
are influenced by thermal convection inside of the receptor.

Many attempts were made to solve the problem of the sample
temperature. Measurements with W-Re-thermocouples were not
successful for temperatures above 2100 °C. However good values
were obtained from the total amount of evaporated tungsten
(determination as described for other elements below). By using
functions as given by Jehn (5) the calculated temperatures were
always within ±50 °C of the temperature measured at the receptor.

Finally it was assumed, that the receptor-wall temperature
is also a good measure for the sample temperature. In this paper
therefore always receptor temperatures are given. One should keep
in mind however, that the real sample temperature could be somewhat
lower.

3.3 Low temperature part

The low temperature part may be considered to begin at the
evaporator, which has a constant inside temperature of
1050 to 1100 °C. Up to this part sodium will be in the vapor
phase.

From the sample evaporating substances get transported by the sodium
vapor and the Ar-transport gas first to a condenser, and further
to parallel filter lines. The filters consist of sintered
stainless steel plates or cylinders with a pore size (nominal)
of 0.5 μ (SUPRAMESH of PALL-company). In case of sodium
experiments these filters are pre-heated to 350 °C to avoid
clogging. The efficiency of a filter-line is better than
99.99 % . Parallel lines could be used by opening or closing
ball valves at a manifold from outside of the SUSI-vessel.

4. Experimental conditions

4.1 Samples

For the experiments artificial fuel with fission products
(FISSIUM) , and UO_2 - pellets from the NUKEM-company,
are used. It is not possible to use plutonium in our laboratory.
FISSIUM is an artificial mixture of UO_2 and fission products,
simulating a burn-up of 10% , sintered at a temperature of
1500 °C for 2 hours. It was fabricated in the facility FIFA (6).
For some fission product elements radioactive tracers are used,
e.g. for Zr, Nb, Ce and Ru.
In addition pure UO_2 pellets were fabricated by the same method
as the FISSIUM and used in some experiments.
(To simulate the destroyed core, stainless steel 1.4970 was added
in some experiments. Results of such experiments were not reported
in this paper.)

The sample weight was between 10 and 70 grams. Normally standard
size pellets were used. In some experiments however broken
pellets were used to increase the surface.

4.2 Atmosphere

The experiments were performed at a total pressure of 1 bar.
In the tests the atmosphere at the samples was either
Ar-6.0 or an Ar-6.0/ Na-vapor mixture with a sodium partial
pressure of 0.2 to 0.3 bars. The gas was blown to the surface
with a rate of about 25 l/min. The gas stream is not hitting the
surface directely. It is leaving the Ta-tube very turbulent
and just filling the lower part of the receptor with 'stirred' gas.
Fig.3 shows a photography of the sodium-vapor leaving the Ta-tube.

The oxygen partial pressure may be an important parameter in the
experiments. In case of no sodium attempts were made to measure
it with an electrochemical analyzer (ENDOMESS). It has been
always below 0.1 ppm by volume (below 10^{-7} bar). The coil of
the evaporator is made of stainless steel, and traces of oxygen
react probably with the chromium . In experiments with
sodium the partial pressure could not be measured. The
oxygen concentration of the used sodium was in the range of
10 ppm by weight. It can be assumed, that also in experiments
with sodium, because of the reaction with the chromium of the
steel, the oxygen partial pressure was below 0.1 ppm by volume.

(The fact that temperatures could be calculated from the tungsten
evaporation rates (above) confirm the very low oxygen partial
pressure.)

4.3 Procedure

The samples got heated to the wanted temperature with a heating
rate of 50 °C per minute, then the temperature was kept constant
for the predetermined time, and finally the cooling rate was
100 °C per minute. In some earlier experiments the temperature was
raised stepwise.

In experiments with sodium the sodium stream was started
at a sample temperature of about 1400 °C. The temperature was
kept at this level until a thermocouple at the evaporator showed
that the sodium reached the receptor (usually after 2 to 3 minutes).

Figure 4 shows schematically the total procedure prior, during and
after the experiment in the SUSI-vessel.

The most time consuming steps are after the heating. Sodium from
all parts of the condenser and filter lines have to be converted to
NaOH and $NaNO_3$, using the technique of conversion by N_2/steam
or converting it with methyl-alcohol. All surfaces were washed
with nitric acid and treated in an ultrasonic bath. Often large
volumes of solutions were obtained, containing corrosion products
from the used stainless steel parts as well. The solutions were
prepared for the chemical analysis, which was done using an ICP-6000
of Perkin Elmer. Some elements had to be determined by neutron
activation analysis, e.g. cesium. For some elements evaporation
rates were too low and ICP-detection limits to high. For these
elements radioactive tracers were used and gamma spectroscopy
applied.

Uranium, which is the only element described in this report,
was determined always by ICP.

4.4 Performed experiments, range of parameters

So far results could be obtained from 34 experiments, with
temperatures between 1400 and 2800 °C. The other parameters
were : with or without sodium and steel, flow rate of the
atmosphere, heating and cooling rates, specific surfaces of the
sample, the amount of the samples and the sample material (UO_2
of different type and FISSIUM).

5. Results

5.1 Density and open pores

Because of the fabrication process and the high concentration
on fission products, the used FISSIUM had a geometrical density of
7.5 g/cm^3, and contained 28 % open pores. Pellets of pure UO_2
were fabricated by the same process, they had a densy of 9.8 and
only 2.7 % open pores. Comparative tests were done also with
different types of UO_2 pellets from RBU, they had densities
of 10.3 and less than 1 % open pores.

Within the error limits density and open pore volume did not change,
regardless if or not sodium was used. This result has been
surprizing and can not be explained completely, because the
metallographic picture especially of the FISSIUM was drastically
changed. Probably there are compensating effects of densification
and creation of new pores by evaporation.

5.2 Rate of evaporation

Evaporation rates of UO_2 at high temperatures were reported
in the literature several times (e.g. 7 and 8). However the
experimental conditions were always different from these in a
HCDA.

In the described experiments rates were determined by two ways:
weight loss determinations for pure UO_2 pellets, and
determination of the total amount of evaporated uranium from all
parts of the arrangement. The second method was the only one
usable for FISSIUM.

Using the geometrical surface of the pellets or broken pellets,

the evaporation rates as shown in fig.5 are obtained.

In spite of the scattering of the values there is no influence
to be seen by the sodium vapor. That means sodium vapor behaves
like an inert gas. This was to be expected because at the
temperatures of the experiments there are no chemical compounds
between sodium and fuel stable enough to influence the
evaporation rate; and for lower temperatures the evaporation rates
were too small to be measured.

Furthermore within the investigated range of 0.9 to 3 cm^2/g
there is no influence of the specific surface, and within 0.6
to 3 cm no influence of the high of the stacked pellets
or particles.

An Arrhenius-fit gives the evaporation rate as a function of the
temperature as

$$\log ER = 2.29 - 11100/T$$

With this the heat for the total evaporation process is obtained
to

$$\Delta H_{ER} = -212 \ [kJ/mol]$$

In this function the evaporation rate ER is given in $[mg/cm^2 \bullet s]$
and the temperature T is given in K.

5.3 Discussion of the rates of evaporation

The table compares measured values with such from the theory and
the literature.

Using a Knudsen-function (9) for calculating maximum evaporation

rates, with vapor pressure data of Ackermann(10), calculated rates
are in a rather good agreement with data measured by Felix (7)
in vacuum. That means the evaporation rates in vacuum are
controlled by the vapor pressure. The heat of evaporation of
-610 kJ/mol is in agreement with other observations.

In a gas atmosphere evaporation rates are as expected smaller.
Data from Matzke (8), obtained in 0.5 bar hydrogen, are included in
the table. Matzke found a heat of evaporation of -440 kJ/mol.
In this case the total process is composed of several steps.

T °C	vapor press. of UO2 [mbar] (10)	evaporation rate [mg/cm²•s]			
		calc.(max) (9) (vacuum)	Felix (7) (vacuum)	Matzke (8) (0.5 bar)	results (1 bar)
2000	0.0076	0.12	0.1	0.0017	0.0025
2500	1.8	25	50	0.2	0.019
2800	18.0	240	780	1.5	0.048

The evaporation rates from this paper are - with the exception
of the lowest temperature- even smaller than the values of Matzke.
Also the found heat of evaporation is much smaller.

Concerning to Aitken (11) the total evaporation rate above
2000 °C is controlled by the transport in the gas phase.
He estimates viscose flow in tight channels, if the partial
pressure difference on both sides is larger than 1 mbar
(for argon gas). For 2400 °C he calculates a evaporation rate of

$2 \cdot 10^{-3}$ [mg/cm$^2 \cdot$s], a value which is even smaller than
the value of this paper. On the other hand he estimates
heats of evaporation between -50 and -100 [kJ/mol], which
are actually energies for the diffusion process.

The experiments reported in this paper are therefore between the
experiments of Matzke and the model of Aitken. For the case
of a HCDA one has to assume, especially at higher temperatures,
evaporation rates of the fuel which cannot be neglected.

Furthermore Felix found for the release of fission products from
fuel at high temperatures, that the release rates were controlled
by the fuel evaporation, and not any more by the diffusion
inside of the fuel.

If in the case of a HCDA the fuel reaches high temperatures,
evaporation of fuel has to be considered.
Important for this process will be the active
surface. In our experiments the large inner surface in the case of
used FISSIUM was 'not active' for the evaporation, while in
the rubble bed of 3 cm depth all the geometrical surface was
'active'. In a real case there is one more driving force :
the temperature gradient in the fuel because of the intrinsic
heat sources. With the model of Aitken and the observations in this
work, the evaporation rate will be not increased.

5.4 Transport of evaporated UO$_2$

As described in the previous chapters evaporating UO$_2$ will
leave the surface of the pellets. A smaller fraction diffuses
into the thoria material, an other fraction condenses in the
crucibles, the remaining part gets plated out at the high
temperature part of the arrangement or is transported to the
filter lines. The fractions of the evaporated UO$_2$ found in
the various parts af the arrangement were very scattered from

experiment to experiment.

5.4.1 Diffusion into thoria

The used thoria curcibles were obtained from the ZIRCOA -company,
ordered as 'fine grains' with high density. The density measured
in our laboratory was 9.38 g/cm^3, with less than 0.1 % open pores.

Diffusion in solids is a very complex process, even if one
considers a nearly ideal systems like the ThO_2 - UO_2
mixture (12). It was not the goal of this work to study this
diffusion, some observations however are worth to be discussed.

The diffusion depends on the (chemical) activity of the two
oxydes, which is proportional to their vapor pressures. Because
the vapor pressure of thoria is much lower than this of UO_2,
diffusion was only observed for UO_2 into the thoria, and not
for thoria into UO_2. (The latter was checked by chemical
analysis of the samples after the experiments.)

After removing the pellets from the crucible, 'foot prints' can
be seen (fig.6). The deepest colored part were at the pellet-
positions. There the UO_2-partial pressure in the space between
the oxydes has the highest value. The density of the thoria
at such position after an experiments was found to 9.59 g/cm^3,
clearly showing the influence of the UO_2. The thoria surface
at such positions was very rough if compared with areas apart of it.

If viewed by dark-field optical microscopy or with polarized light,
cross sections of the crucible at such position show a penetration
profile with a sharp boundary (fig.7).

Actually there is no really boundary, as seen by microprobe
analysis. The seen boundary reflects only a certain concentration
of uranium in thoria. Nevertheless this boundary is used for the

further considerations, because it was found in all experiments.

The boundary moves into the thoria with the square root of the
time as expected for diffusion controlled processes. Fig.8 shows
the results for the temperature of 2450 ±50 °C. No differences
are seen between different types of UO_2-pellets or FISSIUM.

It is remarkable from fig.8, that the penetration rate is higher
in experiments without sodium compared to those with a sodium
vapor atmosphere. Because the vapor pressure of UO_2 is the
same in both atmospheres, surface effects are probably responsible
for this behavior.

This was the only influence of the sodium vapor atmosphere
seen so far in our experiments. If or not such an effect could have
an influence on core-catcher materials is not sure, however it has
to be taken into consideration.

5.4.2 UO_2 crystals and erosion products

Evaporation rates of UO_2 get high at higher temperatures.
In such cases, well shaped crystals are growing at the wall
around the upper edge of the thoria crucible. Fig.9 shows some
SEM-pictures. The crystals were condensed from the gas phase,
sometimes they were up to 0.5 mm in size.

The crystals undergo obviously an erosion process. Edges were
smoothed and finally very courious figures were found (fig.10).

Chemical analysis of the crystals and erosion products showed
more than 95 % UO_2 and less than 1 % ThO_2, and the
concentration on fission products (in the case of FISSIUM pelltes)
was neglectable. From this analysis it may be assumed that pure
UO_2 was condensed.

5.4.3 Deposition at high temperatures

Above a temperature of 1000 °C the sodium remained in the vapor
phase. This part of the arrangement is called 'high temperature
part'. The most important section of it was the tungsten cylinder.
Along this the temperature dropped from the sample temperature
to the temperature of the sodium evaporator. 31 ± 24 % of the
evaporated UO_2 was found in this section, sometimes in a
narrow band, sometimes smeared over the whole cylinder surface.
No effect of the experimental conditions on this distribution
was detected, and the sodium had no influence.

Because no clear 'thermal gradient tube'-effect was found,
and because the UO_2 in this part of the facility is already in
a condensed form, aerosol deposition on the surface is considered
to be responsible for the observations.

5.4.4 Transport into filter lines

On top of the evaporator (fig.2) and before the condenser there
is a small space, where the temperature drops within less than
0.5 seconds to below 500 °C. In case of experiments with sodium,
this is condensed in this area, probably starting at 'seeds'
of other aerosols. 25 ±15 % of the evaporated UO_2 was
always found in this area, independent if or not sodium was
used in the experiment.
The remaining UO_2 is found in the sintered metal filters.

5.5 Thermal reduction

Heating pure UO_2 in an oxygen-free atmosphere will cause
the loss of some oxygen from the oxide (13,14). Such an effect

can be expected because of a relative high oxygen potential
at high temperatures. If oxygen is lost the remaining oxygen-
deficient oxide has a lower oxygen potential. This may have an
influence on the behavior of the nuclides.

In the experiments with FISSIUM this effect cannot be seen
because of the many phases. In pure UO_2 pellets however
the oxygen deficient oxide is formed.

Metallographic specimen of samples, heated for 1 hour to 2500 °C
in an argon or sodium/argon atmosphere, show the occurence of a
metallic phase in form of bright spots with a diameter of 1 to 6 μ
(fig.11). The pictures are comparable to those of Rothwell (13).

The original oxide was stoichiometric $UO_{2.00}$, the
final composition was $UO_{1.95}$ in the case of a LWR-specific
pellet and $UO_{1.96}$ in the case of a FBR-specific pellet.
(These values were obtained by picture analysis). These are the
final composition for a thermodynamic equilibrium , no further
reduction is expected when heating for a longer time (11).

From the literature (13,14) one may expect a final composition
of $UO_{1.97}$ for a temperature of 2500 °C. The found values
are in the same range, the used method for the calculation
however to coarse to allow a more precise evaluation

In any case thermal reduction has to be taken into account when
considering the rubble bed of fuel. Especially the breeding blanket
material, which will be one component of the rubble, will cause
by this way a reduction of the overall oxygen-potential of the
system.

5.6 surface structur

Evaporation of substances from solid bodies starts at sharp
edges and corners, which gets smoothed by this process. In some
cases even grain-boundaries may be startpoints for the evaporation.

Originally the surface of the used pelltes was smooth from the
fabrication, even if the pellets were not grinded or polished.
After an experiment the surfaces were completely restructured,
consisting of small spheres and agglomerates, the spheres with
diameters up to 100 μ . Fig.12 shows some examples. The new
surface was formed at a temperature of 2400 °C within 10 minutes,
at lower temperatures after a longer time. It was formed as well
on FISSIUM and on UO_2 surfaces, independent on the used
atmosphere.

The diameter of the spheres was the same at all temperatures
and independent on the time. Sometimes however, especially
at higher temperatures, columns of the same diameter were
found on the surface.

Obviously the new structure is formed by evaporating oxide.
The size of the spheres is larger than the grain size
of the original material, which was in the case of FISSIUM
90% below 30 μ . Therefore it is assumed that the material
is a condensate, especially if one considers the fact, that
these spheres and agglomerates could easily be stripped away.
Like the crystals described in chapter 5.4.2 the spheres
consist of UO_2.

The new formed surface is obviously much larger than the
surface of the original pelltes. However it was not an active
surface in the sense of chapter 5.3, and does not increase the
evaporation rate. There were no signs for any influence of the
new surface on the behavior of other nuclides.

6. Summary and discussion

An experimental arrangement was developed to study the behavior
of UO_2 and FISSIUM in a sodium vapor atmosphere at temperatures
just below the melting point of the UO_2. Experiments were
performed up to 2800 °C and up to 2 hours holding time. A maximum
of 70 grams of sample materials was used in a 3 cm high stack.

Density and open pore volume did not change during an experiment.
There were probably compensating effects of densification
and creation of new pores because of evaporation of fission
products and UO_2.

The evaporation rate of UO_2 was high at higher temperatures
and reached 0.1 [mg/cm²•s] at 2800 °C. Only an 'active'
geometrical surface participated in this process, no evaporation
occured from inner pores (inspite of 28 % open pores in the case
of FISSIUM).However in a 'rubble bed' of smaller pieces of broken
pellets, all the geometrical surface was active.

The obtained evaporation rates were between values measured by
Matzke (8) and evaluated by Aitken (11). Also the found heat
for the total evaporation process of -212 [kJ/mol] was smaller
than values of Matzke, but larger then these of Aitken. That means
that in a small rubble bed the evaporation rate is not only
controlled by the diffusion in the gas phase, but also by the
evaporation itself.

For a rubble bed of a HCDA-reactor evaporation rates as described
in this paper have to be expected. Near the melting point of the
UO_2 the rate is 0.07 [mg/cm²•s].

Evaporation of UO_2 creates a new pellet surface, consisting

of small spheres and agglomerates of pure UO_2. However
no new 'active' surface is formed.

Evaporated UO_2 is transported away from the oxide surface.
A small fraction diffuses into the thoria of the crucible.
Inspite of no or very small temperature gradients one other
small fraction condenses inside of the crucible. In case of high
temperatures well shaped UO_2 crystals, as well as curiously
shaped erosion products of such crystals were found. No such
particles were found inside of a bed of UO_2 pieces.

One third of the evaporated UO_2 was plated out on surfaces
with temperatures higher than 1000 °C. In this part of the
experimental arrangement the sodium is still in the vapor phase,
aerosol deposition may be assumed.

In the area after this high temperature part the temperature
drops to below 500 °C within less than 0.5 seconds. Here
around 25 % of the evaporated UO_2 was always found. Finally
sintered stainless steel filters, SUPRAMESH of the Pall-company,
removed more than 99.99 % of the oxide aerosol from the sweep-gas
stream. In the case of experiments with a sodium vapor atmosphere,
the filters were heated to about 350 °C to avoid clogging. The
efficiency has been the same.

For the case of a HCDA-reactor therefore one can expect, that
all of the evaporated fuel will be condensed within the sodium
pool. Release to the environment can only occure together with
the release of sodium or during a sodium fire.

The oxygen-potential of the fuel may influence the release of
radionuclides and fuel. It was found, that under the conditions
of an LMFBR - accident core a thermal reduction with the effect
of reducing this potential occures. The final composition
was found for 2500 °C to be $UO_{1.95}$.

All results described so far in this summary were obtained
independent if or not the experiments were performed in a pure
argon or an argon/sodium-vapor atmosphere. An influence of the
sodium was only seen for the diffusion of UO_2 into the
thoria of the crucible. In the case of sodium vapor, the
diffusion rate was reduced. This effect will have to be considered
when studying core catcher questions.

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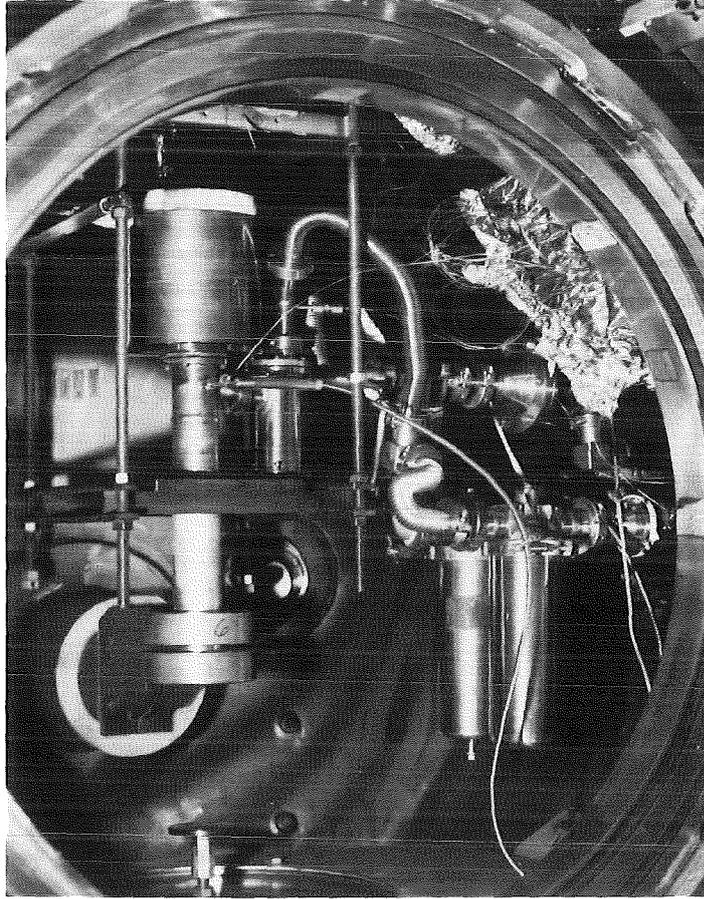
Fig.1: Experimental arrangement in the
SUSI-Vessel
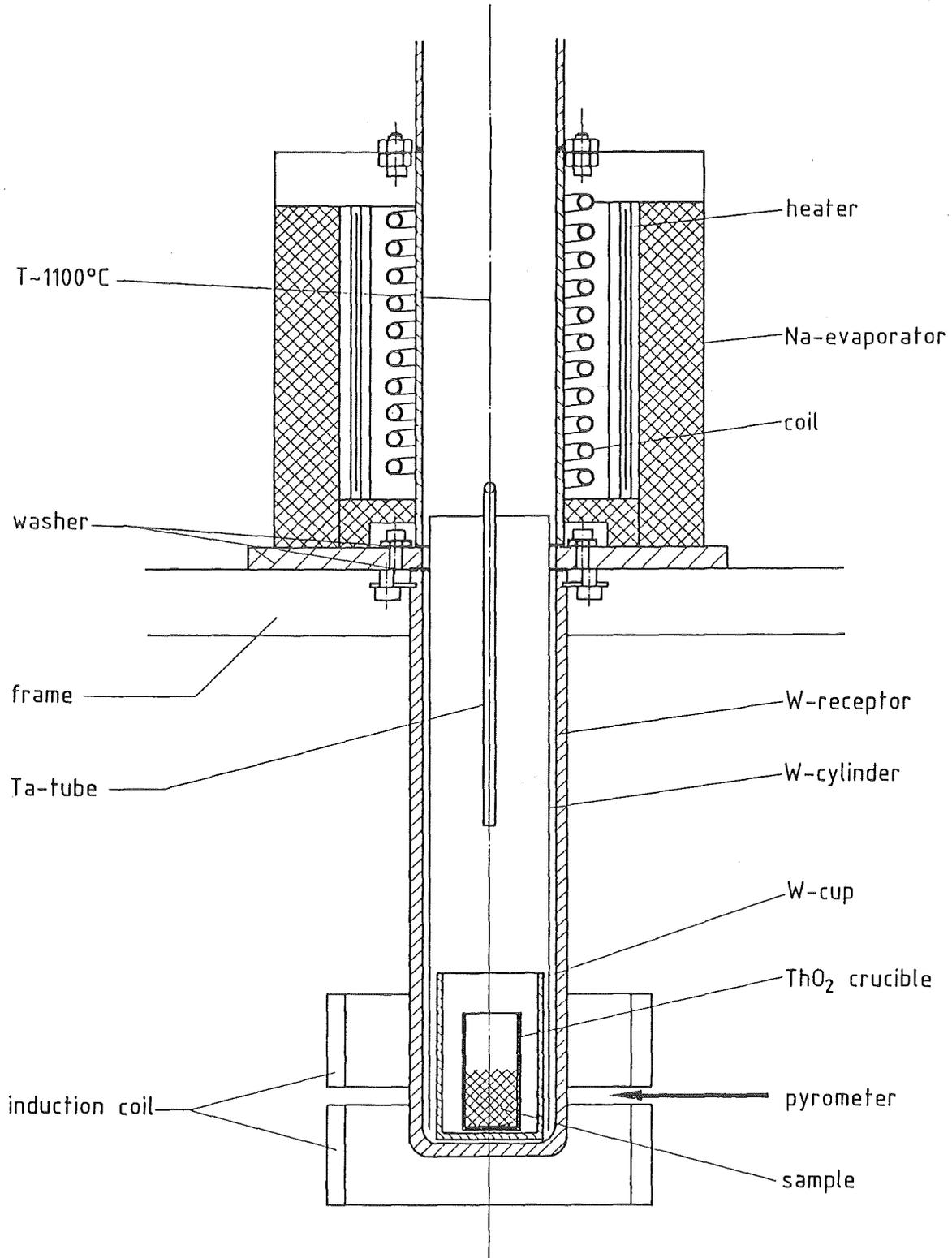
Fig.2: High temperature part of the experimental arrangement
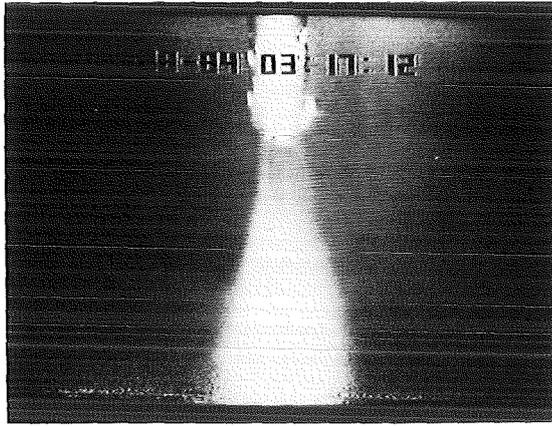
Fig.3: Beam of sodium vapor leaving the tantalum tube
(about 2x)
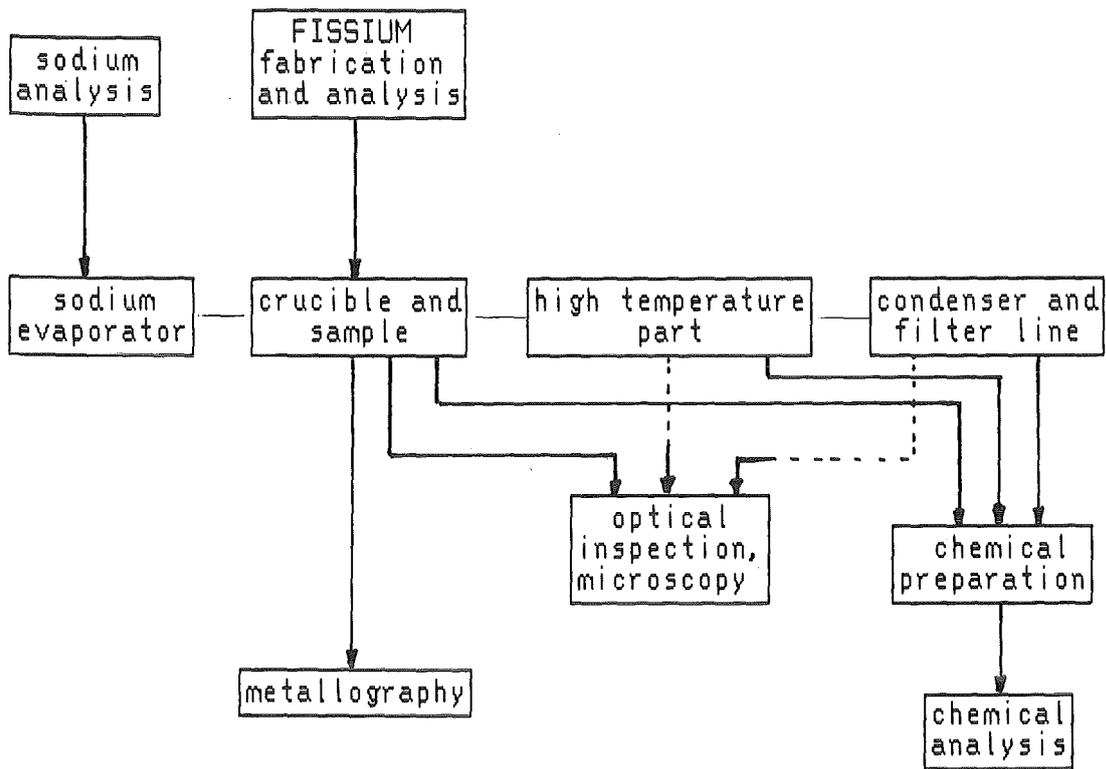
Fig.4: Schematic diagram of the investigations prior and after
the heating
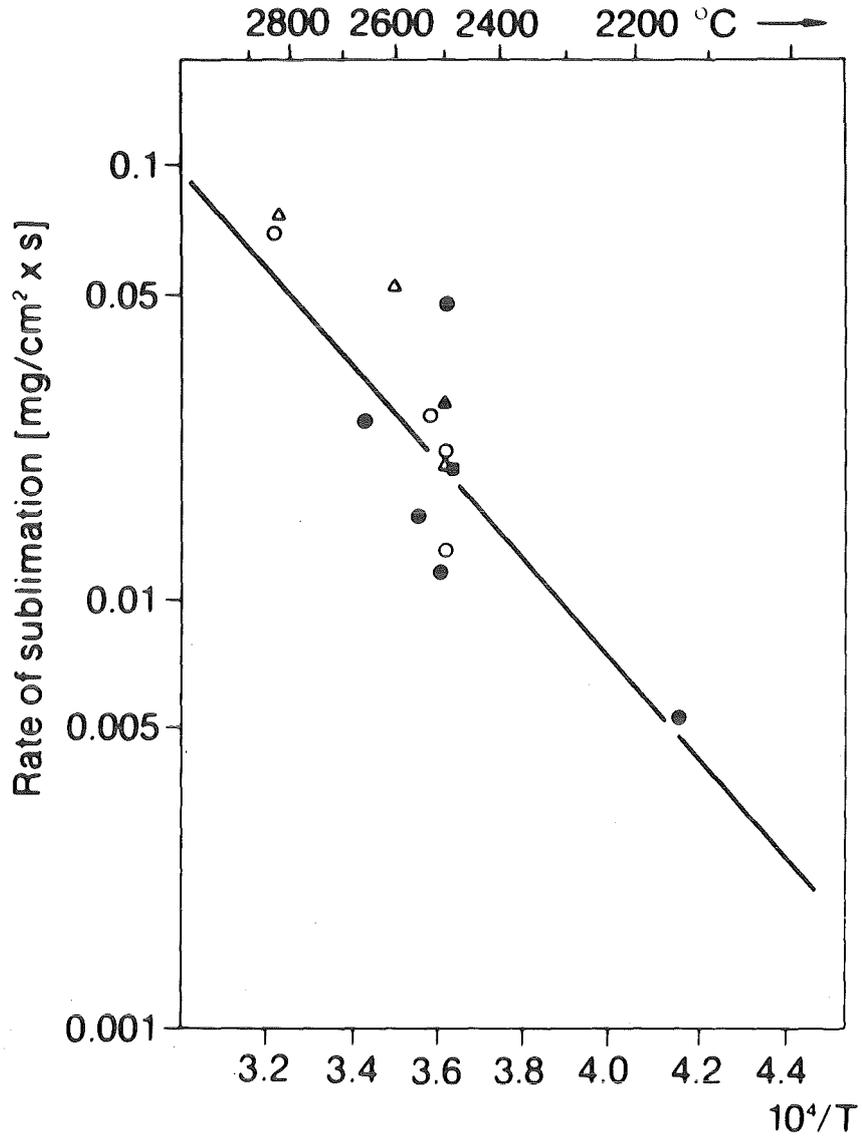
Fig.5: Evaporation rate of UO_2.
Closed figures = experiments with sodium vapor.
Open figures = experiments with argon only.
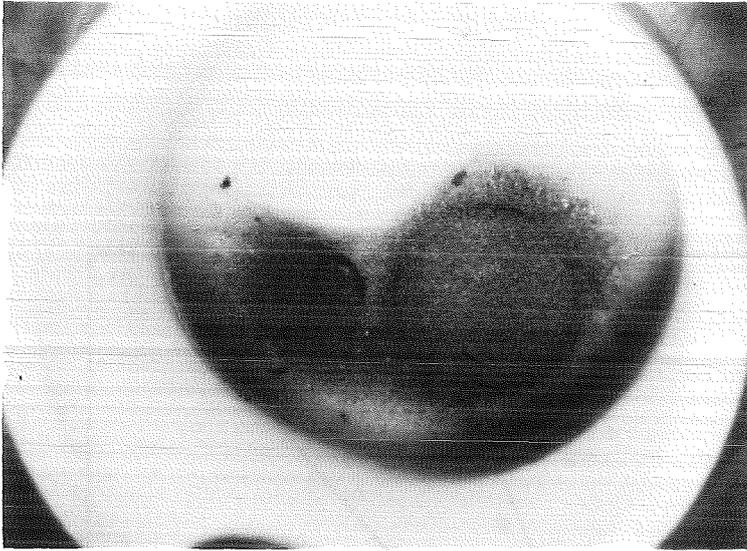
Fig.6: 'Foot prints' of UO$_2$-pellets in the thoria
crucible (about 2x)
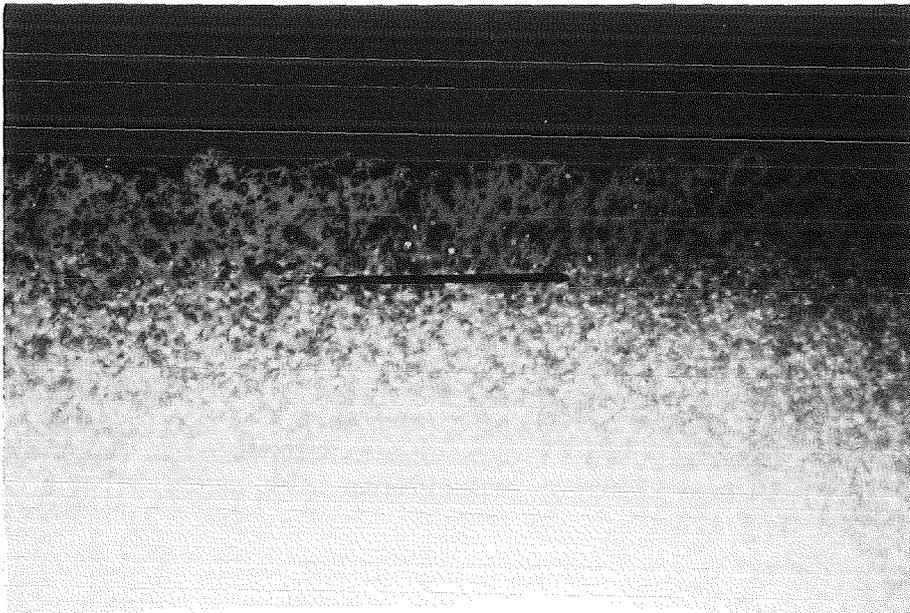
Fig.7: Diffusion zone of UO$_2$ into thoria; 1 hour at 2500°C,
sodium vapor atmosphere.
(100 x)
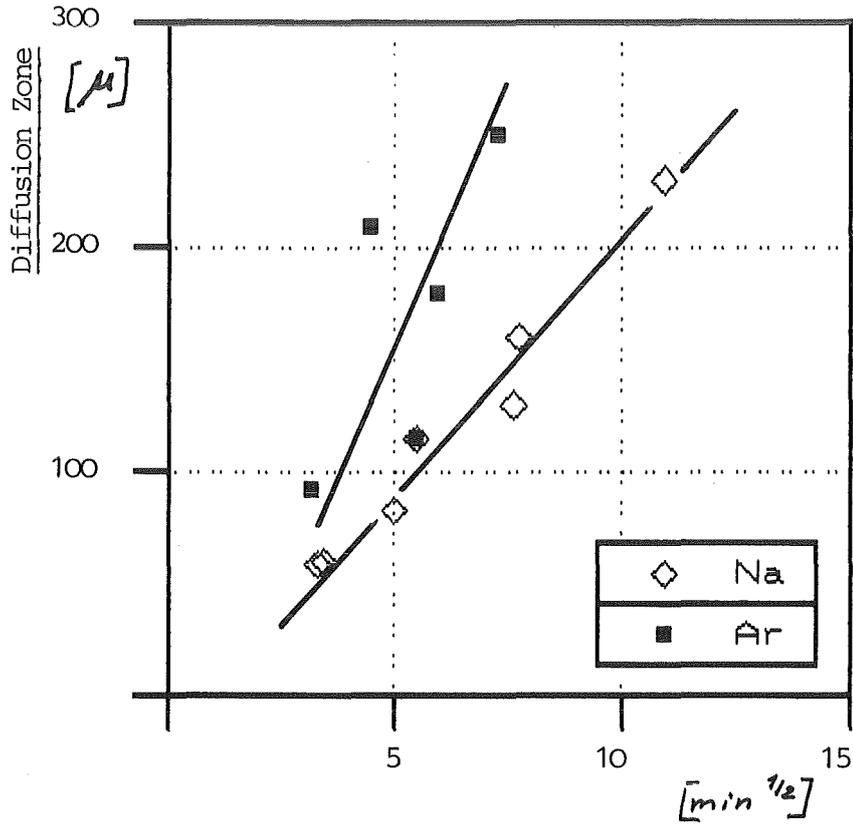
Fig.8: Diffusion of UO_2 into thoria.
Open figures : experiments with sodium vapor.
Closed figures: experiments with argon only.
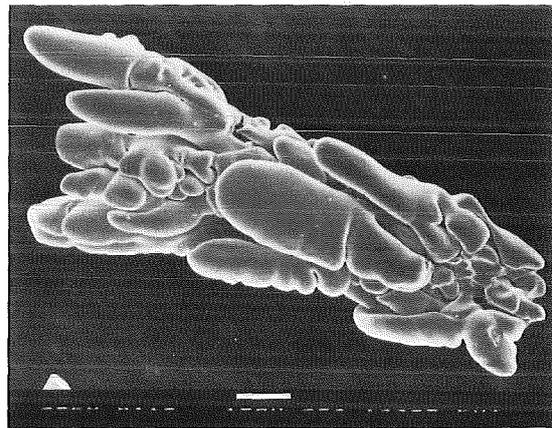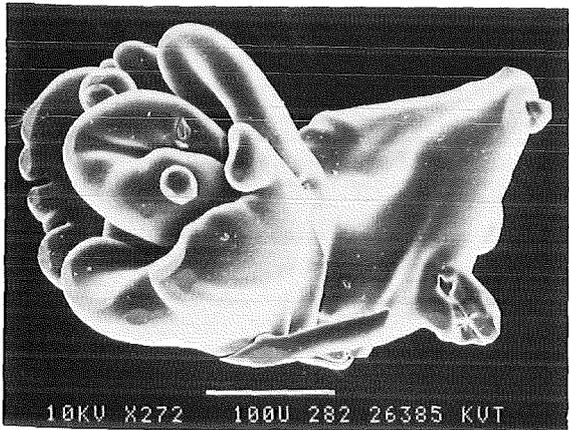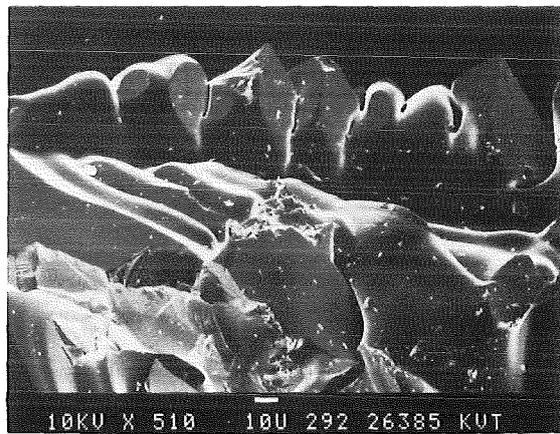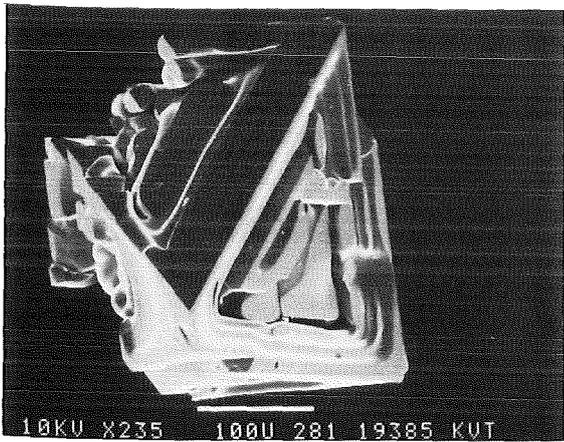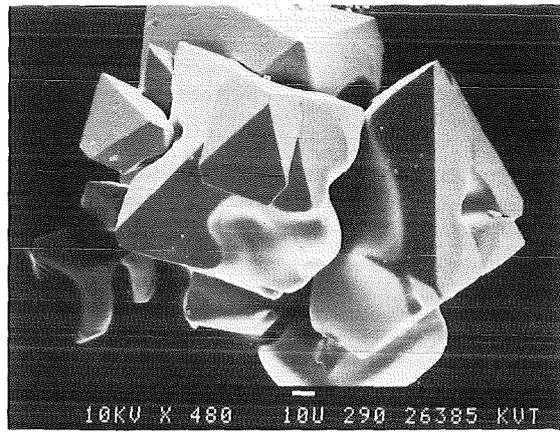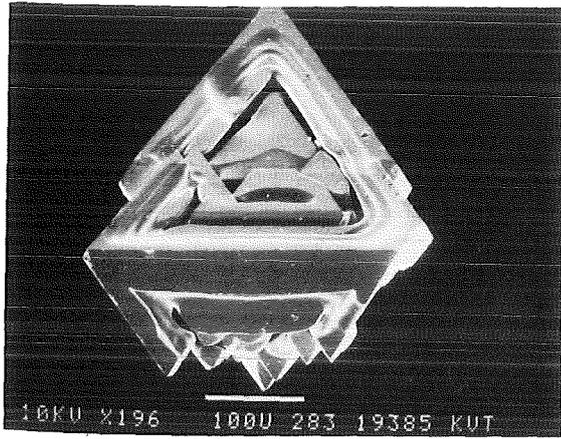
Fig.9 and 10: Crystals of UO_2 , condensed inside of the thoria crucible. Lower two figures erosion products.
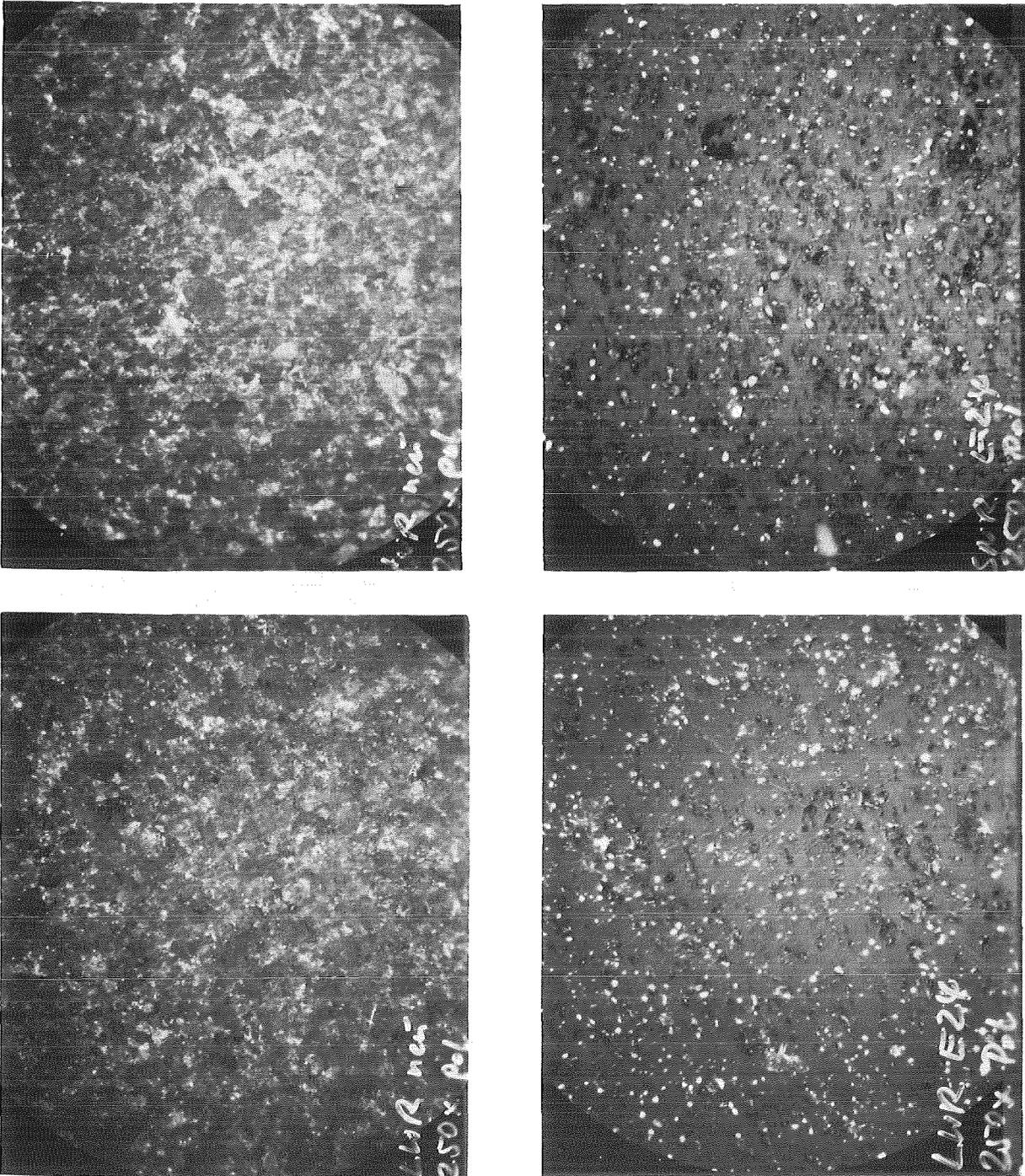
Fig.11: Thermal reduction of UO_2.
Upper two figures FBR-specific pellets, lower two figures
LWR-specific pellets. Left two figures before, right after
heat treatment for 60 minutes to $2500^{\circ}C$.
(Polarized light, 250x), white inclusions are metal rich
phases.
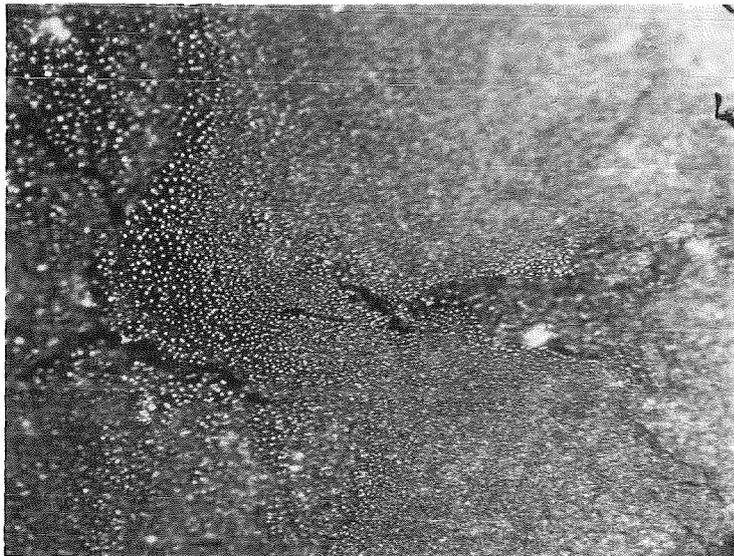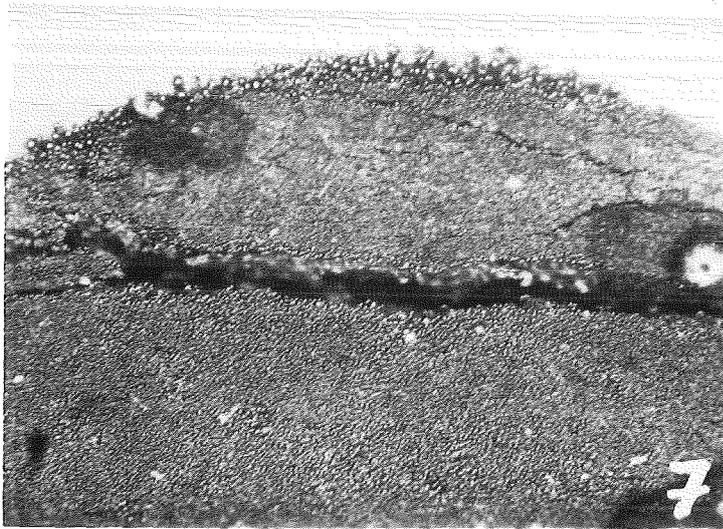
Fig.12: Surface structure of UO$_2$ pellets after exposure
to sodium or argon for 15 minutes   2400°C.