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Fuels and targets for incineration and transmutation of actinides: the ITU programme

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ABSTRACT

The ITU programme for the development of fuels and targets for transmutation of actinides is presented. The fabrication of various types of oxide fuels/targets by dust-free processes is described. Selected results of post-irradiation examinations of irradiation experiments (SUPERFACT, TRABANT-1, EFTTRA-T4) are presented to demonstrate the irradiation behaviour of these fuels/targets. Finally, the future developments at ITU in this field are described, including the new shielded facility (the MA lab) for fabrication of minor actinide fuels.

INTRODUCTION

The development of fuels and targets for transmutation of actinides is one of the most important issues in the P&T proposals for nuclear waste management. It is a time consuming process, which, depending on the level of innovation, can amount to tens of years between initial laboratory studies and industrial qualification.

Different requirements for the fuel and target design can be defined, depending on the fuel cycle strategy. For example, with multiple recycling and use of fast reactors, the high fissile content and the possibility to reprocess the fuel are of key importance. As the material is reprocessed, the fuel or target could be based on a uranium or thorium matrix. On the contrary, in a once-through transmutation scenario, the stability of the material at high burn-up and as spent material for geological disposal is crucial. As the extent of transmutation must be as high as possible, uranium-free targets are preferred.

As a result of these different options, various fuel/target forms are currently under consideration, metals, oxides, and nitrides. For oxide fuels, the following fuel forms can be distinguished: minor actinides in mixed oxide (MINOX), mixed transuranium oxide (MIMOX), inert matrix mixed oxide (IMMOX), and composite fuels such as ceramic-ceramic (CERCER) and ceramic-metal (CERMET). Only the first of these should contain uranium.

At the Institute for Transuranium Elements (ITU), an active programme on the fabrication and post-irradiation examination of fuels and targets for transmutation of actinides has been executed for more than 15 years. The research activities have been part of several international collaborations in which the ITU contribution played a central role. As part of these collaborations ITU participated in the following irradiation experiments:

- The SUPERFACT experiment was initiated in the mid-eighties in collaboration with the Commissariat à l'Energie Atomique (CEA). In this experiment the addition of minor actinides

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to mixed oxide fuel for fast reactors was explored (MINOX fuel type). Eight pins were fabricated and irradiated in Phénix.

- In the TRABANT-1 experiment MOX and inert-matrix fuel with high Pu-content were investigated in collaboration with Forschungzentrum Karlsruhe (FZK) and CEA.
- In the EFTTRA-T4 experiment an inert-matrix target concept for americium transmutation was fabricated (CERCER), and examined following irradiation. This programme was performed in collaboration with the EFTTRA partners (CEA, EDF, FZK, NRG and JRC-IAM).

In the present paper, ITU activities on incineration and transmutation of actinides are described and the main achievements are discussed. The implications of minor actinides on fuel fabrication are indicated, and new directions of the research outlined.

FABRICATION TECHNOLOGY

Fabrication of targets for transmutation and incineration of actinides and fission products requires more stringent radiation protection measures than currently necessary for the manufacture of conventional UO_2 and MOX fuels. Whereas MOX can be fabricated in glove boxes, the handling of minor actinides requires extra shielding in the form of lead (for gamma radiation) and water (for neutron radiation), and remote operation. As a result, process simplification and automation are necessary. In addition, the waste generated should be minimised. The fabrication procedure should not generate dust, which could collect on the surfaces of the glove boxes and the equipment therein, so that operator intervention is facilitated and radiation exposure to the personnel minimised.

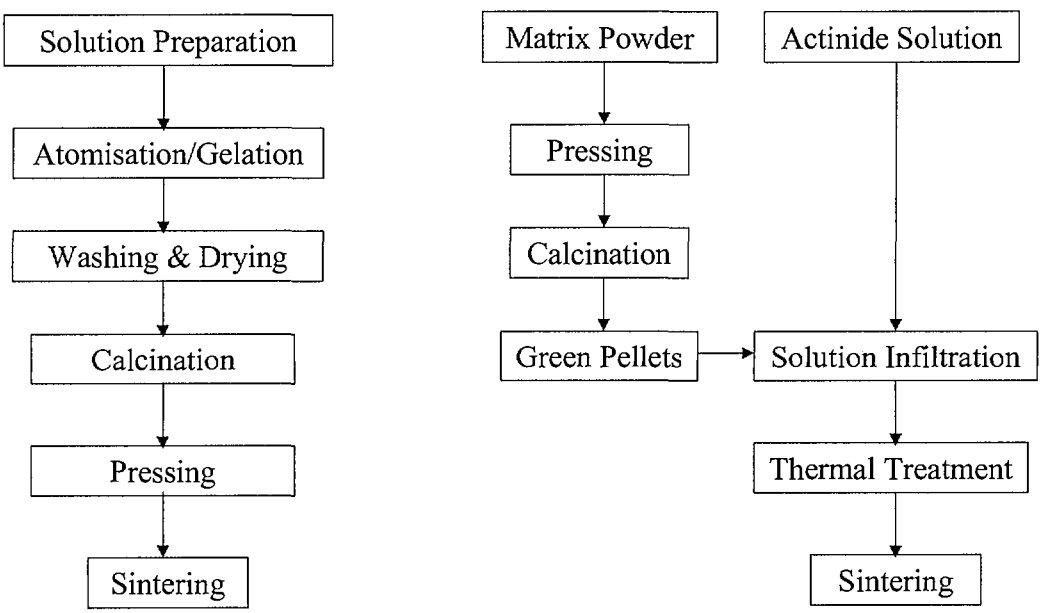

Figure 1. Flow sheets of the fabrication processes: SOL-GEL (left), Infiltration of Radioactive Materials (INRAM) for compacts (right).

For these reasons, dust-free fabrication has become a major development topic at the ITU. The techniques developed are based on liquid processing, in particular, the fabrication of free-flowing powders by SOL-GEL and the infiltration of actinide solutions into compacts or beads (INRAM) (see Figure 1) [1-5].

The principal device at the heart of the SOL-GEL droplet to particle conversion process is the rotating cup atomiser. Droplets generated by passing a feed solution over the edge of a cylindrical cone rotating at high speed (see Figure 2) are collected in an ammonia bath where gelation occurs. Following washing, drying and calcination steps, spherical particles with a polydisperse size distribution (20 -150 μm) are obtained. These spheres are free-flowing and are pressed directly into compacts and sintered to obtain product pellets. With this method a homogeneous distribution of the elements in the final material is obtained. A disadvantage of this method is the relatively large amount of liquid waste that is produced, but this could be recuperated and recycled in an industrial plant.

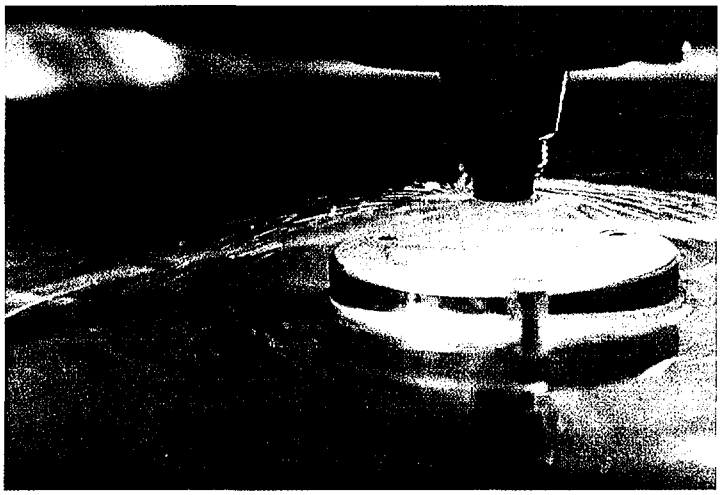

Figure 2. Droplets produced by a Rotating Cup Atomiser

In the SUPERFACT experiment, the SOL-GEL process has been used to fabricate mixed-oxide fuels with varying concentrations of minor actinides: $(U_{0.55}Np_{0.45})O_2$, $(U_{0.60}Np_{0.20}Am_{0.20})O_2$, $(U_{0.74}Pu_{0.24}Np_{0.02})O_2$ and $(U_{0.74}Pu_{0.24}Am_{0.02})O_2$. The density of all materials was greater than 95% TD. For the TRABANT experiment this method has also been used to produce mixed oxide fuels with high Pu content: $(U_{0.55}Pu_{0.45})O_2$ and $(U_{0.55}Pu_{0.40}Np_{0.05})O_2$. In addition, a uranium-free pin, using cerium oxide as a matrix, was prepared by the SOL-GEL method.

Table 1: Fuels and targets irradiated in the frame of the ITU programme on transmutation of actinides.

Programme	Reactor	Fuel	Fabrication method	Type
SUPERFACT	Phénix	$(U_{0.55}Np_{0.45})O_2$	SOL-GEL	MINOX
		$(U_{0.60}Np_{0.20}Am_{0.20})O_2$	SOL-GEL	MINOX
		$(U_{0.74}Pu_{0.24}Np_{0.02})O_2$	SOL-GEL	MINOX
		$(U_{0.74}Pu_{0.24}Am_{0.02})O_2$	SOL-GEL	MINOX
TRABANT-1	HFR	$(U_{0.55}Pu_{0.40}Np_{0.05})O_2$	SOL-GEL	MINOX
		$(Ce_{0.53}Pu_{0.47})O_{2-x}$ [two O/M]	SOL-GEL	IMMOX
EFTTRA-T4	HFR	$MgAl_2O_4$-12 wt%Am	INRAM	CERCER

For the EFTTRA-T4 experiment, a target containing about 12 wt% americium in a $MgAl_2O_4$ matrix was fabricated using an infiltration method (INRAM). In the variation of the infiltration process [3] (see Figure 1), porous pellets (P ≈ 50 %) were immersed in a concentrated americium nitrate solution. Following infiltration, they were dried, calcined to convert the americium nitrate to the oxide form and then sintered at elevated temperatures to give the final pellets. The process has the advantage that the steps involving the handling of radiotoxic materials are minimised, and as no precipitation or washing steps are required, the radioactive wastes produced are negligible. This

method yielded a homogeneous distribution (microdispersion) of the americium in the trial pellets. It was observed, however, that the distribution was not uniform in the final pellets (see Figure 3). This inhomogeneity could have been caused by a non-uniform porosity of the uninfiltrated pellets, or to a diffusion process during the thermal treatment in the steps involving liquid to solid Am nitrate, and / or Am nitrate to oxide conversion.

Further development of this process showed that homogeneity of the actinide distribution can be guaranteed when a porous matrix powder (beads generated by the SOL-GEL method) is infiltrated instead of the pellet [4]. Due to the higher porosity of the beads ($P \approx 70$ %), higher actinide contents can be achieved. Fabrication of $MgAl_2O_4$ with 20 wt% Am, and $(Zr,Y,An)O_2$ targets, with metal concentrations up to 33% (corresponding to $(Zr_{0.68}An_{0.20}Y_{0.12})O_2$) have been obtained [5].

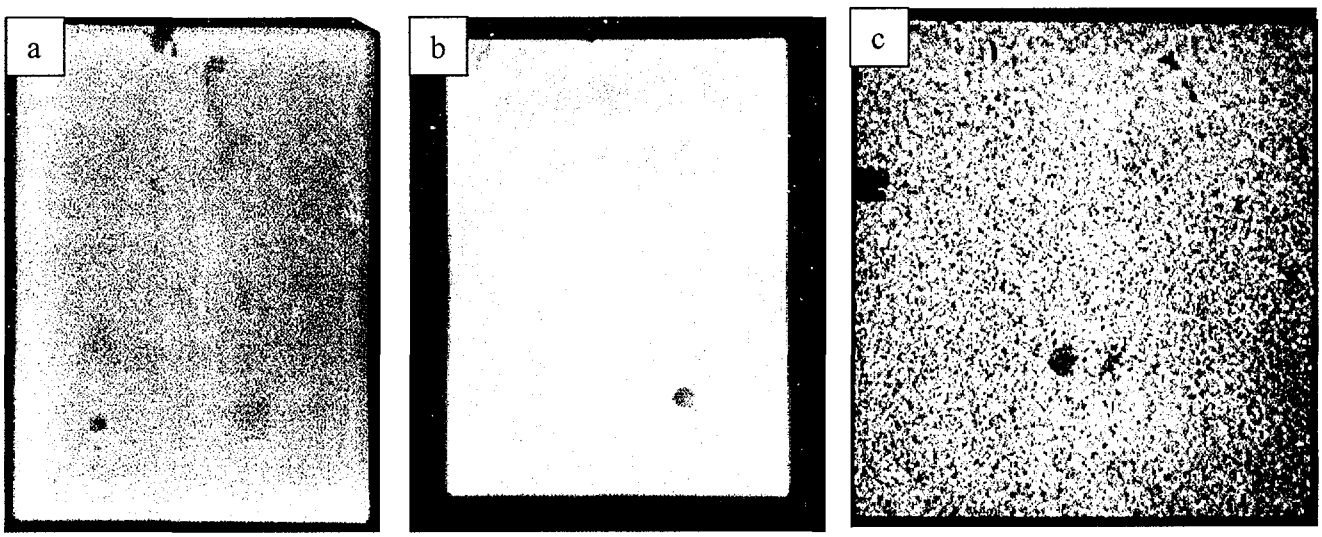

Figure 3. α-autoradiographs of $MgAl_2O_4$ pellets containing Am prepared by INRAM: (a) the EFTTRA-T4 pretest (with 7.7 wt% Am) produced by infiltation of pellets (b) EFTTRA-T4 (12 wt% Am) also prepared by infiltration of pellets, (c) with 20 wt% Am prepared by INRAM of beads.

POST-IRRADIATION EXAMINATIONS

Good in-pile performance of fuels fabricated by the sol-gel technique has been demonstrated in the SUPERFACT experiment. The non-destructive examinations of the 4 pins (see Table 1) did not show any anomaly in their behaviour. In particular, no accelerated corrosion was observed. The high concentration of americium in the $(U_{0.60}Np_{0.20}Am_{0.20})O_2$ fuel led to an important increase of the fuel column length and to a more significant diametrical deformation of the cladding. This is probably due to the beginning of a mechanical interaction between the oxide fuel and the cladding. The fission gas release rates (60-80% of the theoretical yield) were in good agreement with those of standard fuels, even for the fuels with a high concentration of minor actinides. The interpretation of the physico-chemical and ceramograpic examinations of the fuels led to the following conclusions:

- A beginning of a pellet-cladding mechanical interaction was observed for the $(U_{0.60}Np_{0.20}Am_{0.20})O_2$ pins.
- Cesium was found at the end plugs. Its accumulation is probably due to the particularity of the pins with high Am and Np contents (short fuel column).
- The cesium-profiles for the neptunium-containing fuels showed anomalous behaviour of this fission product compared to the other pins.
- The corrosion depth was as expected.
- The fuel temperature was probably higher for the americium-containing fuel.

- The fission gas production and release were as predicted for the operating power.
- The Am pins had a higher helium production, mainly due to the daughter products with high specific alpha activity (e.g. ^{242}Cm and ^{238}Pu). The released helium contributed to an increase of the internal pressure of the pin. In addition, higher porosities and swelling, probably due to the helium still confined in the fuel, were found.

The analysis of samples of fresh and irradiated SUPERFACT fuels performed by ITU are in excellent agreement with the observations made at CEA [6]. The neptunium measurement is difficult (not available at CEA) and has a higher uncertainty than the measurement of the usual nuclides. The measured extent of transmutation (CEA and ITU) of americium at the maximal flux level is 31.5% for the fuels with high minor actinide content and 28% for the fuels with low minor actinide content. From neptunium analyses an average extent of transmutation of 30% was determined for the three samples. The comparison of the measured values with calculations is satisfactory in the case of americium. In the case of neptunium, comparison with calculations showed less agreement for the fuels with low minor actinide content.

Also the SOL-GEL $(U_{0.55}Pu_{0.40}Np_{0.05})O_2$ fuel in the TRABANT-1 experiment, which reached a burn-up of 9.3%, showed a good in-pile performance as derived from the non-destructive analysis [7]. In contrast, the behaviour of the ceria-based fuels was less promising. The $(Ce_{0.53}Pu_{0.47})O_{2-x}$ fuel with a low O/M ratio melted, indicating a poor thermal conductivity for this fuel material. The $(Ce_{0.53}Pu_{0.47})O_{2-x}$ fuel with higher O/M ratio fuel behaved better [7]. Further post-irradiation examinations of the TRABANT-1 fuels are still in progress and will reveal more details about the in-pile behaviour of these pins.

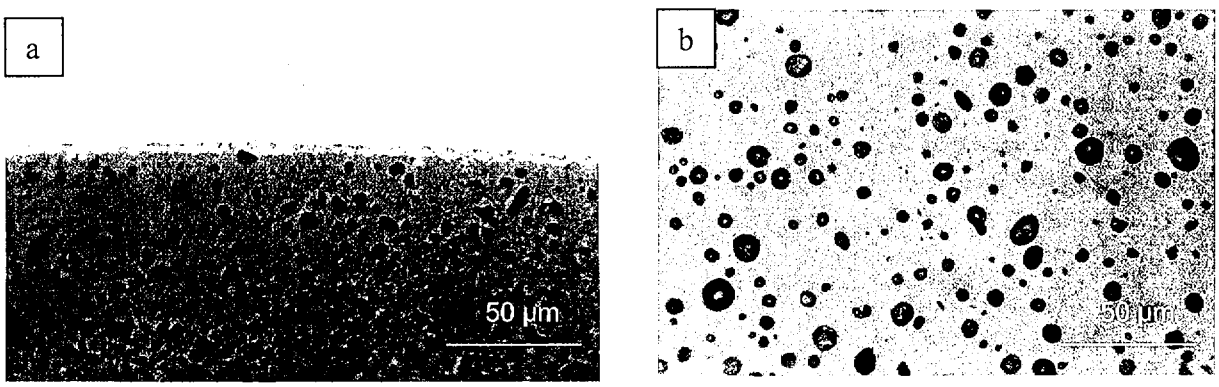

Figure 4. Ceramographs of the EFTTRA-T4 sample showing (a) the porosity at the rim of the pellet (r/r_0=1) and (b) at the centre of the pellet (r/r_0=0).

Only one irradiation test of fuels/targets prepared with the INRAM fabrication method has been performed up to now: the EFTTRA-T4 experiment. The target was irradiated in HFR Petten during 358 full power days, during which 96% of the initial ^{241}Am was transmuted, and 28% of the initial americium atoms fissioned. The irradiation performance of the target was not optimal, however, swelling (up to 18% in volume) was observed during the non-destructive analysis performed by NRG [8]. The cause of this swelling was revealed by the destructive analysis at ITU, where ceramographs indicated a porosity increase from 3% in the unirradiated to about 18% in the irradiated pellet. This can be attributed to the accumulation of helium produced by alpha decay of ^{242}Cm (a product in the transmutation chain of ^{241}Am) in gas bubbles (see Figure 4).

Table 2. Characteristics of the EFTTRA-T4 and SUPERFACT irradiation experiments

		EFTTRA-T4	SUPERFACT-1/pin 6
Reactor		HFR Petten	Phénix
Irradiation Time (Full Power Days)		360	358.4
Composition		$MgAl_2O_4$+AmO_x	$(U_{0.6}Np_{0.2}Am_{0.2})O_2$
Am-content (g cm^{-3})		0.4	1.66
Extent of transmutation		94.3	31.5
Extent of fission		27.9	n.a.
Central temperature (°C)		<1050	2200
Axial expansion of the column (%)		4.5	2.3
Radial expansion of the pellets (%)		6.7	3.3
He-production (mol cm^{-3})	End of irradiation	$8.49\ 10^{-4}$	$7.12\ 10^{-4}$
	End of cooling	$1.07\ 10^{-3}$	$1.36\ 10^{-3}$
He release (%)		20	100 [a]
Fission gas release (%)		5	62

[a] not measured but assumed on basis of gas-puncturing results at end-of-cooling

The SUPERFACT (pin-6) and EFTTRA-T4 experiments have shown that for americium-containing fuels/targets the accumulation of helium is an intrinsic problem. In spite of differences in Am-content, neutron fluence and the concomitant difference in extent of Am transmutation (Table 2), the helium production per unit of volume was about equal in the two experiments. In contrast, the swelling was much higher in the EFTTRA-T4 experiment: the axial and the radial expansion were about 2 times higher than in SUPERFACT (pin-6). Apart from the different fuel concepts (composite vs. homogeneous) and the properties of the matrix ($MgAl_2O_4$ vs. UO_2), this can be understood by consideration of the fuel temperature. In the T4 experiment the temperature of the target was rather low and, as a consequence, the He mobility also. The majority of the helium is therefore still present in the pellets, mainly accumulated in gas bubbles or else dissolved as atoms in the lattice. In the SUPERFACT-1 experiment the temperature was considerably higher. As a result, the helium atoms produced migrate through the pellet and accumulate in gas bubbles, which eventually form gas tunnels at grain boundaries. During and after the irradiation, these gas tunnels act as paths for He release, limiting its accumulation in the pellets and, hence, the swelling.

FUTURE DEVELOPMENTS

The research programme at the ITU is currently focussed on tailoring the fuel/target design of uranium-free fuels/targets for transmutation of larger quantities of minor actinides. A point of attention is the problem of He production and how to deal with it: release or retention. Release can be achieved by deliberately using low density (<85% TD) pellets to provide open porosity, but this would lower the thermal conductivity of the fuel. Retention of He could be achieved if low operating temperatures are maintained (e.g. in a CERMET, or in a CERCER composite) and the fuel is produced with gaps between the inclusion and the matrix to accommodate gas accumulation and swelling during irradiation, for example the so-called "jingle concept" developed by the CEA [9,10]. This option, however, must consider the response of the fuel under transient conditions (i.e. "burst release").

At present an extensive programme is being pursued on the fabrication of $(Zr,Y,An)O_2$ IMMOX fuels by infiltration of the actinide solution into porous yttria-stabilised zirconia (YSZ) beads, produced by the SOL-GEL technique. The feasibility of obtaining good pellets of sufficient density has been demonstrated in the cold laboratory using Ce as an actinide stand-in [5]. A major issue of this fuel type is its poor thermal conductivity, which might limit its application. For this reason, the

composite fuels are not rejected, although the highly localised fission density and gas build up may cause local stresses and irradiation instability; rather they are being pursued as a second priority. The fabrication of the MgO-(Zr,Y,An)O_2 CERCER composite is being investigated as is the fabrication of the Mo-(Zr,Y,An)O_2 CERMET composite. Both fuels are fabricated by blending (Zr,Y,An)O_2 spheres with the matrix powders. The results obtained so far indicate significant difficulties in obtaining good quality pellets, especially when the size of the inclusions is greater than 50 μm. This is due to local stresses during sintering as a result of the different material properties of matrix and inclusion.

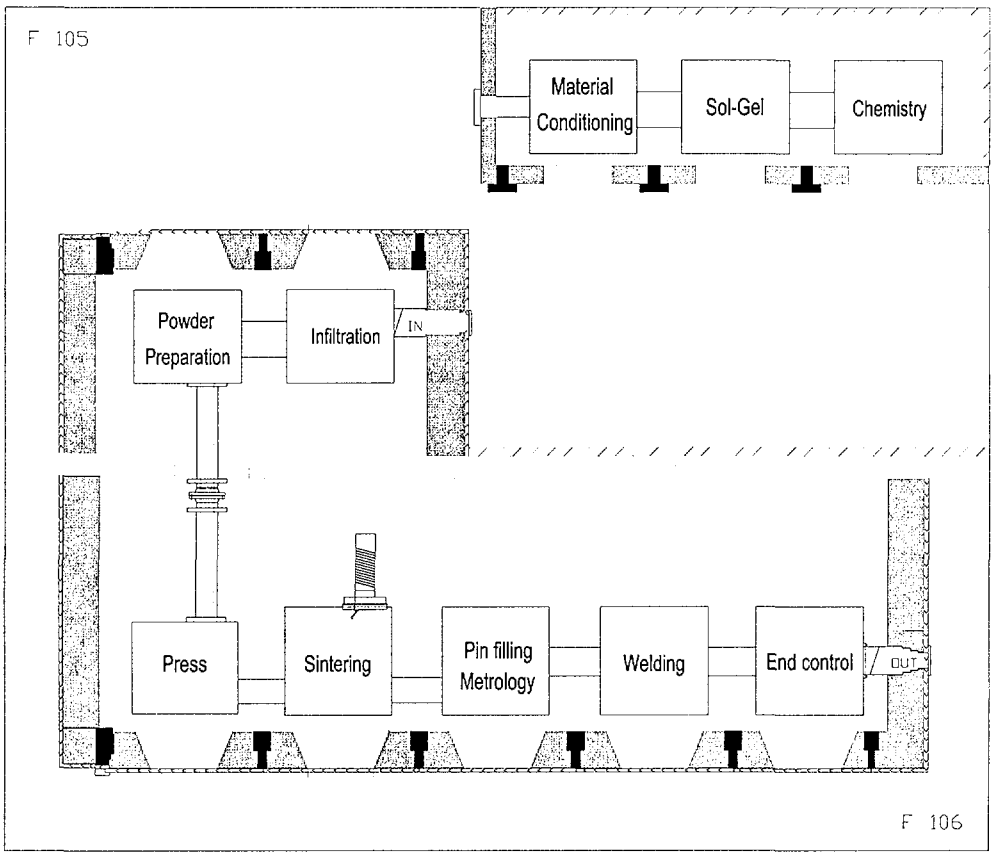

Figure 5. Schematic drawing of the Minor Actinide laboratory at ITU.

The next step in this work is to adapt the processes and validate them in a new shielded laboratory that is being constructed for handling minor actinides in relatively large quantities. In this so-called Minor Actinide laboratory (MA-Lab), two glove-box chains are being build to fabricate fuels and targets containing minor actinides such as americium and curium (see Figure 5). The main chain consists of 7 glove boxes that are shielded by a steel wall containing 50 cm water and 5 cm lead, for which the limiting masses are 150 gm of ^{241}Am or 5 gm of ^{244}Cm. Here the fabrication process is restricted to infiltration, pellet pressing, sintering, metrology, visual inspection, pin filling (maximum length 1000 mm) and pin welding. Operation is achieved not only by telemanipulators, but also by extensive use of robots and remote control. In the case of intervention or repair, the highly active material can be isolated, and conventional glove box practices employed.

In addition to the seven glove boxes that form the core of the MA-lab, a separate chain of three glove boxes for (chemical) preparation of powders containing americium or dirty plutonium by the SOL-GEL technique will be installed. The shielding of these glove boxes, which are also operated with manipulators, is much less: 5 mm lead and 20 cm polyethylene which yield a limiting mass of

50 gm of ^{241}Am. The powder produced in this chain can be transferred to the main chain where it can be processed further. The main chain of the MA-lab is expected to become operational in 2001, the second chain in 2002.

It should be realised that in addition to the traditional pellet fuel forms discussed above, other fuel concepts will be considered. Especially particle fuels are highly interesting as the dust-free fabrication methods like INRAM and SOL-GEL being developed at ITU are admirably suitable for producing such fuels. Particle fuels in sphere-pac pins provide lower smeared densities for helium release. In contrast, gas (helium) retention could be achieved by coating the spheres, which could be consolidated in sphere-pac pins, or embedded in another type of matrix. Such fuel forms are a logical extension of the current research at ITU and will be investigated in the future.

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