INTERNATIONAL CONFERENCE
ON NUCLEAR POWER AND ITS FUEL CYCLE
SALZBURG, AUSTRIA • 2–13 MAY 1977

IAEA-CN-36/477

PLUTONIUM USE - PRESENT STATUS AND PERSPECTIVES

J. van Dievoet, E. Fossoul, E. Jonckheere and E. Vanden Bemden
Belgonucleaire (Brussels)

1. INTRODUCTION

The nuclear power growth in the world (not included USSR, China and Eastern Europe) could result in a nuclear generating capacity of 1700 GW by 2000 and 3500 GW by 2010.

From now up to 2010, the share of the LWR's in the nuclear power will remain very important (probably 80 to 90%) and the fissile plutonium produced, if not used for recycling or for FBR's, will build up a plutonium inventory of more than 5000 T at the end of the period.

The maximum penetration of the FBR's corresponding to that plutonium production could result in a FBR generating capacity of 860 GW by 2010, i.e. 25% of the nuclear capacity at that time. The expected FBR penetration on an industrial basis, if similar to the previous evolution of the LWR capacity is much less than that maximum and a considerable surplus of plutonium is available for recycling in the LWR's. In that case, about 15% of the LWR's energy would be produced by the plutonium.

In the meantime, the very problem is that of reprocessing. If positive decisions are taken, not only the FBR's growth as a commercial reactor type will be possible but also the best solution will be obtained in the intermediate period.

2. PLUTONIUM FUEL PERFORMANCE

2.1. Pu use in thermal reactors

2.1.1. Short review of plutonium fuel irradiation

Numerous irradiations of fuel bundles containing mixed oxide fuel rods have been performed in LWR's, both pressurized and boiling. The general trend has been towards the island concept for cruciform control rods reactors, and towards the all mixed oxide concept for rod cluster control reactors. The all mixed oxide concept is advantageous because it concentrates all the Pu bearing rods in a definite number of bundles of the reload, and subsequently avoids mixing still somewhat enriched uranium with the depleted uranium of the mixed oxide rods during the reprocessing operation. On the other hand, it implies the cost of a larger number of plutonium concentrations in the fabricated mixed oxide rods.

2.1.2. Scheme of utilization

The many experiments which have been conducted in both critical facilities and reactors, have shown that the calculation tools and methods developed for uranium reloads are quite satisfactory to design plutonium fuels.

In particular, the initial reactivity measurements and the measurement of the power distribution in the reactor have shown a very good agreement between theory and experience.

The methods of design are thus suitable for calculating accurately a scheme of Pu utilization and allow to choose the optimum one for the reactor and the type of fuel policy adopted in a particular country. For example, it is possible to recycle self-generated Pu in LWR's (about 20 to 30% of the rods enriched with Pu) with minor modification to the core and control management. By modifying the control strategy and using an adequate number or enrichments, particular reactors can recycle Pu in a proportion of 70% of the fuel reloadings and this figure could even be increased to 100% in reactors specialized for such a fuel. A particular scheme can be adopted on the basis of Pu availability and return to fully uranium enriched core is always possible.

2.1.3. Problems in Pu utilization in LWR's

The fuel designer will have to solve problems which are connected to the use of Pu in LWR's on an industrial scale.

The origins of Pu could be different for the same reload if a large amount of Pu is recycled in the reactor (e.g. for more than the self-generated amount). In that case the isotopic composition might be different and the physics designer will have to optimize the Pu distribution already in the assembly itself and afterwards in the reload scheme.

Furthermore, the introduction in LWR's of second or third generation plutonium must be carefully studied because of the isotopic composition of Pu (which leads to high plutonium content) and U (which leads to isotopes of high activity, difficult to handle). The conclusions could be in this particular case that it is more beneficial to store these second generation fuels before reprocessing for later use in FBR's.

The designers and the customers have to perform the safety analysis and present the safety report to the licensing authorities.

For nominal operation, the design methods and the Pu behaviour are sufficiently known to consider the Pu reload design on the same quality level as the uranium assembly one.

But the introduction of Pu in a core will probably be considered by some licensing authorities as a major perturbation and a safety analysis by difference will not be considered as sufficiently convincing. Thus a complete safety analysis must be foreseen by the customer assisted by the fuel designer. So, evaluation of incidents such as rod ejection, steam line break and loss of coolant will be probably asked even if in this latter case the physics properties and the flexibility of the plutonium reloads can lead to a safer situation (cf. GESMO).

2.2. Pu use in fast reactors

If in the early designs of FBR fuel, U-Pu metallic alloy was considered, the mixed oxide $(U\text{-}Pu)O_2$ has been very quickly recognized as the reference material for the present generation of fast reactors.

An attractive alternative for large size commercial fast reactors is the mixed carbide (U-Pu)C but its development has not yet reached a state which would permit an adequate assessment of its use in commercial FBR's.

The main differences between the LWR recycle fuel and the FBR fuel are:

- high enrichment
- high power rating (in W/g)
- high fuel burn-up
- high neutron fluence in the structural materials
- high cladding temperature

The current values for the existing and planned reactors are summarized in Table 1 hereafter.

The main criteria considered for the design of the fuel pin are deduced from safety and reliability considerations:

-Stability of fuel material in the pin (assuming as low as possible redistribution of Pu in the pin). This leads to a "no melting" criterion which requires a fuel temperature below 2750°C.

- Like in the thermal reactors, the pin cladding constitutes the first barrier for Pu and fission products . This leads to limit the elastic strain of the cladding to 0.2%, the thermal creep strain to 0.2% and the cladding corrosion attack to 100 microns.

- Stability of the bundle configuration to allow coolant passage. This leads to an expected neutron induced swelling of less than 5%.

For the future commercial large breeders, the anticipated technical performance will be substantially similar to the ones of the existing prototypes:

- the rating will remain essentially constant, i.e. about 450 W/cm maximum nominal, except if a carbide fuel is selected;

- the expected average burn-up should increase to 80,000 or 100,000 MWd/t, thanks to improvements in cladding and structural material having considerably better swelling behaviour and improved corrosion resistance both to Na and to fission products.

Considering the economics, important aspects are the breeding gain, the Pu inventory and the doubling time.

The in-pile Pu inventory is reduced by increasing the linear rating (if the central temperature can be limited), by decreasing the pin diameter (but at the expenses of increased fabrication cost and decrease of breeding gain) and by an improved power distribution within the reactor. In the future, one can expect a value of about 3000 kg of fissile Pu per GW.

The internal breeding gain is improved by an increase of the pin diameter, of the fuel density and a decrease of the structural material fraction. The external breeding gain is improved by the increase of axial and radial blanket thickness, and of UO_2 volume fraction in the blanket. One can expect in the future a breeding ratio of 1.2 to 1.22 for a mixed oxide core and a ratio of 1.25 to 1.30 for the carbide core.

One has to mention here the mixed oxide breeder with the "heterogeneous" concept, i.e. containing inner blanket material in the core regions. This concept could lead to improve both safety and economic aspects. If the trend presently observed in the studies is confirmed, this breeding gain could be of 1.30 to 1.40.

The consequent doubling time which combines breeding gain and total Pu inventory would be around 20 years in case of an homogeneous mixed oxide core, less than 15 years for an heterogeneous mixed oxide core and 10-11 years for a carbide core.

3. PLUTONIUM PRODUCTION

3000 tons heavy metal per year is the rated capacity of existing facilities for the reprocessing of LWR oxide in the world (without China, USSR, and Eastern countries), corresponding to a production capacity of some 20 tons of fissile plutonium per year. Yet their cumulative throughput is barely above 500 THM to-date - or 1,5 year of production of the oldest facilities put into service 10 years ago - while hundreds of tons of fuel are tied up in storage pads and the reprocessing of 100 tons in 1977 could be considered as a real achievement. In fact, 5 of the 7 existing facilities will not be operated in the 70's, if ever. To make the future still a little more confusing, the plutonium contained in spent LWR fuel could not, if reprocessed, be utilized for lack of adequate conversion and fabrication capacities.

This is not the place to analyse the reasons of such a warped situation. Some of these reasons are certainly technical and the transition of low burn-up, low Pu content metallic fuel to high burn-up, high Pu content oxide fuel has quite normally brought some problems; these have materialized in particular in the form of insoluble compounds associated with solvent radiolysis.

However, many reasons of the present situation are of political or administrative origin and some are directly linked with the very aim of reprocessing, the recovery of plutonium.

There is no doubt that all these difficulties will finally be resolved, so that the benefits of reprocessing and Pu utilization for energy economy and later energy autonomy will accrue while the risks of the out of pile Pu cycle are minimized, e.g. by such concepts as co-located facilities, master blends, etc... But this will take time in practice since it will take at least 10 years for new projects to materialize; effective capacity will thus lag behind demand at least until the beginning of the 90's.

When the present difficulties are resolved, LWR fuel reprocessing will in fact bring back to the reprocessing facilities a few years later, new technical problems in the form of irradiated mixed oxide fuels. They will arise both at the head-end (dissolution of Pu dioxide) and at the tail-end (concentration and activity of plutonium).

Those problems will be still more acute for FBR plutonium fuel which will bring the specific aspects of sodium contamination and high decay heat. In addition, reprocessing is essential to the breeder concept while LWR Pu elements can, without large difficulty or penalty, be stored even for a long period. However, the different time scales of Pu LWR recycle and commercial FBR introduction quite normally call for LWR Pu reprocessing as an intermediate step in the implementation of a complete industrial Pu cycle, in addition to the continuing R & D efforts on specific aspects of FBR fuel. It is clear that both routes should be actively pursued right now if previous mistakes are to be avoided and if a complete FBR fuel cycle is to be available at the time of commercial introduction of the FBR's, in the mid 90's.

4. PERSPECTIVES of PLUTONIUM UTILIZATION

4.1. Short and medium term period (1976-1990)

4.1.1. Reprocessing facilities and Pu oxide production

At the present time, the only operating facilities for LWR fuels are La Hague and WAK; as to GCR fuel, Marcoule, La Hague and Windscale are in operation. The corresponding fissile Pu production amounts to a total of 3 tons in 1977.

The increasing capacity of La Hague, the start up of the PNC plant in Japan and the reprocessing of AGR fuel in Great-Britain could result in a fissile Pu production of 10 tons in 1980. A further expected development of the reprocessing capacity in the world (Barnwell, NFS and EXXON plants in the U.S.A, BNFL THORP plant in the U.K, PWK plant in FRG, Belgoprocess in Belgium, PNC commercial plant in Japan, Trombay plant in India) could increase that production up to 40-50 tons of fissile plutonium. By 1990, such an evolution in reprocessing capacity corresponds to a cumulative fissile Pu production by reprocessing plants of 300 tons. At that period, the cumulative fissile Pu production by the LWR's and GCR's (including AGR's) would amount to 500 tons.

4.1.2. FBR development

In 77, the FBR's on line as demonstration plants are PHENIX and PFR, corresponding to $\pm$ 0.5 GW. With SNR, that power will be $\pm$ 0.8 GW around 1980; the introduction of SUPERPHENIX , MONJU in Japan and CLINCH RIVER breeder reactor in the U.S.A, will add $\pm$ 1.9 GW around 1985. One can expect a further development (SNR 2 in FRG, CFR 1 in the U.K, SAONE in France, Prototype large breeder reactor in the U.S.A) so that the FBR capacity at the beginning of the 90's might be 7 to 8 GW.

Such an evolution corresponds to a cumulative fissile Pu demand of 55-70 tons in 1990 (R & D needs included), to be compared to 500 tons produced in the thermal reactors and 300 tons available ex reprocessing plants. Therefore, there remains a considerable Pu surplus available for further FBR development or for recycling in thermal reactors.

4.2. Long term strategies (up to 2010)

4.2.1. Extreme situations

In a first step, the extreme situations have been studied, before taking into account the technical, industrial and political constraints as well as the intermediate strategies.

The strategies are studied in terms of Pu needs and availability to minimize the plutonium inventory as soon as possible after the insertion on a commercial scale of FBR's and for recycle. The total LWR + FBR capacity is supposed to be $\pm$ 3000 GW in 2010 (cfr. 1. Table II).

The annual fissile Pu production and needs are taken as follows (kg/GW-year):

	LWR U	LWR 1st recycle	FBR (Breeding gain: 0.2)
Production	160	550	1110
Needs	-	880	1000

These figures take into account fabrication and reprocessing losses. The case of Pu recycle is based on a mixture of Pu with depleted uranium, in order to compare Pu recycle with FBR's and to determine the part of nuclear capacity supported by the Pu. The initial inventory for FBR's is 3 tons fissile Pu and, in case of FBR fuel reprocessing, the out of pile inventory is 2 tons. The delay between end of reprocessing and insertion in a reactor is 1 year, as well as between fuel discharge and end of reprocessing. The beginning of operation, not on a demonstration scale but on a commercial scale, is 85 for recycle and 90 for FBR's insertion. Table II and Figure 1 give the results of the study:

- In the extreme situation where neither FBR insertion nor recycle occurs, 5000 tons of fissile Pu would have to be stored in 2010.

- If one considers the maximum insertion of FBR's in 2010 more than 25% (860 GW) of the total capacity (3160 GW) would be FBR, if FBR fuel is reprocessed (curve 1) and half of this if fuel is not reprocessed (curve 5). If the FBR's are just self-sufficient (net production equal to zero, taking into account the losses), the FBR capacities must be reduced by 10%.

- In case of recycle of the 1st generation Pu, the corresponding share of LWR-Pu in total capacity is 11 to 15% (zone 6) according to whether FBR's are introduced or not (curve 4). In the latter case, 1750 tons fissile Pu will be stored in 2010. The insertion of FBR with the 2nd generation LWR-Pu results in a share of FBR's capacity of respectively 12% and 5% whether the FBR fuel is reprocessed or not.

- The curves 2 and 3 show the total capacity corresponding to FBR plus LWR-Pu fuels.

4.2.2. Possible strategies from an industrial point of view

From the results of the extreme situations study, it becomes obvious that, if the LWR-U fuel is reprocessed, the plutonium availability is not a constraint on the FBR's development, even if the FBR fuel is not reprocessed during a long period. The real constraints will be of technical, industrial or political nature. Due to these constraints, it seems reasonable to assume that a maximum FBR introduction would follow the same pattern as the now historical LWR one before. Estimates for FBR capacity in 1995 are 35 to 40 GW (65% in Europe, 20% in the U.S.A, 15% in Asia) which corresponds to the LWR capacity in 1977 (39 GW); as well as 5 GW

FBR in 1990 corresponds to 5.3 GW LWR in 1969. The expected growth of LWR's being respectively 24%, 16% and 13% / year for the periods 1975-80, 1980-85, 1985-90, the corresponding FBR generating capacity should be ± 110 GW by 2000, 240 by 2005 and 420 by 2010. The zone 7 on Figure 1 shows a possible zone for FBR penetration with such figures as maximum curve.

From the Table II and the Figure 1 , it is easily shown that for such a penetration of FBR's in the nuclear generating capacity, there still remains Pu available for recycle, especially if one considers that the commercial penetration of FBR's from 1990 must normally be followed by FBR fuel reprocessing much before 2010.

The economic impact of recycling the first generation Pu is large, since it allows not only substantial savings of U_3O_8 and SW requirements, but also as to Pu storage problems and cost. In addition, it immobilizes the huge Pu inventory safely in power reactors.

5. FABRICATION of Pu FUEL

5.1. The present situation

In Europe, for thermal reactors, the BN plant at Dessel and the Alkem plant in Hanau have almost the same 40 T/year range capacity. On the fast reactor fuel side, in addition to these plants, which are able to produce either thermal or fast reactor fuel, the CEA plant in Cadarache and the BNFL plant in Windscale have both FBR fuel capacities of about 10 T/year. In the U.S.A, three pilot plants are presently operational : the Babcock & Wilcox plant at Apollo, the Exxon plant at Richland and the Westinghouse plant at Cheswick.

All these capacities are still in the industrial pilot range but the experience gained with their operation since several years allows without any real major problems to extrapolate to larger industrial capacities.

Probably, the most important problem that such Pu fuel fabrication plants have to face is the personnel formation and training. This is one of the major safety aspects of the plant operation. It is quite impossible to start a large Pu plant without an operating crew having several years experience in the field of Pu handling. After the start up of such a plant, only a progressive and slow personnel increase can be allowed. This essential safety consideration induces a practical limitation in the number of plants which can be licensed at any given time to fabricate Pu fuel.

5.2. The future trends

In spite of the fact that there exists a substantial difference between thermal and fast nuclear fuel, the general process is about the same ; only fabrication step capacities are different according to the

difference in the specification of both fuels. For instance, the number of pins per kg fuel is much higher for fast reactor fuel requesting a bigger rod filling and end cap welding capacity.

Also, for both types of fuel, mechanization is a necessity:

- Even when the annual capacity of the production line is as small as 40 tons of fuel, the total amount of material circulating in the glove box system is several hundreds kg per day and is too large to be transferred by hand.

- The γ and η doses to personnel, induced by the Am and Pu isotopes must be minimized: a reliable mechanization in that respect reduces personnel exposure and allows n gher throughputs.

In addition to this, shielding will be requested where pure plutonium or high plutonium content materials are handled. This means practically that a fast reactor fuel production line will be fully shielded when using LWR plutonium; on the contrary, a thermal reactor fuel production line will require shielding only where pure PuO_2 is handled, since once mixed, self-shielding of the blend itself reduces substantially the radiation problems.

In general, distant operation will be really needed for fast and thermal reactor fuel lines with an appropriate shielding where pure or high plutonium content oxides are processed.

As far as the shielding is concerned, it is neither necessary nor advantageous to place the equipment in heavily shielded cells in which the visibility is poor and the accessibility difficult; it is better to redesign the equipment itself so that the product circulates for instance in a tight channel inside the glove-box with small lead or iron screens. disposed close to the plutonium fuel, the visibility being achieved by means of mirrors. This reduces also the dust formation in the glove-box and the associated problems (dust deposition on glove-box pannels, leading to increased personnel exposure, and α contamination in case of incident). This concept leads in addition to much easier maintenance of the equipment itself.

5.3. Organization

No fixed limit exists between reprocessing and fuel refabrication flow sheets. The delimitation must be fixed on the basis of the location of the plants (co-location or not) and the technical responsibilities involved (integration of plants or not).

In general, the reprocessor will prefer to transfer a product which can be easily analysed but also easily transported if the location of the refabrication plant requires it. On the other hand, the refabricator prefers to start its process with materials having all the reproductibility characteristics necessary to guarantee the product quality. This is the reason why practically all the uranium fuel manufacturers fabricate their

own enriched uranium oxide powders starting from UF_6. In the case of plutonium, the refabricator prefers also to start with a product which liberates a minimum amount of dust.

According to this principle, the Pu fuel production should start from Pu nitrate and UO_3, both being produced by the reprocessing plant.

If the reprocessing and the refabrication plants are not co-located, transportation of Pu on large distances has to be contemplated. A proposal in order to improve safeguards is to blend the plutonium oxide with uranium oxide, or even to co-precipitate them. In addition, the blended oxides may be stored over a much longer period than pure PuO_2 thanks to the shielding effect of UO_2.

The necessity to protect the nuclear fuel production site favours co-location which reduces also the number of transports and increases therefore the safety aspect of the system.

Co-location of reprocessing, refabrication and waste management plants allows also to combine most of the auxiliary services (analytical laboratories, other test laboratories, work-shops, special nuclear material accountability, wardening, etc...) and therefore favours the economy of the co-located plants.

6. CONCLUSIONS

Plutonium use in power reactors as such does not raise any major problem. This has been demonstrated technically in light water reactors and in fast reactors as well. Its economic incentive is high, both in the short term through yellow cake and separative work savings, and in the long term through the offering of a new energy resource.

Obviously, the decision not to proceed in the way of reprocessing reactor fuel is equivalent to preclude all future plutonium use. Reprocessing light water reactor fuel is thus an absolute prerequisite to the following statements:

1. Plutonium availability is not a constraint whatsoever on fast reactors development for the next 30 years.

2. Plutonium will be available during the 80's in large quantities for recycling in light water reactors.

3. Plutonium recycling in light water reactors will in all probability go on for a long time during fast reactors implementation.

4. Reprocessing techniques for plutonium bearing fuel can be developed on a timely basis without putting too heavy constraints on plutonium use.

5. A most important problem to be faced by the plutonium industry lies in the setting up and training of highly experienced working teams. This means a need for continuity in the plutonium programme.

6. Concentration of back-end cycle production units deserves attention for safeguards, safety, and economy.

TABLE I : FBR fuel characteristics

DATA	CRBR	RAPSODIE Fortissimo	PHENIX	SUPER PHENIX	DFR	PFR	CFR	SNR 300		SNR 2 (1300 MWe)	BN 300	BN 600
								Mk Ia	Mk II			
Reactor power (MWe)	380	-	233	1200	15	254	1300	300	300	1300	150 +	600
Fuel pin diameter (mm)	5.84	5.1	6.55	8.6	5.84	5.84	5.84	6	7.6	7.6	6.1	6.9
Na outlet T° (°C)		514	560		400	577		546	546	540		
Maximum nominal linear power (W/cm)	475	430	430	450	435	460	445	355	450	440	440	530
Nominal midwall clad T° (°C)	640	650	640	620	620	613	-	620	620	660		
Average burn-up (MWd/kg)	50	-	50	70		75		57	57	57	50	50

TABLE II : Pu utilization strategies.

	1980	1990	2000	2010
1. LWR only				
- capacity (GW)	155	585	1540	3160
- cumulative Pu production (LWR + GCR) fissile T	95	515	1830	5070
2. LWR + FBR - no recycle				
2.1. FBR fuel reprocessed				
- LWR capacity (GW)	155	560	1220	2300
- FBR capacity (GW)		25	320	860
2.2. FBR fuel not reprocessed				
- LWR capacity (GW)	155	580	1420	2760
- FBR capacity (GW)		5	120	400
3. LWR Recycle - no FBR				
- LWR-Pu capacity (GW)		90	200	450
- cumulative Pu production (LWR 2d generation + GCR) fissile T	25	100	590	1760
4. LWR + FBR - recycle				
4.1. FBR fuel reprocessed				
- LWR capacity (GW)	155	575	1420	2780
of which LWR-Pu capacity (GW)		90	190	400
- FBR capacity (GW)		10	120	380
4.2. FBR fuel not reprocessed				
- LWR capacity (GW)	155	580	1490	3000
of which LWR-Pu capacity (GW)		90	195	430
- FBR capacity (GW)		5	50	160
5. FBR possible capacity from an industrial point of view	.8	5	80-110	320-420

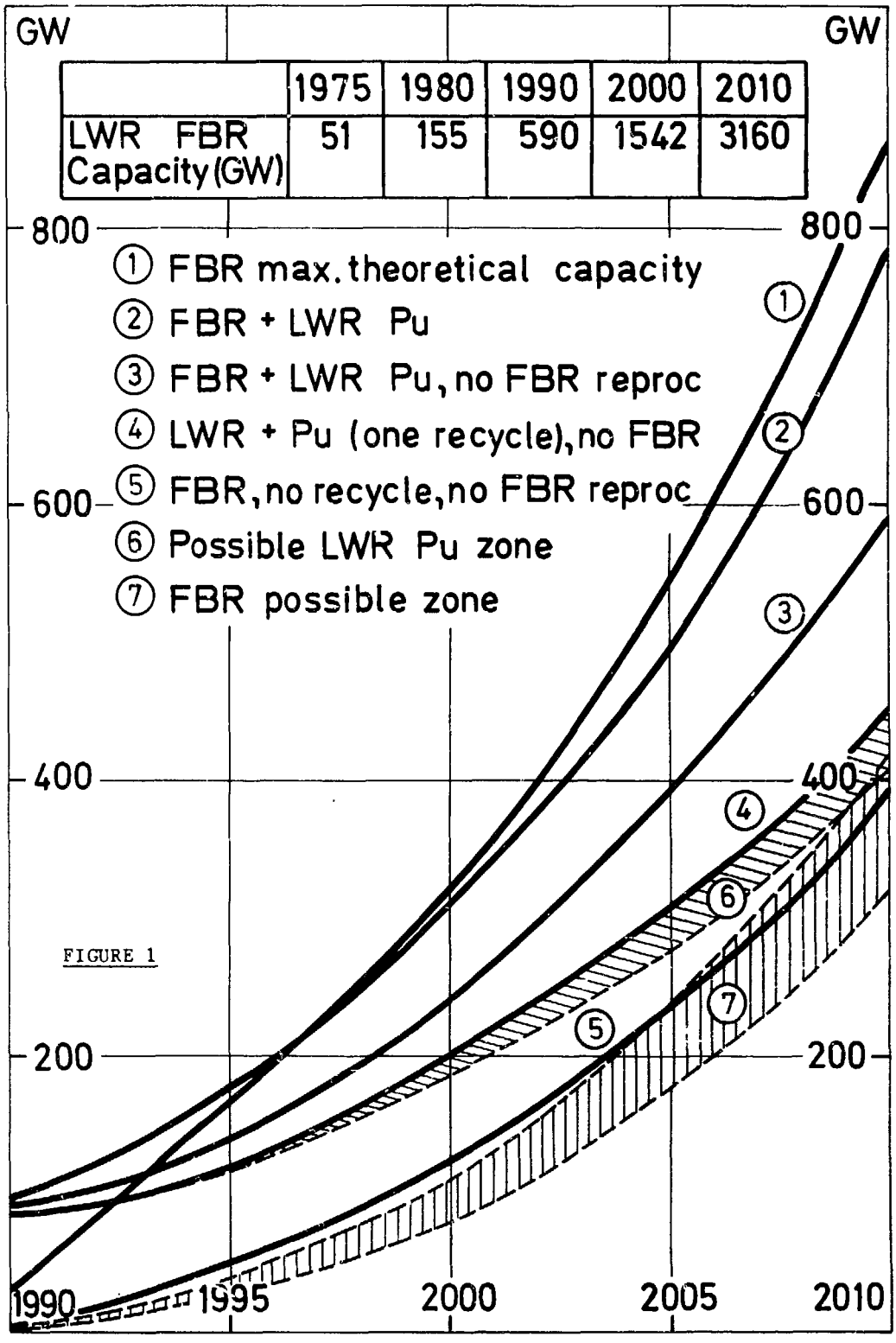
GW
GW
1975 1980 1990 2000 2010
LWR FBR Capacity (GW) 51 155 590 1542 3160
800
600
400
200
① FBR max. theoretical capacity
② FBR + LWR Pu
③ FBR + LWR Pu, no FBR reproc
④ LWR + Pu (one recycle), no FBR
⑤ FBR, no recycle, no FBR reproc
⑥ Possible LWR Pu zone
⑦ FBR possible zone
1990
1995
2000
2005
2010

FIGURE 1