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Disposition of Separated Plutonium

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In the immediate term, plutonium, recovered from dismantled nuclear warheads and from civil reprocessing plants, will have to be stored securely, and under international safeguards if possible. In the intermediate term, the principal alternatives for disposition of this plutonium are: irradiation in mixed-oxide (MOX) fuel assemblies in commercial unmodified light-water reactors or in specially adapted light-water reactors capable of operating with full cores of MOX fuel or incorporation into a matrix with high-level waste (HLW). Of these three options, blending plutonium into HLW as it is being glassified for final disposal is probably the least costly and the least burdensome to safeguards resources.

INTRODUCTION

Today there is a growing world surplus of separated plutonium. An imbalance between supply and demand in the commercial sector is being compounded by deep cuts in the CIS and US nuclear arsenals, with a concurrent release of large quantities of weapons plutonium. This paper analyzes alternative approaches for dealing with this material, focusing principally on economic and security considerations.

The nuclear warheads that are to be dismantled without replacement contain perhaps 150–200 tonnes of plutonium. Dismantlement has already begun and is expected to continue for at least a decade. At the same time, pro-

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duction of new weapons plutonium has almost ended in both the US and the Commonwealth of Independent States (CIS), and the rate of production in other countries is low.

By contrast, commercial separation of civilian plutonium is undergoing major expansion. During the 1990s, some 200 tonnes of civil plutonium are due to be separated from power-reactor fuel originating in 14 countries. Most reprocessing will take place in three nuclear-weapon states (the UK, France, and Russia). If all of the recovered plutonium is returned to the countries of origin—about half to non-weapons states—this would greatly increase the transport and handling of separated weapons-usable plutonium around the world.

The impending surpluses of both separated weapons and civil plutonium present a global security problem. All plutonium (except relatively pure Pu-238) is weapons-usable.[1] In periods of crisis, stocks in non-weapons states could greatly lower the threshold to nuclear weapons proliferation. In weapons states, plutonium stockpiles could lower the threshold to "breakout" from reductions agreements. In any country, they pose a severe challenge to physical-security systems. The diversion of even a few kilograms of plutonium to a terrorist group could cause a major international crisis.

In this paper we assess three basic alternatives for disposition of weapons plutonium and already-separated civil plutonium:

- Safeguarded long-term storage.
- Irradiation in nuclear-reactor fuel.
- Mixing with high-level radioactive waste as it is glassified in preparation for geological disposal.

We begin, however, by describing the sources of the separated plutonium and current plans for its storage.

SOURCES OF SEPARATED PLUTONIUM

Table 1 gives an overview of the distribution of world plutonium stocks. Separated plutonium is accumulating in large quantities from two sources: the dismantlement of CIS and US warheads, and large-scale chemical reprocessing of spent fuel from civilian nuclear power reactors.

Dismantled Warheads

Under the reciprocal reductions announced by Presidents Bush, Gorbachev

Table 1: Summary table of world plutonium stocks and surpluses at the end of 1990.[a]

	Nuclear-weapon countries	Non-nuclear weapon countries	Non-NPT countries	Total
		tonnes		
In weapons[b]	260	0	< 1[c]	260
Civil				
In irradiated fuel	296[c,d]	218	17[e]	531
	366[f]	*148*[f]		
Recycled in MOX	31[g]	18[h]	0	49
Stored as oxide	64[i]	8[j]	< 0.5	72
Total (by ownership)	651	244	18	913
Total (by location)	***721***[f]	***174***[f]	**18**	**913**

a. Source: D. Albright, F. Berkhout and W. Walker, *World Inventory of Plutonium and Highly-Enriched Uranium* (Oxford: Oxford University Press, 1993) table 12.3.
b. Includes associated stocks. These figures have much larger error margins (± 20 percent) than the rest of the table (± 10 percent).
c. Includes Israel and India.
d. Includes an estimated five tonnes of plutonium contained in East European and Finnish spent fuel sent to Chelyabinsk under "take-back" arrangements (adjacent figure of 218 tonnes in the next column excludes this amount).
e. Includes India, Pakistan, Brazil, Argentina and Taiwan.
f. Figures in italics take account of plutonium held in store in France and the United Kingdom in spent fuel or as separated plutonium, but belonging to utilities in non-nuclear weapon states customer countries.
g. Comprises 25.3 tonnes of civil plutonium recycled in fast reactors and other R&D facilities (5 tonnes in the UK, 12.5 in France, 0.5 in the CIS, and 6.7 tonnes in the US—the last figure includes material imported from the UK), and 5.8 tonnes of plutonium recycled in thermal reactors in France. Four and one half tonnes of foreign-owned plutonium in Superphénix fuel is not included under this column, but in the adjacent column.
h. Comprises 4.5 and 2.5 tonnes of plutonium recycled in Japanese and German fast reactors respectively, 4.5 tonnes of plutonium owned by Italian, German and Dutch utilities loaded into Superphénix, and 6.5 tonnes of plutonium recycled in German, Swiss and Belgian thermal reactors.
i. Includes two tonnes of plutonium separated at Sellafield and La Hague, but not yet returned to owners in non-nuclear weapon states.
j. Comprises 27.8 tonnes of plutonium separated from non-nuclear weapon states fuel, less 18 tonnes of recycled material, less two tonnes held in store (see note g).

and Yeltsin, Russia and the US are each expected to retain no more than 1,000–2,000 tactical nuclear warheads. In parallel, the Bush-Yeltsin framework agreement of June 1992 envisions limiting the strategic arsenals to 3,000–3,500 warheads each by approximately the year 2000.* If the overall cuts to approximately 5,000 total warheads each for the US and Russia are carried through, we estimate that over the next decade the US will retire approximately 15,000 warheads, and Russia about twice as many.

Modern thermonuclear warheads contain a fission explosive "primary" and one or more "secondaries" that typically contain fission as well as fusion fuel. Plutonium is contained in the fissile "pit" of the primary. The Nagasaki fission weapon contained six kilograms.[2] Today's thermonuclear warheads contain less plutonium; we assume three kilograms on average.[3] (They also contain significant quantities of highly enriched uranium [HEU] in both their primaries and secondaries—about 15 kilograms on average.[4])

Therefore, projected cuts in the nuclear arsenals would release about 45 tonnes of plutonium from US weapons and some 90 tonnes from the CIS arsenal. Additional quantities of surplus material in plutonium components from already dismantled warheads and in unprocessed production scrap could bring the totals up to about 70 and 120 tonnes, respectively.

Warhead dismantlement involves the removal of electronics, the separation of the primary and secondaries, and then removal of the high-explosive implosion system from around the plutonium-containing pit. US nuclear warheads are currently dismantled at the Pantex facility near Amarillo, Texas at a rate of about 2,000 per year.[5] Plutonium components from these warheads are not being further processed. The Rocky Flats Plant in Colorado, to which they would have been shipped previously for extraction of the plutonium, storage and perhaps fabrication into new warhead pits, has been shut since November 1989 due to environmental and safety concerns. In the absence of an alternative, recovered plutonium pits from dismantled warheads are therefore being stored intact at Pantex in heavily protected bunkers ("igloos"). There are 60 igloos at Pantex, but as of early 1992, only 18 have been qualified to store pits. Each of these igloos can currently hold up to 240 plutonium pits. The possibility of extending this capacity to 400 warheads is being examined. Storage at the Los Alamos National Laboratory and at the former plutonium-

* All of the other former Soviet republics have agreed to become non-nuclear-weapon states. However, Belarus, Kazakhstan, and the Ukraine each retain joint ownership with Russia of the warheads formerly located on their territories while the warheads are being dismantled in Russia, and of the recovered fissile materials. We therefore will refer to the warheads of the former Soviet Union that are to be dismantled and the recovered fissile materials as CIS warheads and materials.

production sites at Hanford, Washington and Aiken, South Carolina is also being explored.[6]

Arrangements for dismantlement and storage of CIS warheads are less clear. Dismantlement can apparently be carried out at four sites with a combined maximum rate of 5,500–6,000 warheads per year.[7] However, in 1992, the actual dismantlement rate was reportedly about the same as in the US—2,000 per year. Weapons components are being temporarily stored at the dismantlement sites. At the insistence of other CIS states, weapons components removed from Ukraine and stored in Russia are being jointly monitored by both countries under an agreement signed in April 1992. The Russian government has said that it would consider similar agreements with Belarus and Kazakhstan.[8] It has also expressed a willingness to comply with a US Congressional requirement that the US be able to verify the subsequent peaceful use of any weapons materials in any storage facility constructed with US assistance.[9] Thus far, however, the US has refused to consider reciprocal safeguards on surplus US weapons materials.[10]

Russia has proposed that eventually the fissile materials recovered from CIS warheads be stored at a single secure underground facility, probably near the Tomsk-7 weapons plutonium-production complex, and it has requested US financial assistance for its construction. As of June 1992, the facility plans called for an initial capacity sufficient to hold about 45,000 storage containers. Each container would hold either fissile components of dismantled warheads or fissile material reduced to subcritical metal cylinders ("pucks") containing 4–5 kilograms of plutonium or 10 kilograms of HEU.[11] A contemplated second stage of construction could increase the storage capacity to about 100,000 containers. This expansion may be unnecessary, however, if, as has recently been agreed in principal, the surplus HEU is sold to the US for dilution to low-enriched uranium for use as a power-reactor fuel.[12]

The dismantlement of CIS and US warheads is being accompanied by an almost complete halt in production of fissile material for weapons. Production of HEU for weapons ended in the US in 1964. Plutonium production for weapons in the US ended in 1988 when the last four operating US production reactors (out of a maximum total of 14) were shut down for safety reasons. In July 1992, President Bush announced that the US would not resume production of either plutonium or highly enriched uranium for weapons.[13]

Production of HEU for weapons ended in Russia in 1989. Russian production of weapons plutonium continues, but at a low level. (All CIS weapons material production facilities are located in Russia.) So far, 10 out of 13 plutonium-production reactors have been shut down.[14] The three remaining reactors continue to produce fresh weapons plutonium while supplying heat and

electricity to nearby towns in the Tomsk and Krasnoyarsk regions.[15] President Yeltsin has reconfirmed a 1989 commitment by President Gorbachev that all Russian military plutonium production will end by the year 2000.[16]

Reprocessing of Power-Reactor Fuel

By the end of 1990, just over 120 tonnes of civilian plutonium had been separated from fuels discharged from the world's power reactors. Of this, some 50 tonnes had been recycled as nuclear fuel—mostly for demonstration fast-neutron plutonium-breeder reactors. The remaining 70 tonnes was stored, principally at four reprocessing plants: Sellafield in the UK, Chelyabinsk-65 in Russia, and La Hague and Marcoule in France. Most is stored as sintered plutonium oxide powder canned in subcritical masses.

With substantial new reprocessing capacity coming on line in the UK and France, about 190 tonnes of plutonium is scheduled to be separated in commercial reprocessing plants by the year 2000. (Table 2 lists the world's commercial reprocessing plants.)

Some of this plutonium will be recycled in fast-neutron reactors, and a larger amount is planned to be recycled as mixed-oxide (uranium-plutonium oxide) fuel (MOX) for light-water power reactors (LWRs). Given likely delays in the construction and operation of MOX fabrication facilities and in the licensing of reactors to accept MOX fuel, it appears unlikely that more than 60–70 tonnes of plutonium will be used in fuel through 2000. Therefore, unless reprocessing activities are slowed, the surplus of stored civil plutonium is likely to increase to about 200 tonnes by the turn of the century. Appendix A provides a detailed presentation of world balances of civil plutonium as of 1990 and as projected to the year 2000.

As noted in appendix A, about half of the separated civil plutonium arising in the 1990s will belong to non-weapon states—principally Japan and Germany—that have sent their spent fuel to Britain and France to be reprocessed. Most of these non-weapon states have explicit policies against stockpiling plutonium, consistent with the statutory position of the International Atomic Energy Agency (IAEA) that national stockpiles of excess weapons-usable fissile material should be avoided.[17] Notwithstanding this policy, Japan recently announed that it intended to store for at least three years plutonium brought back this year from France. In general, however, the non-weapon states therefore may have no practical alternative to storing their excess plutonium in Britain and France until it can be disposed of in a responsible way.

As discussed in appendix B, there is currently no economic rationale for plutonium recycling, which appears motivated today primarily by the desire of

Table 2: Civilian reprocessing plants around the world.[a]

Country	Location	Owner/ Operator	Facility	Fuel	Capacity $HM\ yr^{-1}$	Operated
Belgium	Mol	Eurochemic	Mol	oxide	30	1966–1974
France	Marcoule	Cogema	UP1	metal	400	1958–2000?
	La Hague	Cogema	UP2	metal + oxide	400	1966–1987 (metal)
	La Hague	Cogema	UP3	oxide	800	1990–
	La Hague	Cogema	UP2-800	oxide	800	1993–
Germany	Karlsruhe	KfK/DWK	WAK	oxide	35	1971–1990
India	Tarapur	DAE	PREFRE	oxide	30–150	1982–
	Kalpakkam	DAE		oxide	100–200	1993/4?-
Japan	Tokai-mura	PNC	Tokai	oxide	90	1981–
	Rokkasho-mura	JNFS	Rokkasho	oxide	800	2002?–
Russia[b]	Chelyabinsk-65	MinAtom RF	Mayak	oxide	600	1978–
United Kingdom	Sellafield	BNF plc	B205	metal	1,500	1964–2010?
			B204/205	oxide	300	1969–1973
			THORP	oxide	700	1993–
	Thurso	UKAEA	DNPDE	oxide (MTR)	< 1	1959–
				oxide (FBR)	7	1980–1997?
United States	West Valley	NFS	West Valley	oxide	300	1966–1972

a. HM = tonnes heavy metal in fuel; MTR = materials test reactor; FBR = fast breeder reactor; UP = Usine de Plutonium; KfK = Kernforschungszentrum Karlsruhe; WAK = Wiederanfarbeitungsanlage Karlsruhe; DAE = Department of Atomic Energy; PREFRE = Power Reactor Fuel Reprocessing; PNC = Power Reactor and Nuclear Fuel Development Corporation; JNFS = Japan Nuclear Fuel Service Company; BNF = British Nuclear Fuels plc; THORP = Thermal Oxide Reprocessing Plant; UKAEA = UK Atomic Energy Authority; DNPDE = Dounreay Nuclear Power Development Establishment; NFS = Nuclear Fuel Services; DWK = Deutsche Gesellschaft Für Wiederanfarbeitung von Kernbrennstoffe; MinAtom RF = Ministry of Atomic Power, Russian Federation.

b. Construction was begun on another plant at Krasnoyarsk in Siberia (Dodonovo-27), but halted when about 30 percent complete. We assume the plant will not be completed.

electric utilities in a few industrialized countries to rid themselves of their accumulated and anticipated stockpiles of separated plutonium. It now appears possible that, in light of widespread utility disinterest and even opposition to recycling, commercial reprocessing will be phased out over the next decade. If so, the world will be left with the daunting, but *finite* problem of disposing safely of a few hundred tonnes of separated civilian and weapons plutonium.

SAFEGUARDED STORAGE OF SEPARATED PLUTONIUM

Unless plutonium is completely fissioned or launched into the sun, a large fraction will remain for a long time in one form of storage or another, whether as separated plutonium, in spent fuel, or fixed in another matrix such as high-level waste glass. Storage in spent fuel or high-level waste glass would make the storage form self-protecting to a significant extent because of the intense gamma radiation that associated fission products would emit for hundreds of years.[18]

Stored plutonium metal (in intact pits or pucks) or plutonium oxide requires far more organizational vigilance because the protective radiation from pure plutonium is relatively weak.[19] In the short term, all separated plutonium will be stored in one of these forms. We discuss in this section the economic and security aspects of plutonium metal and oxide storage. Then, in the following section, we consider ways to fix already separated plutonium into more proliferation-resistant forms.

International Safeguards

As noted above, Russia has indicated a willingness to have recovered weapons material be monitored under bilateral verification arrangements. To further lock in nuclear-weapon reductions, the US and Russia (and eventually the other weapons states as well) could move beyond ad hoc bilateral arrangements to guarantee peaceful uses for recovered weapons materials and accept international (i.e., IAEA) safeguards as well.

For such a regime to be credible, *all* civil nuclear materials would probably have to be brought under safeguards. Safeguards coverage could come about in either of two ways. First, under Article XII.A.5 of the IAEA Statute, member states may transfer stocks of fissionable material to the control of the IAEA, which is then responsible for storing and protecting them.[20] Alternatively, international safeguards on plutonium stores could be extended to all weapons- and non-weapons-states as part of a universal ban on the production

of unsafeguarded civil fissile materials. Under such circumstances, if surplus weapons plutonium were declared to be "civil," it would automatically come under safeguards. These extensions of IAEA safeguards could only be done, however, if the participating parties guaranteed sufficient additional resources for the new tasks.

Currently, stores of civil plutonium in Britain and France are not fully subject to IAEA safeguards although they are safeguarded by Euratom.* Stores of civil plutonium in Russia are not yet subject to any international oversight.

In addition, plutonium stores must, of course, be made relatively resistant to clandestine diversion by subnational groups. Strict physical and administrative control must be maintained by keeping a constant heavy guard, severely restricting access to the store and requiring that those who enter the store exit through portals equipped with detectors sensitive to the neutrons emitted by plutonium.[21] Plutonium containers could be tagged and sealed after their contents have been assayed and their gamma emissions measured to assure without a new assay being required that their contents have not been tampered with in storage. Such arrangements would effectively address subnational threats and, with regular international inspection, should inspire confidence in the international community that no state diversion is taking place.[22]

Costs of Storage

Storage of plutonium will be costly. The storage facility must be able to resist penetration by explosives, have fire suppression and cooling systems (especially if the plutonium is in metal form), and be equipped with a variety of sensing systems. In addition, there will be continuing high labor costs due to the large guard force. However, very little specific information is publicly available on the costs of large plutonium stores such as those at La Hague and Sellafield. Costs of $1–2 per gram of plutonium per year have been published, but without further explanation.[23] Information gleaned from interviews with utilities suggests that, in practice, the prices charged by reprocessors for plutonium storage may be higher than this, even approaching $4 per gram per year.

A 1990 US study estimated that a facility with capacity for 50 tonnes of plutonium oxide stored in 12,500 shipping canisters, each holding four kilograms of plutonium, would have a capital cost of about $240 million (1987$).[24]

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* All nuclear material in Western Europe declared as being civil is subject to Euratom safeguards. Japanese plutonium stored in Britain is subject to IAEA safeguards.

Table 3: Isotopic makeup and physical characteristics of plutonium.

Isotope	Half-life	Reactor-grade[a]		Weapons -grade[d]	Decay heat	Neutron emission rate[e]	Delayed neutron fraction[f] (thermal)
		Spent LEU fuel[b]	Spent MOX fuel[c]				
	years	*percent*			*kW t^{-1}*	*s^{-1} kg^{-1}*	
Pu-238	87.7	1.3	2.3	0.012	560	2.6×10^6	
Pu-239	24,100	60.3	38.1	93.8	1.9	22	0.0021
Pu-240	6,560	24.3	32.7	5.8	6.8	9.1×10^5	
Pu-241	14.4	8.3	16.9	0.23	4.2	49	0.0049
Pu-242	376,000	5.0	8.3	0.022	0.1	1.7×10^6	
Am-241	430	0.8	1.7	0.13	114	1,200	
Radiation hazard relative to Pu-239		190	290	34			
Maximum storage of separated Pu[g]		7	3	No limit			
Neutron emission rate ($s^{-1}\ kg^{-1}$)		3.3×10^5	5.0×10^5	5.3×10^4			
Decay heat ($kW\ t^{-1}$) of Pu after 10 years storage		14.3	24.4	2.4			

a. Plutonium stored for two years after separation, *Plutonium Fuel: An Assessment* (Paris: OECD/NEA, 1989) Table 12A.
b. Fuel irradiated in a PWR to 33 MWd kg^{-1} and stored for 10 years before reprocessing, *Plutonium Fuel: An Assessment*, (Paris: OECD/NEA, 1989) Table 9.
c. Fuel irradiated in a PWR to 43 MWd kg^{-1} and stored for 10 years before reprocessing, *Plutonium Fuel: An Assessment*, (Paris: OECD/NEA, 1989) Table 12B.
d. Weapon material stored for 10 years. Source: N.J. Nicholas, K.L. Coop and R.J. Estep, *Capability and Limitation Study of DDT Passive- Active Neutron Waste Assay Instrument*, Los Alamos National Laboratory, LA-12237-MS, 1992.
e. *Reversing the Arms Race*, F. von Hippel and R. Sagdeev, editors, (New York: Gordon and Breach Science Publishers, 1990) p. 315.
f. The fraction of neutrons yielded per fission of the isotope that are delayed neutrons. Source: G.R. Keepin, *Physics of Nuclear Kinetics*, (Reading: Addison-Wesley, 1965) p. 102.
g. The period of time separated plutonium could be stored before becoming unacceptable at the new MOX fabrication facilities. Assuming plutonium separated after 10 years fuel storage and a maximum Am-241 concentration of 2.5 percent.

With the addition of operating costs, the discounted storage cost would be about $0.84 per gram-year when the facility was full.[25] Another recent estimate attaches a higher capital cost to a storage facility of $1.5 billion for a store with a 90 tonne capacity, and proposes an undiscounted annual storage cost of $1–2 per gram of plutonium.[26]

An additional penalty associated with storage of reactor-grade plutonium is that the radiation hazard increases with time. About nine percent of the plutonium in 10-year-old LWR spent fuel is Pu-241, which decays with a 14.4 year half-life to americium-241 (see table 3). Americium-241 is a strong X-ray and gamma-ray emitter and its ingrowth will approximately double the radiation hazard from reactor-grade plutonium in two years. MOX fuel fabricators currently will not accept plutonium that contains more than 1.5–2.5 percent Am-241 (corresponding to about 4 to 8 years in storage for standard LWR plutonium). The cost of removing Am-241 chemically has been estimated at about $20 per gram of plutonium.[27]

The costs of storage are small compared to the value of the electricity that was generated as the plutonium was produced. At a storage cost of $1.5 per gram per year, each year of storage would add only one-tenth of one percent to the delivered cost of the electricity.* Nor are the costs high in comparison to the amounts that industrialized nations are accustomed to spend on national security. At $1.5 per gram per year, the storage of 50 tonnes of plutonium would cost $75 million per year.

Nevertheless, utilities tend to make decisions on the basis of small marginal costs. Even where there is a significant economic penalty to the utility for using plutonium in light-water reactors, cumulative plutonium storage costs will eventually exceed whatever economic penalty is incurred.† As long as utilities believe that the only reliable way to get rid of separated plutonium is in MOX fuel, plutonium storage costs will push them to recycle. If this is viewed as undesirable, governments will have to make available alternative methods of plutonium storage and disposal. They will also have to offer incentives to store surplus plutonium for an interim period, or impose their preferred approach by regulation.

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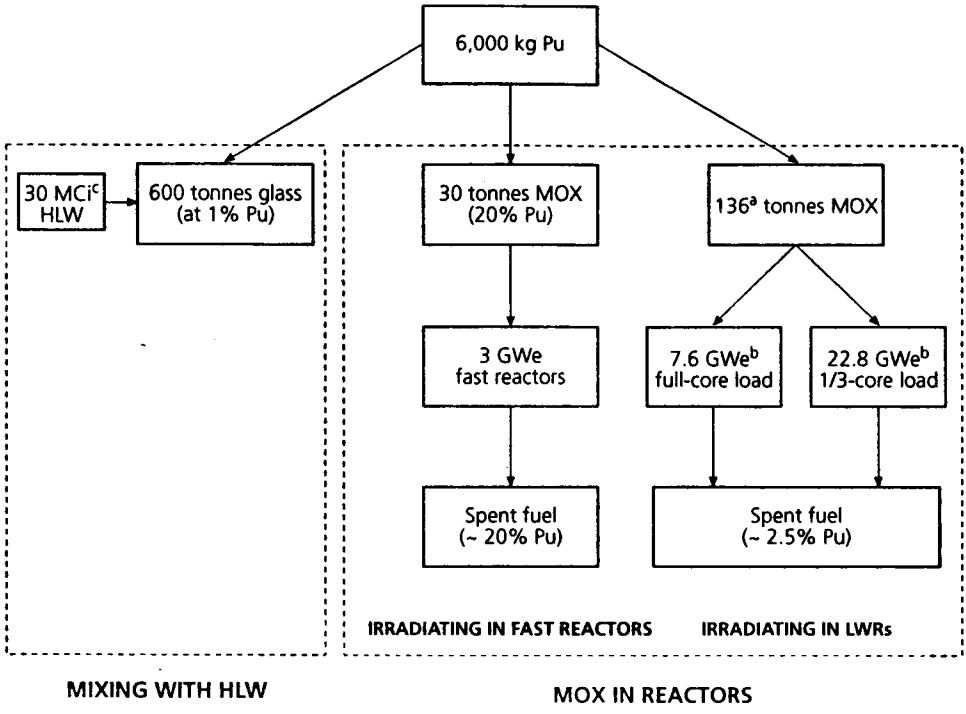
* For a fuel burnup of 33 megawatt-days per kilogram of heavy metal (MWd kg HM^{-1}) and a heat-to-electricity conversion ratio of one-third, approximately 260,000 kilowatt-hours (kWh) of electric energy would be generated from each kilogram of fuel and nine grams of plutonium would be produced. Thus about 30,000 kWh of electric energy would be generated for each gram of plutonium. At an electricity price of $0.05 per kWh, each gram of plutonium would be associated with $1,500 worth of power.

† As is shown in the next section, the mid-range cost penalty for recycling plutonium in thermal reactors is $5 to 12 per gram. At a storage cost of $1–2 per gram per year, plutonium storage costs would exceed this penalty within a few years.



a. Assuming 4.4% Pu in MOX.
b. Assuming 0.7 capacity factor, 0.33 thermal efficiency, 43 MWd kg^{-1} burnup.
c. Assuming 0.05 Ci g^{-1} high-level waste (HLW) in glass.

Figure 1: Alternative paths to dispose of 6,000 kilograms of weapons-grade plutonium per year.

ALTERNATIVE DISPOSITION OPTIONS FOR SEPARATED PLUTONIUM

Separated plutonium in retrievable storage could support a national nuclear weapon program at short notice in both weapon and non-weapon states. As explained above, indefinite storage would also be costly. Therefore, consideration should be given to alternative methods of making separated plutonium less accessible for weapons use. The principal alternatives we consider are (see figure 1):

- *MOX in Unmodified LWRs*: Irradiation of plutonium in commercial unmodified LWRs.
- *Full-Core MOX*: Irradiation of plutonium in specialized reactors.

- *Anti-Reprocessing*: Incorporating plutonium into a matrix with high-level waste.

In addition, we will briefly discuss other options that have been proposed: immobilization in materials without mixing in high-level waste, glassification with rock using underground nuclear explosions, space disposal, and plutonium fissioning using accelerator-driven subcritical assemblies.

In considering the various options, we should not exaggerate the implications for nuclear power. Four hundred tonnes of plutonium recycled in LWRs would fuel the current world LWR capacity (about 280 gigawatts-electric [GWe]) for about 1.5 years.[28] By contrast, a world LWR capacity of 400 GWe operating on a once-through fuel cycle could be sustained by known, conventional low-cost uranium (less than $130 per kilogram U) for nearly 100 years.[29] A capacity four times larger could be supported for this period if speculative low-cost uranium resources were included.[30] Uranium is not a scarce commodity.

Conversion of separated plutonium to a less accessible form would also not preclude a potential long-term future role for plutonium-breeder reactors. If world nuclear capacity grows so large that breeders are needed, the plutonium required for breeder start-up inventories could be derived from stored LWR spent fuel. For example, at a world nuclear capacity of 2,000 GWe, the smallest capacity for which a shift to breeder reactors might be considered, LWRs operated on a once-through fuel cycle would discharge about 240 tonnes fissile plutonium *annually*.[31] On this scale, the fate of 300–400 tonnes of separated plutonium currently under discussion here does not loom very large.

MOX in Unmodified LWRs

Up until the mid 1970s, the primary justification for separating civil plutonium was to fuel fast-neutron plutonium-breeder reactors. Since then it has become increasingly evident that these reactors are expensive to build and difficult to operate. Commercialization is now considered decades away, even by the most hopeful. Today the only planned large-scale use of plutonium is as mixed-oxide fuel (MOX) in light water reactors (LWRs). As described in table 4 and in appendix A, recycling programs have been proposed in six countries: Belgium, France, Germany, Russia, Switzerland, and Japan. In the short-term, LWR-MOX is the only way in which plutonium can be converted to a less accessible form at a rate of ten tonnes or more per year.

There is no question that recycling plutonium in LWRs could effectively convert the plutonium to a more proliferation-resistant form. It would reduce the amount of fissile plutonium in the fuel by about 40 percent[32] and mix the

Table 4: Plutonium and uranium recycle policies in Europe and Japan.

Country	Plutonium recycle	Reprocessed uranium recycle	LWR-MOX program
Belgium	Yes?	?	Plan to load 2 reactors with MOX, beginning mid 1990s. MOX fabricator.
France	Yes	Yes[a]	First phase (8 reactors) to be loaded with MOX by 1993. 8 more reactors to be loaded by late 1990s. MOX fabricator.
Germany	Yes	Tested with MOX	Leader in MOX experience. Eighteen reactors to be loaded with MOX.[b] MOX fabricator.
Italy	No[c]	No	No operating reactors.
Japan	Yes	?	Demonstration program (2 reactors) planned 1994–1997. Commercial program due to start in 1995, rising to 12 LWRs loaded with MOX by 2003. MOX fabricator.
Netherlands	Yes[d]	No	MOX R&D at Dodewaard currently suspended.
Russia	Expected	Yes	Uranium separated from VVER fuel recycled in graphite-moderated reactors (RBMKs).
Spain	No	No	
Switzerland	Yes	?	Two reactors loaded with MOX.
United Kingdom	No	Yes	Planning to become an MOX fabricator.

a. Official French policy is to recycle uranium recovered by reprocessing spent fuel (U_{rec}), either by re-enriching the uranium, or by using the U_{rec} as the matrix for LWR-MOX manufacture. However, the low price of natural uranium has meant that the Electricité de France has shown little practical interest in uranium recycle.

b. At present only ten reactors have been awarded licenses to load MOX fuel. MOX has been loaded at seven reactors.

c. A small MOX test program ran at Garigliano in the 1970s. Most Italian separated plutonium has been used to fuel Superphénix.

d. A small MOX test program ran at Dodewaard in the 1970s and 1980s. Dutch separated plutonium has been used in the Superphénix and Kalkar fast-reactor cores.

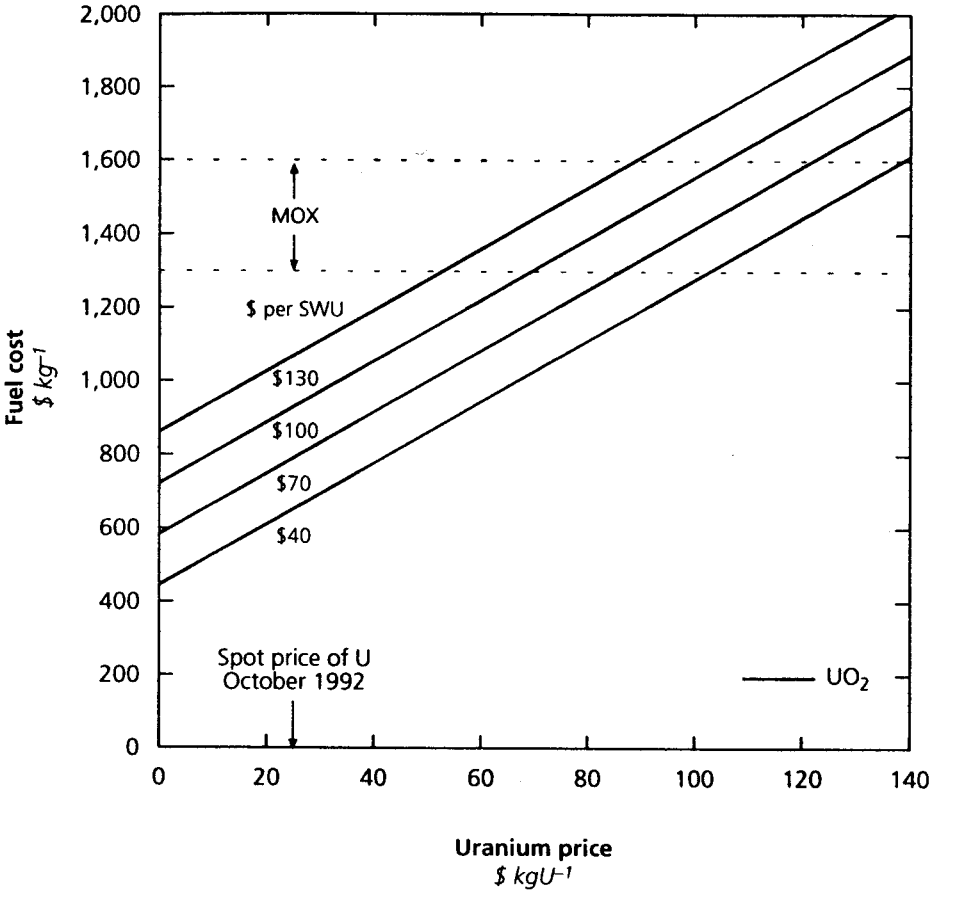


Figure 2: Sensitivity of fuel costs to uranium price, and SWU costs, using free plutonium. Assumed burnup is 43 MWd kg^{-1}. UO_2 fabrication cost is $200 per kilogram; UF_6 conversion cost is $7 per kilogram of uranium fuel; 0.3% tails assay.[35]

plutonium with intensely radioactive fission products in the spent fuel. Irradiation in LWRs will also convert "weapons-grade" plutonium (< 6 percent Pu-240) to "reactor-grade" (> 19 percent Pu-240). Reactor-grade material is less desirable for weapons use because of its higher output of spontaneous fission neutrons and radioactive decay heat (see table 3.)

However, commercial recycling of plutonium raises troubling economic and security issues. As shown in figure 2, even if plutonium is considered a "free good" (so none of the costs of reprocessing are charged to the cost of the MOX fuel), a utility using MOX will incur a cost penalty of $200–500 per kilogram of MOX, assuming a uranium price of $40 per kilogram. The cost differential is based on a cost of a kilogram of low-enriched uranium (LEU) fuel of

about $1,100 and a cost range for a comparable kilogram of MOX fuel of $1,300–1,600. (See appendix B for details.) This cost differential implies a cost of recycling 100 tonnes of weapons-grade plutonium in MOX of about one-half to one billion dollars.

More importantly, plutonium recycling will involve a great increase in safeguards and physical security requirements to protect against the diversion of plutonium. The director of the Euratom Safeguards Directorate has stated that inspection of large reprocessing and MOX fabrication facilities will require a "quantum leap" in inspection effort.[33] A large reprocessing plant is estimated to require 2,000 man-days of inspection time per year (two to three inspectors present around the clock), or about one-fourth the total inspection effort of the IAEA today. Extra inspection effort and physical security would also have to be extended to reactors using MOX fuel. Whether either Euratom, the IAEA, or national security forces will be given the resources to take on these new tasks without diverting resources from other essential safeguards activities remains to be seen.

Even with improved safeguards, given the vast throughputs of material and intrinsic measurement uncertainties, material accountancy at plutonium reprocessing and MOX fabrication plants will probably be inadequate to detect the diversion of one or more fission bomb equivalents of plutonium per year per plant.[34] And, even if the thousands of kilograms of plutonium produced, fabricated, and transported annually in a full-blown recycling economy could be completely secured, the commercial use of plutonium in Europe and Japan would make it more difficult to restrict plutonium separation and use in other regions that are currently of more proliferation concern.

Furthermore, even apart from economic and security concerns, commercial recycling appears currently to be incapable of absorbing plutonium at the rate at which it is to be separated. In unmodified LWRs, MOX use is generally restricted to one-third of a reactor core because of the different nuclear properties of plutonium compared with U-235. (Neutron absorption cross sections in plutonium fuel are higher than in uranium fuel, which decreases the effectiveness of the control rods.)

The annual loading of fissile plutonium for a 1-GWe LWR operating with one-third MOX fuel is about 0.26 tonnes.[36] This translates to 0.38 tonnes of reactor-grade plutonium or 0.28 tonnes of weapons-grade plutonium per year. If one-third core recycling were carried out in order to process 100 tonnes of weapons-grade plutonium released from dismantled CIS and US nuclear warheads, about 360 GWe-years of LWR capacity would be required. Over ten years, this would require over 10 percent of total current world LWR capacity disregarding the capacity needed to dispose of civil plutonium. In practice,

civil plutonium recycling will be the priority, and even here the rate of separation is likely to outpace use.[37] As shown in appendix A, expected MOX recycle capacity will only be able to process about one-half of the *civil* plutonium becoming available over the next decade. And countries driven to recycling because of their growing civil plutonium surpluses are therefore unlikely to accept plutonium released from CIS and US warheads.

Irradiation in Specialized Reactors

LWRs with Full Cores of MOX Fuel

One way to speed plutonium use in reactor fuel without increasing the number of reactors and sites involved would be to modify a set of reactors to accept a full-core of MOX fuel. To address the problem of the shorter mean free path of neutrons in plutonium fuel, the number of control rods would have to be increased. In some reactor designs there is space available in the cores. In others, fuel rods would have to be removed to make space available.

Another potential control problem that could arise with weapons-grade plutonium is related to that fact that the fraction of delayed neutrons among the fission neutrons produced by Pu-239 is only about one-third that for U-235. (Delayed neutrons are neutrons that are released by fission products on average about ten seconds after fission. About 0.64 percent of neutrons produced by the fission of U-235 are delayed.) If the neutron multiplication factor in a reactor is not larger than unity by an amount equal to this delayed neutron fraction, the time constant for power changes will be determined by the delayed neutrons and will be slow enough to be managed by mechanically operated control rods. Because Pu-239 fission has a lower delayed-neutron fraction, this margin of safety in a reactor fueled with weapons-grade plutonium will be lower than for one fueled with U-235, and the control system required to keep the neutron multiplication rate within the required range would have to work faster.*

If dedicated LWRs designed for full-core MOX could be installed, about 0.78 tonnes of weapons-grade plutonium could be irradiated per GWe-year. One hundred tonnes of weapons plutonium could be irradiated in about 120 GWe-years. This would require just 12 large LWRs operating for some 10 years. The same approach could be used to deal with surplus civil plutonium. The throughput of plutonium in LWRs could be further increased if the LWRs

* This problem is somewhat reduced for reactor-grade plutonium, where the higher delayed-neutron fraction of Pu-241 partially compensates for the smaller one for Pu-239 fissions.

were operated at lower burnups than commercially optimal. For example, at a burnup of 20 MWd kg^{-1} instead of 43 MWd kg^{-1}, about 1.5 tonnes of plutonium could be irradiated annually in an LWR employing full-core MOX. (Even at this relatively low burnup, the Pu-240 content in the spent fuel would be over 24 percent.)

Fast-Neutron Reactors

A fast-neutron reactor operating on a once-through MOX fuel cycle could process still larger amounts of plutonium than an LWR of equal power output because the percentage of fissile plutonium in their fuel is about four times higher for the same burnup. For example, a 1-GWe fast-neutron reactor (operating with a 40 percent thermal efficiency, 70 percent capacity factor, fuel burnup of 50 MWd kg^{-1}, and a plutonium fuel fraction of 20 percent), would have a throughput of plutonium of about 2.4 tonnes per year.

The isotopic mix of plutonium irradiated in fast reactors would not be altered as much as in LWRs, however. This is because the neutron capture-to-fission ratio of Pu-239 is much smaller for fast neutrons than for thermal neutrons and also because the fraction of fissile plutonium nuclei fissioned in fast-reactor fuel is lower than in LWRs operated at the same fuel "burnup." As a result, in spent MOX fuel that originally contained weapons-grade plutonium, the percentage of Pu-240 in the plutonium of fast-neutron reactor fuel after an irradiation of 50 MWd kg^{-1} would be only 12 percent, compared to 25 percent for LWR fuel at 53 MWd kg^{-1}.[38] This, along with the fact that fast-reactor MOX contains a higher proportion of plutonium (15 to 20 percent) than LWR-MOX (5 to 7 percent), would make spent fast-neutron reactor fuel more attractive as a plutonium "mine."

In any case, the world's fast-neutron reactor capacity is very small.[39] At the end of July 1992, following the decision by the French government not to re-licence the Superphénix reactor, world fast-reactor capacity stood at about 1.8 GWe. The future of the Russian fast reactors must also be deemed uncertain,* while the British Prototype Fast Reactor at Dounreay is scheduled to close in 1994. That would leave just two fast reactors with significant capacity, the Phénix in France and Monju in Japan, with a combined capacity of about 0.5 GWe.

New fast reactors could be constructed to irradiate plutonium on a once-through fuel cycle, but would require considerable modifications from current designs if the reactors were operated as plutonium burners rather than breed-

* The existing Russian and Kazakh fast reactors have been fuelled principally with enriched uranium rather than MOX.

Table 5: World vitrification facilities.

Country	Location	Facility	Operated	Throughput *kg glass per hour*	Waste form[a]	Capital cost *$10^6 1990*	Waste to be treated[b] *megacuries*	Mean waste loadings in glass *curies per gram*
Belgium	Mol	PAMELA	1985–1991	12–30[c]	BSG			
China	Beijing		1994–	27	BSG			
France	Marcoule	Piver	1969–1973	13	BSG			
	Marcoule	AVM	1978–	15	BSG		100	
	La Hague	R7	1989[d]–	3 · 30	BSG		550[e]	
	La Hague	T7	1992–	3 · 30	BSG	180		
India	Tarapur		early 1980s–	2 · 2	BSG			
	Trombay		1993–	2 · 2	BSG			
	Kalpakkam		late 1990s?	2 · 2	BSG			
Japan	Tokai		1992/3–	~30[f]	BSG	290		
Russia	Chelyabinsk-65		1987[g]–	2 · 45 (max)	PG		300	0.1
	Chelyabinsk-65		mid 1990s?	25-35	PG?			
UK	Sellafield	WVP	1991–	2 · 25	BSG	410	330	0.4
US	Hanford	DWVP	late 1990s?	100	BSG	1,400	190	0.018
	Savannah River	DWPF	1993–	103	BSG	2,100[h]	260	0.05
	West Valley		1996–	45	BSG			

a. BSG = borosilicate glass; PG = phosphate glass.

b. Sr-90, and Cs-137, as of 1990.

c. Marples reports that in its first two years of operation, PAMELA produced 86.4 tonnes of glass. Over six years of operation, 500 tonnes of glass were produced at PAMELA containing 12 megacuries of β/α activity. This gives a mean activity of 0.024 curies per gram, see M. Odell, "Vitrification—A World Review," *Nuclear Engineering International*, June 1992, p. 51; and J.A.C. Marples, "The Preparation, Properties, and Disposal of Vitrified High Level Waste from Nuclear Fuel Reprocessing," *Glass Technology* **29** (6) December 1988, p. 231.

d. M. Odell, (op. cit.,) pp. 51–52.

e. La Hague total.

f. Design throughput is set at 700 kilograms per day. Assuming round-the-clock operation of the melter during casting campaigns, this is a rate of 29 kilograms per hour.

g. The plant operated smoothly from for 13 months until 1988 when it failed, having produced about 160 tonnes of glass. It was re-opened in 1991. Source: T.B. Cochran and R.S. Norris, *Russian/Soviet Nuclear Warhead Production, Nuclear Weapons Databook*, (Washington DC: NRDC, NWD 92-1, 1992) pp. 40–41.

h. Additional supporting facilities, which DWPF cannot operate fully and reliably, will cost an additional $1.8 billion, making a total of $3.9 billion. Source: US General Accounting Office, *Nuclear Waste—Defense Waste Processing Facility—Cost, Schedule, and Technical Issues*, (Washington DC: GAO/RCED-92-183, June 1992.).

ers. In these circumstances, the uranium in the reactor blanket would be replaced by non-fertile material, such as stainless steel; and uranium in the core would also be replaced by non-fertile material (beryllium in some advanced designs).

Even if based on old designs, the use of fast reactors to burn weapons plutonium would appear to be a costly option. For example, the Japanese Science and Technology Agency has proposed to build a $4 billion 0.8 GWe fast breeder in Russia capable of processing two tonnes of weapons plutonium per year.[40] If this total cost was attributed solely to plutonium burnup with the cost of the reactor depreciated over a 30-year period, the cost of irradiating 60 tonnes of plutonium over a 30-year period would be over $7 billion.[41]

If the reactor produced electricity about one half of the costs of plutonium irradiation could be offset, but they would still be much higher than for the other alternatives considered in this paper.[42, *]

Because of the long delays required for their design and construction and their high cost, plutonium irradiation with fast reactors, therefore, seems unlikely on a large scale.

Conclusion

If separated plutonium is to be processed into spent fuel through irradiation in reactors within the next two decades, the only practical option appears to be conventional light-water reactors. This is the option that the nuclear industry has chosen for separated civilian plutonium. However, the industry's approach is to use unmodified LWRs with cores restricted to containing one-third MOX fuel. If the MOX-fuel option is pursued on a large scale for a prolonged period of time, we believe that it would be easier to safeguard the fresh plutonium fuel if MOX-fuel use were concentrated in a smaller number of dedicated LWRs with their control systems adapted for 100 percent MOX loadings. Below, however, we examine an alternative approach to plutonium disposal that may be both less costly and easier to safeguard.

* This assumes that the fabrication costs of MOX fuel for the fast reactor would be roughly comparable to that for an LWR, even though the plutonium content of the MOX for a fast reactor would be about four times greater; if the fast-reactor fuel costs were significantly higher than the LWR costs, the fast-reactor penalty would, of course, be even greater.

Anti-Reprocessing

Glassification with High-Level Waste

Plutonium in spent fuel is in a highly proliferation-resistant form by virtue of being intimately mixed with highly radioactive fission products. But this mixing can be accomplished without irradiating plutonium in reactors, simply by mixing the plutonium with some of the huge quantities of fission products that have been produced as a byproduct of past reprocessing. Because the mixing of the plutonium with the fission products is the opposite of what is done in reprocessing, we call this approach "anti-reprocessing." (See figure 3.)

The safest time to mix plutonium in with high-level waste (HLW) is when the HLW is being incorporated into a solid waste form for final disposal. Currently, glass if the preferred matrix. HLW glassification is being carried out on a significant scale at La Hague and Marcoule in France; Chelyabinsk-65 in Russia; and Sellafield in the UK. Additional facilities are scheduled to open during the 1990s at Tokai, Japan; and Savannah River, West Valley and Hanford in the US (see table 5). As a benchmark of an acceptable concentration of plutonium in glass we take the range of concentrations in LWR spent fuel—from about one percent by weight in spent low-enriched uranium fuel up to five percent in spent LWR-MOX fuel.[43]

The advantages of glassification with HLW are:

- The plutonium would be made relatively inaccessible in a leach-resistant, radioactive matrix.
- The disposal of the plutonium could be accomplished at little incremental cost.
- The vitrification of plutonium would be relatively simple and carried out at a small number of sites, thus minimizing safeguards problems.

In the glassification process, the liquid HLW is converted to glass in a series of steps: the liquid (nitric acid solution) is evaporated; the nitrates of the waste compounds are calcined to oxides; the resulting waste oxides are melted with nonradioactive oxides to form a glass; and the glass is cast into containers for storage. (Some of these steps may be combined.) At all vitrification plants except Chelyabinsk-65, borosilicate glass (BSG) is the glass of choice.[44] Since the boron in this glass is a neutron absorber, it would insure against criticality accidents.

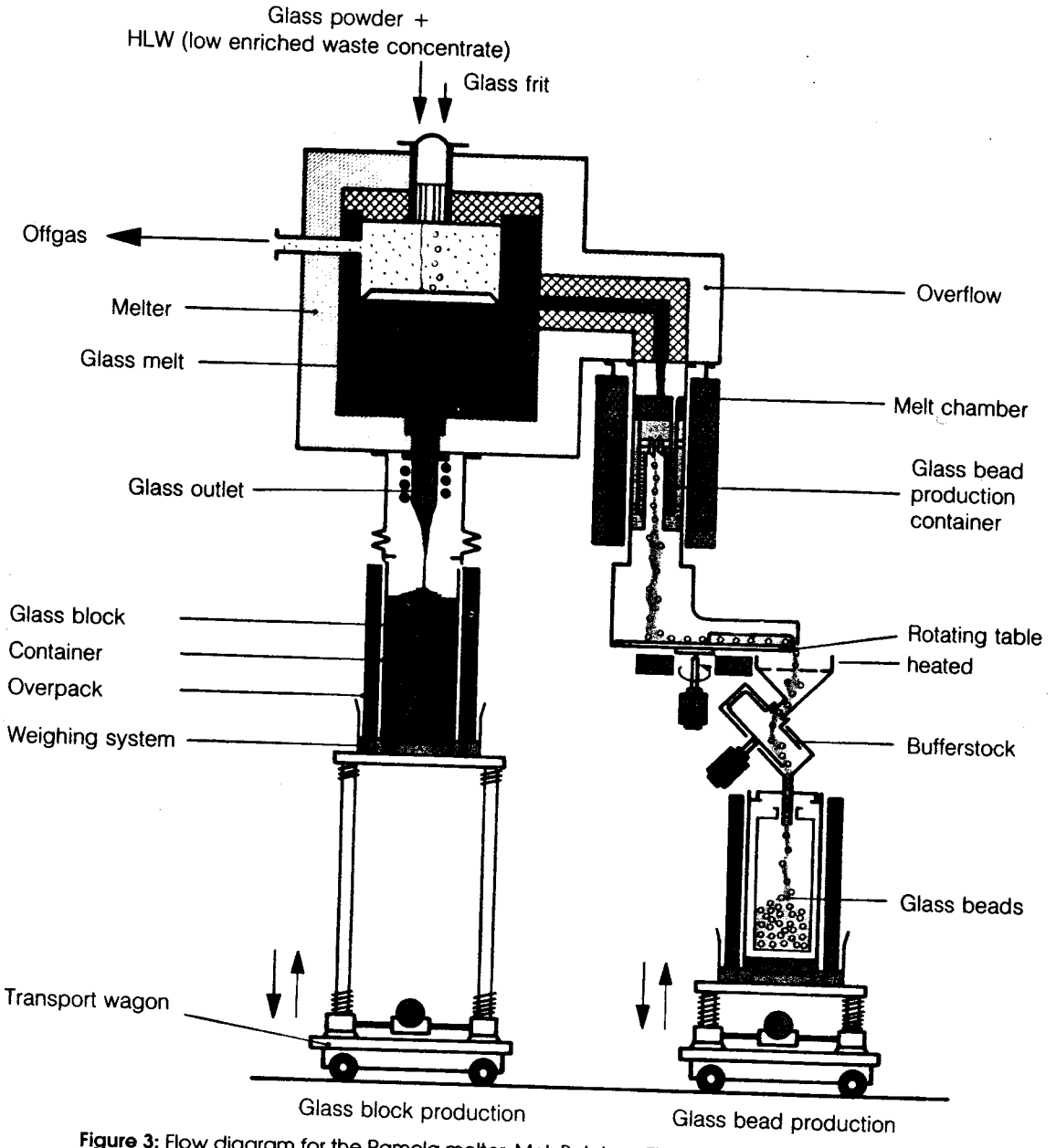


Figure 3: Flow diagram for the Pamela melter, Mol, Belgium. The Pamela vitrification plant operated between October 1985 and September 1991, when it was closed down for refurbishment. During six years of operation two ceramic melters vitrified just over 900 m^3 of HLW, with β/γ activity of 12 megacuries and α activity of 41 kilocuries, in almost 500 tonnes of glass. The vitrified product is stored in about 2,200 stainless steel drums containing about 100 tonnes of waste oxides at a dedicated waste storage facility also located at Mol.

Technical Considerations

A preliminary investigation has revealed no technical obstacles to inclusion of plutonium in HLW glass at plutonium concentrations of up to a few percent with regard to plutonium solubility in the glass, the effects of radiation and heat on the long-term integrity of the glass, or plutonium criticality.

The first assessment of this approach was carried out by a group at Pacific Northwest Laboratory (PNL).[45] The PNL group believes that there should be no difficulty in dissolving plutonium in borosilicate glasses at levels up to 2 percent by weight plutonium.[46] A German experiment found a solubility of 4.5 percent, even allowing for the addition of up to 20 percent by weight of fission product oxides.[47] In contrast, a few Russian experiments using both borosilicate and phosphate glasses found plutonium solubility limits as low as 0.1–0.3 percent for both glasses.[48] A possible reconciliation of these very different results based on the different chemical compositions of the German and Russian glasses is put forward in appendix C.

With regard to potential radiation damage to the HLW glass over time, the alpha-decays of plutonium are potentially more damaging than the beta- and gamma-decays of fission products because of their more energetic nuclear recoils and because alpha particles are converted into helium gas in the glass matrix. However, it appears that, beyond concentrations of 0.1 percent alpha-emitters by weight (about one quarter the level already in vitrified HLW), alpha-radiation damage saturates and does not seriously degrade the integrity of most glasses.[49] The helium is expected to remain trapped in the glass matrix at ambient temperatures and therefore should not cause gas-pressure buildup.[50]

Decay heat produces stresses associated with the difference between the centerline and the surface temperature of the glass block. Waste-glass forms are relatively standardized as cylinders with radii of about 20 centimeters. Depending upon whether the plutonium was weapon- or reactor-grade, one percent plutonium would increase the decay heat of a tonne of high-level waste by between 25 and 250 watts per tonne (see table 3). The rate of heat loss that the glass would experience if it were cooled at a rate of 1°C per hour would be about 200 watts per tonne.[51] Actual rates of cooling after casting exceed 15°C per hour.[52] The added thermal stresses due to plutonium decay heat could therefore easily be avoided if necessary by a slight decrease in the cooling rate.

Criticality also does not appear to pose a problem. Since a fuel containing over two percent fissile plutonium is necessary to drive a light-water reactor core critical in an optimal geometry, criticality would be unlikely to arise at a concentration below two percent plutonium by weight in any material, even if

the material crumbled and mixed with water. As indicated above, the neutron poison, boron, in borosilicate glass would prevent criticality in any case.

Quantities of Glass

In this section we consider on a country-by-country basis the amounts of glassified HLW expected to be produced from existing liquid HLW and compare these with the quantities of glass required to absorb surplus plutonium at different concentrations. We count only the activity of Sr-90 and Cs-137 because these radioisotopes, which have half-lives of about 30 years, dominate the radioactivity of the high-level waste for hundreds of years starting about five years after fission.[53]

♦ *United States*

As shown in table 5, most of the HLW from US plutonium and tritium production is located at the Savannah River, South Carolina (260 megacuries of Sr-90 plus Cs-137 in liquid HLW) and the Hanford, Washington (190 megacuries) sites.[54]

At the Savannah River site, some 4,000–5,000 tonnes of HLW is scheduled to be glassified beginning in 1993 at a plant that is currently projected to cost about $4 billion.[55] The glass is to have a mean concentration of Sr-90 plus Cs-137 of about 0.05 curies gm^{-1}.[56] (For comparison, the corresponding concentration in spent fuel is about 0.2 curies gm^{-1}.[57]) At a capacity factor of 50 percent, the output of the glassification plant would be 440 tonnes of HLW glass per year, enough to hold 4.4 tonnes of plutonium at a concentration of one percent. In ten years, when glassification of the Savannah HLW is due to be completed, 40–50 tonnes of plutonium could have been glassified—proportionately more at concentrations higher than one percent. Currently, the actual start-up date and performance of the facility are quite uncertain, due to a number of technical problems.[58]

At the Hanford site, construction of the planned HLW-glassification plant has not yet begun. The current plan is to incorporate HLW into 25,000 tonnes of glass, with concentrations of Sr-90 plus Cs-137 ranging from 0.004–0.03 curies gm^{-1} and averaging about 0.018 curies gm^{-1}.[59] The planned production capacity at the Hanford glassification plant is the same as at Savannah River. At the same output as at Savannah River—440 tonnes per year—it would take over 50 years to glassify the Hanford HLW at the proposed concentrations.

♦ *Russia/CIS*

Russian/Soviet production of plutonium and tritium for weapons has taken place at three sites in Russia: Chelyabinsk-65, Tomsk-7, and Krasnoyarsk-26.[60] High-level waste is being vitrified only at the Mayak combine at Chel-

yabinsk-65. At the other sites HLW has routinely been injected into deep wells in liquid form.

As of 1990, about 300 megacuries of Sr-90 and Cs-137 were stored in the HLW tanks at the Mayak facility in Chelyabinsk-65.[61] Since 1987, an HLW-glassification plant has been in operation at Chelyabinsk-65 with a nominal production capacity of about one tonne of HLW glass per day. By the beginning of March 1992, approximately 460 tonnes of HLW glass had been produced containing a total of about 45 megacuries at a concentration of about 0.1 curies gm^{-1}.[62] At this waste loading, the current HLW inventory at Chelyabinsk-65 would be incorporated into a total of 3,000 tonnes of glassified HLW. This much glass could contain from 30 to 120 tonnes of plutonium at loadings of one to four percent by weight.

Reprocessing continues at Mayak at a rate of about 250 tonnes of fuel per year, leading to a separation of approximately 2.5 tonnes of plutonium per year. Assuming a mean burnup of 33 MWd $kgHM^{-1}$, the on-going reprocessing would add about 50 megacuries of Sr-90 and Cs-137 to the HLW each year—corresponding to another 500 tonnes of HLW glass per year at past waste loadings.[63] If the annual plutonium increment now being separated is later mixed back into the associated HLW as the waste is vitrified, it could be incorporated into the waste at a concentration of about 0.5 percent. It should be noted, however, that a stock of about 30 tonnes of separated civilian plutonium had accumulated at Chelyabinsk-65 by mid 1992.[64] If this plutonium is incorporated into the 3,000 tonnes of glass expected to be produced from already existing HLW, it would by itself raise the plutonium concentration in the glass to about one percent.

♦ *United Kingdom*

At the end of 1992, Britain will have reprocessed Magnox[65] fuel containing about 83 tonnes of fission products at Sellafield.[66] This would result in an inventory of about 330 megacuries of Sr-90 and Cs-137 (taking into account decay).

Britain and France are both expected to produce glass with waste loadings of about 0.4 curies gm^{-1}.[67] At this concentration about 825 tonnes of glass could incorporate 330 megacuries. One industry source claims a somewhat lower waste loading of 0.28 curies gm^{-1}.[68] At this loading, about 1,200 tonnes of HLW glass would be produced at the British plant from existing waste. Twelve hundred tonnes of vitrified HLW at a loading of one percent plutonium would accommodate 12 tonnes of plutonium—considerably more than the UK weapons inventory (about three tonnes),[69] but much less than the 37 tonnes of civilian plutonium now stored at Sellafield. As in the Russian case, either

higher concentrations would be required or a more dilute glass.

♦ *France*

At the end of 1992, France will have reprocessed at Marcoule and La Hague Magnox and LWR fuel containing a total of about 125 tonnes of fission products.[70] Allowing for decay, this would correspond to an accumulated HLW inventory containing about 550 megacuries of Sr-90 plus Cs-137 at La Hague and 100 megacuries at Marcoule. Assuming 0.4 curies gm^{-1} of glass, about 1,600 tonnes of glass would be produced out of this waste. At a one percent concentration of plutonium in the glass, this would accommodate about 16 tonnes of separated plutonium, compared to the total estimated French weapons inventory of about six tonnes. Only a limited amount of civil plutonium is currently stockpiled at French reprocessing sites.[71]

♦ *Summary*

Although the HLW inventories at the British and French reprocessing plants are comparable to the US HLW inventory (measured in curies) the activity concentrations in the glass being produced at those sites is higher than is planned for US weapons-production facilities. This is mainly because of the more complex chemistry of high-level wastes stored at Hanford and Savannah River. It is also important to note that, while European HLW-glassification plants are all operating, the US facilities are still some years away from completion. Plant adaptations are therefore probably more easily accomplished in the US.

Safeguards and Security

Because of assay inaccuracies arising from inhomogeneity and other characteristics of the glassified plutonium-HLW product, safeguards for plutonium glassification would have to rely heavily on containment and surveillance. The same is true for a reprocessing plant because of uncertainties in the amount of plutonium in the incoming spent fuel. The HLW-glassification process itself would be much simpler than the plutonium separation process at a reprocessing plant.

The glassified waste form would place the plutonium in a relatively inaccessible radioactive matrix at low concentrations. In principle, the plutonium could be re-extracted, but this would probably be a more difficult and costly undertaking than extracting plutonium from spent reactor fuel. To remove the plutonium from the silicate matrix, it would be necessary to use a combination of hydrofluoric acid and nitric acid to make the plutonium soluble. This is a slow process that requires large amounts of acid and special coating materials (e.g., teflon, tantalum, or platinum) for dissolving vessels and piping.[72]

Costs

If plutonium is encapsulated in HLW glass at existing or planned vitrification facilities, only the marginal costs of plutonium processing should be attributed to plutonium disposal. The PNL analysis estimates a cost of just $15 million to process 50 tonnes of plutonium metal to oxide and into glass. There would be additional charges for transporting and disposing of the waste in a repository.[73] In contrast, an analysis done at the DWPF vitrification facility at Savannah River suggests that adaptation of that facility to handle plutonium would add up to $100 million (five percent) to its capital cost and up to 30 percent to its operating costs. This translates to an increase in total costs of about $30 million per year.[74] Assuming the DWPF could handle about four tonnes of plutonium per year at a loading of one percent, the marginal unit cost of handling plutonium would be about $7.5 million per tonne. However, even at this high cost, plutonium vitrification would still cost less than the likely subsidy required for its fabrication into MOX fuel.

Matrices without HLW

What about the possibility of imbedding plutonium in specially designed materials without high-level waste? Dilution in large blocks of such material would make the plutonium hard to remove clandestinely and the material could be designed to minimize leaching—a highly desirable characteristic, given the long half-lives of some of the plutonium isotopes. The absence of gamma-emitting fission products would probably also make production of the wasteform less costly. Instead of conducting all production and maintenance operations behind heavy shielding, glovebox-type arrangements would be sufficient. On the other hand, the absence of intense gamma emission would also tend make it less costly to recover the plutonium from this waste form than from spent fuel or glass containing fission products. However, even in the absence of fission products, recovery of plutonium from silicate glass would be hazardous and costly, even prior to final disposal in geological repositories. In all cases, the effectiveness of safeguards would be enhanced by the fact that the plutonium would be diluted in large, heavy blocks, not concentrated in fluids or powders.

Glassification by Underground Nuclear Explosions

One of Russia's nuclear-warhead design laboratories, Arzamas-16, has proposed that the plutonium components of nuclear warheads be glassified *in situ* with underground nuclear explosions. They estimate that 5,000 plutonium components containing 20,000 kilograms of plutonium could be destroyed by a

single 100 kiloton explosion producing a mass of glass with about 0.01 percent by weight plutonium.[76]

The cost of such an explosion would be comparatively low—on the order of a hundred million dollars based on the cost of US tunnel tests—corresponding to a cost of a few million dollars per tonne of plutonium processed.[77] However, the effects of the explosion might compromise the long-term isolation of the glass from ground water, and the long-lived fission product content of the glass would be quite low—less than 0.2 microcuries of Sr-90 and Cs-137 per gram of glass—easing the task of would-be plutonium miners.[78] Finally, a renewal of interest in "peaceful nuclear explosives" would make the effort to achieve a Comprehensive Nuclear Test Ban more difficult.

Other Disposal Options

The alternatives for disposition considered above would incorporate plutonium into more proliferation-resistant forms, but would not get rid of the plutonium entirely. Possible measures to do so include space disposal and the *complete* fissioning (or "transmutation") of the plutonium in special reactors or accelerators.

Space Disposal

The most recent version of this proposal is by Theodore Taylor, who envisions chemically-propelled heavy-lift vehicles carrying plutonium-containing payloads into high earth orbit. The payload-packages would be designed to survive intact the highest-possible velocity impact with the Earth, or an explosion of the rocket. Once in high-earth orbit, a solar-powered tug would transfer the package into an orbit from which it would fall directly into the sun.[79]

Fission in Accelerators

Because of the continuing worldwide impasse over the selection of sites for geological disposal of spent fuel and glassified high-level waste, there has been a renewal of interest in the past few years in the idea of separating plutonium and other long-lived radionuclides out of spent fuel and transmuting them into shorter-lived species.[80] This approach is often called "partitioning and transmutation" (P-T). In the case of the transuranic elements (neptunium, plutonium, americium and curium) transmutation would be done by fission. This fissioning could be accomplished in fast-neutron reactors but other approaches have been explored theoretically as well, including fissioning by spallation neutrons from accelerator-produced high-energy protons colliding with heavy-element targets or by high-energy X-rays from accelerator-pro-

duced high-energy electrons.

At present there are four accelerator P-T projects under investigation—two in the US (at the Los Alamos National Laboratory and at Brookhaven National Laboratory) and two in Japan. Three of the concepts employ proton accelerators[81]; one concept employs an electron accelerator.[82] All are at a very early stage of development, and have so far been unable to attract major funding. The US effort has a total budget for fiscal year 1992 of $2 million.

In view of the expected high cost, it is hard to imagine that accelerator fissioning will be developed for the sole purpose of fissioning already-separated plutonium. Such a solution is only likely if, in the long term, widescale P-T programs are instituted for dealing comprehensively with radioactive wastes. The fissioning of surplus plutonium stocks might be piggybacked onto such schemes. At present however, the costs of P-T are estimated as being some three times higher than direct disposal of spent fuel to a repository, and the reduced health risks over the long term are still seen as marginal.[83]

Summary

In general, both these schemes appear technologically daunting. Beyond this, while the concept of getting rid of the plutonium altogether rather than incorporating it into spent fuel or an equivalent matrix seems attractive, it probably could only be justified if most of the world's spent fuel was to be reprocessed. Currently, it appears likely that about 75 percent of plutonium discharged from civilian power reactors through the year 2000 will remain unseparated in spent fuel (see appendix A). As long as this is the case, it appears pointless to develop exotic techniques to treat the remaining 25 percent.

CONCLUSIONS

Maintaining control over separated plutonium remains as important as ever. The most critical near-term tasks are to ensure that all separated plutonium—both weapons-grade and reactor-grade—is stored securely and under international safeguards.

A second important near-term task is to accelerate the phaseout of plutonium separation—either for weapons or commercial use. With the beginning of large-scale dismantlement of warheads, further production of plutonium for weapons makes little sense. The need for commercial reprocessing has also disappeared.

For the longer term, this paper has proposed alternatives to the conven-

tional approach of disposing of separated plutonium in the fuel of unmodified LWRs. The principal alternatives are:

- Modifying a set of LWRs to be capable of handling full MOX cores.
- Mixing the plutonium into high-level waste as it is glassified.

Both of these alternatives would reduce the number of sites to which separated plutonium or fresh MOX fuel was distributed. Full-core MOX LWRs would reduce by two-thirds the number of reactors to which MOX would have to be delivered. Glassification would confine all plutonium handling to a few facilities, and would involve much less handling of plutonium than at a MOX fuel-fabrication facility. Moreover, it would produce a waste form at least as resistant to clandestine plutonium "mining" as spent LWR-MOX fuel.

The principal disadvantage of the HLW-glass route relative to the MOX route is that, in the case of weapons-grade plutonium, plutonium would remain weapons-grade, while, in spent MOX fuel, it would have been converted to reactor-grade.

With respect to cost, the full-core MOX strategy is probably not much more expensive than the one-third-core MOX strategy. Core redesign and relicensing would be required but safeguards would be required at fewer reactor sites. Of all the options, however, blending plutonium into HLW glass at existing or planned facilities is probably the least costly—especially in the US, which has no established infrastructure for plutonium recycle—because existing or planned HLW-glassification facilities could be used. Table 6 summarizes very roughly the costs of the principal alternatives, disregarding development costs.

Large amounts of plutonium (either glassified or in MOX spent fuel) will ultimately have to be disposed of in geological repositories, as will unreprocessed spent LEU fuel. Although not all questions with regard to plutonium migration from geological repositories have been resolved,[84] it appears that, given a compatible geochemical environment, MOX spent fuel and high-level waste glass containing up to a few percent plutonium differ little from spent LEU fuel with respect to long-term safety.[85]

It is imperative that any strategy for disposing of plutonium be accompanied by the necessary resources for safeguards and physical security, without reducing the resources available for other important safeguards responsibilities.

Table 6: Estimated cost for disposition of 100 tonnes of weapons-grade plutonium.[a]

	$ billion
MOX (one-third core)[b]	~1
MOX (full-core)[b]	~1
Dedicated fast reactor[c]	~5
Glassification with HLW[d]	~0.1–0.75

a. Not including development costs of option.
b. In already deployed LWRs, assuming a cost-penalty of about $500 per kilogram of MOX compared to low-enriched fuel (see appendix B).
c. Assuming a fast reactor dedicated to plutonium conversion.
d. Assuming that plutonium is processed at existing or planned HLW vitrification facilities and that no additional glass is produced.

Appendix A: Present and Future Civil Plutonium Surpluses

Historic and projected plutonium production and consumption data are presented in this appendix as background for the figures used in the paper. All figures given are total plutonium. In general we have assumed recycled civil plutonium to be 75 percent fissile on average, except where otherwise stated.

Historic Plutonium Production and Separation from Power-Reactor Fuel

Table A-1 presents estimates of the amounts of plutonium discharged from power reactors by country, and the amounts of plutonium separated from reprocessed fuel.[86] By the end of 1990 about 120 tonnes of plutonium had been separated from the irradiated power-reactor fuel of 17 countries. However, this represented less than one-fifth of the total amount of plutonium that had been discharged from the world's reactors. Even in those countries with reprocessing policies, the cumulative fraction of plutonium separated was just one-third (119.1 tonnes separated out of 375.2 tonnes discharged).

Although there is due to be a major expansion in the rate of reprocessing over the coming decade, this will have little impact on the fraction of discharged plutonium which is separated. Under current reprocessing plans, about 190 tonnes of plutonium will be separated between 1991 and 2000. This means that by 2000 a cumulative fraction of 23 percent of discharged plutonium will have been separated. The proportion separated for those countries with reprocessing policies will have risen from one-third to two-fifths (314 tonnes out of 800 tonnes discharged). From these figures it is clear that storage, probably with eventual disposal in deep geologic repositories, is now the principal means of managing the world's spent fuel.

Historic Plutonium Use and Balances

Table A-2 indicates plutonium use through 1990. Separated power reactor plutonium has been used principally for fuel for fast reactors (37 tonnes) and thermal reactors (12 tonnes). Small quantities of plutonium separated from power-reactor fuel have been used for weapons, but these are not included in the table.

When the use of plutonium is compared with its separation it will be seen that some 60 percent of the material (over 70 tonnes) separated through 1990 has been placed in store, rather than used. The largest stockpiles exist at Sellafield in the United Kingdom (over 37 tonnes) and at Chelyabinsk in Russia (about 25 tonnes).

Future Plutonium Use

Estimating what future plutonium balances will be is difficult since it depends on the operation of plants not yet built and the implementation of new LWR-MOX programs. Table A-3 shows the capacities of currently operating and planned MOX-fuel fabrication plants. Tables A-4 and A-5 present estimates of "credible" scenarios of plutonium use, based on a variety of data set out in the tables.

MOX Recycle in LWRs

Recycle of plutonium in LWRs is being pursued or is proposed in five countries: Belgium, France, Germany, Japan and Switzerland. The rate at which these programs will use plutonium will be determined mainly by MOX fuel fabrication capacity and the number of reactors licensed to take the fuel. In the analysis presented in table A-4 account has been taken of these aspects. The table presents a scenario for plutonium use in LWR-MOX fuel for the decade 1991–2000. The assumptions used are explained in the footnotes.

Fast-Reactor MOX

During the 1990s it is planned to load plutonium into fast-neutron reactors and an Advanced Thermal Reactor (ATR) in four countries: France, Japan, the United Kingdom and Russia. Russian plans to build a fast-neutron reactor sited at Chelyabinsk-65 have been put on hold and we have not included this reactor in our analysis.

Plutonium irradiation in the five reactors that may be loaded with fresh fuel depends upon whether they operate or not. One of these reactors has not yet started operating (Monju), two are R&D facilities (Joyo and Fugen) and have a history of uneven operation, while Superphénix may be closed down for good. Only Phénix has a reasonable operating record, but even it has outstanding safety problems. Suggesting a scenario for plutonium use at these facilities is therefore fraught with uncertainty.

We have assumed the following scenario for reactor operation:

- The Prototype Fast Reactor (PFR) at Dounreay in the UK is shut-down in 1994.
- Phénix (France) operates until 2005 and Superphénix is permanently shut-down in 1992.

- Joyo and Fugen (Japan) operate until 2000 and Monju operates until 2017 at 60 percent capacity.
- BN350 and BN600 (Kazakhstan and Russia) continue to be fuelled with HEU, and the BN800 is not completed.

Table A-5 shows how much plutonium would be absorbed by these fast reactors under this scenario. Table A-6 then shows the projected world balance of separated plutonium in 2000.

Balances of Separated Plutonium Forecast for 2010

It is probable that surpluses of the magnitude estimated above will not be permitted to arise for a mixture of economic and political reasons. However, it is necessary to show that the time-frame we have chosen does not deliberately underestimate the prospective use of plutonium after 2000. If it were possible to suddenly increase the demand for the material after the turn of the century, the surpluses built up during the 1990s might seem less significant.

With so little certainty about the future of reprocessing, there is little gained in attempting a full-scale comparison of supply and demand here.[87] Instead we can estimate how long it would take to absorb the world stock of civil plutonium (neglecting for the moment the weapons material) which is projected to accumulate by 2000.

First, we assume that plutonium consumption by fast reactors during the decade 2001–2010 will be about the same as we have forecast for the previous decade—about 10 tonnes.* We further assume that the major thermal recycling programs will continue to be in France, Germany and Japan. If the MOX programs announced in these countries all go ahead (16-reactor program in France, 18-reactor program in Germany, 12-reactor program in Japan after 2000), and if we assume that all these reactors are operating with 43,000 MWd t^{-1} fuel burnup, these three countries could absorb a maximum of about 15 tonnes of plutonium per year in 2001–2010.[88] Therefore, even if there was a complete cessation of plutonium separation in 2000, it would still take over a decade to consume the accumulated world surplus of civil plutonium (about 180 tonnes according to our forecast). If surplus weapon plutonium were made available for commercial use (between 130 and 190 tonnes in total, by our estimate made above), then a further ten years of supply would be guaranteed. Under these conditions, by the year 2000, total plutonium surpluses would be sufficient to supply commercial uses for about 20 years.

* This assumes that the Demonstration Fast-Breeder Reactor and Demonstration Advanced Thermal Reactor planned in Japan will not be built.

Table A-1: Estimated plutonium discharged and separated from power-reactor fuel (total plutonium, tonnes).

	through 1990		1991–2000		through 2000
	Pu discharged	Pu separated	Pu discharged (est.)	Pu separated (est.)	Cumulative % of total Pu separated (est.)
Countries reprocessing fuel					
Belgium	11.5	1.2	10	4	24
Bulgaria, Czech., Finland,[a] GDR, Hungary	23.7	5?	38	0?	8
CIS	78.1	20?	78	15?	15?
France	82.2[b]	23.6[b]	105	45	38
Germany	38.8	15.8[c]	45[d]	34	60
India	3.5	0.1	10	2?	15?
Italy	5.6	2.6	<1	2	74
Japan	57.4	6.4	83	54	43
Netherlands	2.4	0.7	1	1	47
Spain	9.7[e]	0	17	1.5	6
Switzerland	9.2	1.1	7	9	63
United Kingdom	53.1	42.5	30	25	80
United States	176.0	1.5	180	0	< 0.5
Countries not reprocessing fuel					
Canada	53.6	0	60	0	
Republic of Korea	8.4	0	23	0	
Sweden	20.7	0[f]	22	0	
Taiwan	7.6	0	11	0	
Rest of the world[g]	12.1	0	16	0	
Total	**653.6**	**120.5**	**737**	**192.5**	**23**

a. The two TVO BWRs in Finland are counted under this category.
b. Includes 4.2 tonnes of plutonium discharged from the Spanish Vandellos 1 Magnox reactor supplied by France together with fuel under a "take back" arrangement under which France retains ownership of plutonium.
c. Includes some 0.5 tonnes total plutonium separated from Swedish spent fuel at La Hague and coming into German ownership under a swap arrangement in 1990.
d. This includes 230 tonnes of fuel due to be discharged from decommissioned reactors in the former GDR, containing an estimated 900 kilograms of plutonium.
e. This does not include Vandellos 1 plutonium discharges (see note a).
f. Some 57 tonnes of Swedish fuel have been reprocessed at La Hague, but the separated plutonium has been swapped with German utilities.
g. Includes Argentina, Brazil, China, Cuba, Mexico, Pakistan, Romania, South Africa and Yugoslavia.

Table A-2: Power reactor plutonium separation and use through 1990 (total plutonium, tonnes).

Country	Plutonium separated	Plutonium use: Fast reactors	Plutonium use: Thermal reactors	Plutonium balance
Belgium	1.2	0	0.3	0.9
Bulgaria, CIS, Czech., Finland, GDR, Hungary[a]	25	< 0.5		24.5
France	23.6[b]	12.6	5.8	5.2[c]
Germany	15.8	2.5	4.9	8.4
India	0.1	0.1		0
Italy	2.6	3.7[d]	0.1	−1.2[e]
Japan	6.4	4.5	0.02	1.9
Netherlands	0.7	0.2[f]	0.2	0.3
Switzerland	1.1	0	1.0	0.1
UK	42.5	5		37.5
US	1.5	6.7		−5.2[g]
Total	**120.5**	**35.7[h]**	**12.3**	**72.5**
		37.3[i]		70.9[j]

a. Reprocessed fuel in East Europe and Finland was owned by the Soviet Union and supplied to clients under "take back" arrangements.
b. Including some four tonnes of plutonium separated from Vandellos 1 fuel at Marcoule and La Hague, which is French property.
c. This is a provisional figure since French plutonium was used in the core for the (never operated) Kalkar (Germany) fast-neutron breeder reactor (about 600 kilograms total plutonium) and in the foreign-owned segment of the Superphénix core (about three tonnes of total plutonium).
d. Plutonium in 33 percent of Superphénix core owned by the Italian utility ENEL.
e. ENEL bought plutonium from a variety of sources to make up its proportion of the first Superphénix core.
f. Material leased for the Kalkar first-core.
g. We assume here that all of this material came from other countries' stocks of civil plutonium—notably Britain.
h. This does not include approximately 1.6 tonnes of plutonium used in the 16 percent segment of Superphénix owned by the multinational consortium SBK, and supplied from a variety of sources, including French magnox plutonium.
i. Adding 1.6 tonnes of unallocated Superphénix plutonium (see note h).
j. Subtracting 1.6 tonnes of unallocated Superphénix plutonium (see note i).

Table A-3: Plutonium fuel fabrication facilities.[a]

Country	Location/Facility	Operator	Operated	Fuel type	Capacity *t MOX per year*
Operating facilities					
France	Cadarache/ATPu	CEA	1970–1989	FBR	15
United Kingdom	Sellafield	BNF plc	1970–1989	FBR	4
Japan	Tokai/PFFF	PNC	1972–	FBR + ATR	1 (FBR) + 9 (ATR)
Belgium	Dessel/DEMOX P0	BN	1973–	FBR + LWR	35
Germany	Hanau/BEW1	Siemens	1974–1992?	FBR + LWR	25–30
Japan	Tokai/PFPF	PNC	1988–	FBR	5
Russia[b]	Chelyabinsk-65 "Granat" "Paket"	MinAtom RF	1988–	FBR	35–40 kg MOX
France	Cadarache/CFCa	Cogema	1990–	FBR + LWR	10 (FBR) + 15 (LWR)
Planned facilities					
Germany	Hanau/BEW2	Siemens	1992?–	LWR	80–120
Japan	Tokai/PFPF	PNC	1993/4	ATR	40
United Kingdom	Sellafield/MDF	BNFL	1993–	LWR	8
France	Marcoule/Melox	Cogema	1996–	LWR	115
Belgium	Dessel/DEMOX P1	BN	mid 1990s?	LWR	35
Russia[a]	Chelyabinsk-65	MinAtom RF	mid 1990s?	FBR	25–30
			late 1990s?	VVER	100–300?
United Kingdom	Sellafield/SMP	BNFL	late 1990s?	LWR	50–70
Japan	Rokkasho-mura	JNFS?	late 1990s?	LWR	100?

a. FRB = fast-breeder reactor; ATR = advanced thermal reactor; LWR = light-water reactor; VVER = light-water reactor (Russia); CEA = Commissariat a l'Énergie Atomique; ATPu = Atelier Trailement Plutonium; PFFF = Plutonium Fuel Fabrication Facility; DEMOX = Dessel MOX; BEW = Brennelementwerke; PFPF = Plutonium Fuel Production Facility; MDF = MOX Demonstration Facility; SMP = Sellafield MOX Plant. See table 2, note a, for explanation of remaining acronyms.

b. Data on Russian plants are from V.N. Solonin, "Utilization of Nuclear Materials Released as the Result of Nuclear Disarmament," paper given to the International Symposium on Conversion of Nuclear Weapons for Peaceful Purposes, Rome, 15–17 June 1992.

Table A-4: Credible scenario for plutonium consumption in LWRs, 1991–2000[a]

Country	MOX fuel matrix	MOX fuel burnup	MOX fuel enrichment[b]	No. of reactors loaded with MOX[c]	Total MOX fuel loaded	Total plutonium consumed
		MW t^{-1}	*% total Pu*		*tonnes*	*tonnes*
Belgium	Assume nat U	33,000	4.1	2	96[d]	3.9
France	Depleted U (0.225% U-235)	33,000	4.8	8 (by 1993)	200 (to 1995)[e]	9.6
		43,000	5.6	16 (by 1998)	310 (to 2000)[f]	17.4
Germany	Nat U	33,000	4.1	10[f]	210 (max)[g]	8.6
		43,000	4.9	10	400 (max)[h]	19.6
Japan	Assume nat U	33,000	4.1	2 (1995-1997) 4 (1998–2000)[i]	140	5.7
Switzerland	Assume nat U	33,000	4.1	2	140[j]	5.7
Total					**1,496**	**70.2**

a. Sources: M. Rome et al., "Plutonium Reload Experience in French Pressurized Water Reactors," *Nuclear Technology* **94**, April 1990, pp. 87–94; P. Schmiedel, "Recycling and its Implication for Uranium Demand: the German Perspective," paper given to the Annual Symposium of The Uranium Institute, London, 5–7 September 1990; K. Samejima, "The Use of Plutonium in Japan," (London: Uranium Institute, September 1990); H.J. Dibbert, "Strategien des Brennstoffkreislauf," *Atomwirtschaft*, February 1991, pp. 83–88; M. Hibbs, "German Utilities Bracing for MOX Fuel Cost Increases," *NuclearFuel*, 6 January 1992.

b. Assuming 33 GWd t^{-1} LWR plutonium extracted from spent fuel stored for 10 years, with a fissile content of 70 percent.

c. Based on "Current Status and Utility Perceptions on LWR MOX Fuel Recycling," *NUKEM Market Report*, March 1992, pp. 8–12.

d. Assuming one-third MOX reloads at Doel 3 and Tihange 2 beginning in 1994.

e. Before the Melox fabrication plant begins operation MOX reloads in France will be limited by the availability of MOX fabrication capacity at Dessel and Cadarache. Electricité de France holds contracts for annual production of about 40 tonnes MOX per year at these plants (25 tonnes at Dessel and 15 tonnes at Cadarache).

f. One-quarter core loading.

g. Recycle uranium has been used and plans also exist for the use of depleted uranium as an MOX matrix.

h. A total of 50 tonnes of MOX are assumed to be loaded between 1991 and 1993. From 1994 production is raised to 80 tonnes MOX per year, the maximum that can be loaded at reactors for which licenses to take the fuel are outstanding. Under existing reactor licences just 30 tonnes MOX could be loaded at German reactors.

i. We assume that the Japanese MOX program follows the plan laid out in Japan Atomic Energy Commission, *Nuclear Fuel Recycling in Japan*, report by the Advisory Committee on Nuclear Fuel Recycling, Tokyo, August 1991.

j. Assuming one-third MOX reloads at Beznau 1 and 2 and at Goesgen (14 tonnes MOX per year) throughout the 1990s.

Table A-5: Credible scenario for FBR-MOX and ATR-MOX plutonium consumption, 1991–2000.

Country	Reactor	Type	Quantity of fuel loaded	Quantity of total plutonium loaded
			tonnes MOX[a]	*tonnes*[b]
France	Phénix	FBR	17	3.9
	Superphénix	FBR	0	0[c]
Japan	Joyo	FBR	8[d]	1.0
	Fugen	ATR	90	1.7
	Monju	FBR	16	3.8
Russia/Kazakhstan	BN350	FBR	5	0
	BN600	FBR	5	0[e]
United Kingdom	PFR	FBR	0	0
United States	FFTF	FBR	0	0
Total			**141**	**10.4**

a. Assuming mean fuelling cycles of 28 months. Mean annual reloads are therefore 40 percent of core.
b. Mean fuel enrichments are taken from D. Albright, F. Berkhout, W. Walker, *World Inventory of Plutonium and Highly-Enriched Uranium* (Oxford: Oxford University Press, 1993) pp. 119–128.
c. Following the French government's decision not to permit the restart of Superphénix, July 1992.
d. Assuming annual full-core reload.
e. Some plutonium use has been planned at BN600, but in the absence of completed MOX fabrication capacity or clearly stated plans we do not attempt to estimate consumption.

Table A-6: Projection of separated plutonium balances in 2000 (total plutonium, tonnes).[a]

Country	Plutonium separated (1991-2000)	Plutonium use: Fast reactors (1991-2000)	Plutonium use: Thermal reactors (1991-2000)	Plutonium balance (1991-2000)	Cumulative plutonium balance[b]
Belgium	4	0	3.9	0.1	1
Bulgaria, Czech., Finland and Hungary	0?	0	0	0?	
CIS	15?	0	0	15?	40?
France	45	3.9	27	14.1	19.3
Germany	34	0	28.2	5.8	14.2
India	2?	0?	0	2?	2?
Italy	2	0	0	2	0.8
Japan	54	6.5	5.7	41.8	43.7
Netherlands	1	0	0	1	1.3
Spain	1.5	0	0	1.5	1.5
Switzerland	9	0	5.7	3.3	3.4
United Kingdom	25	0	0	25	62.5
United States	0	0	0	0	–5.2
Total	**192.5**	**10.4**	**70.5**	**111.6**	**184.5**

a. Figures have been left with four significant figures not to denote precision, but to show fully the results of the assumptions used here.
b. See table A-2.

Appendix B

The Economics of Plutonium Recycle

At current natural uranium and enrichment prices, the value of uranium recovered in reprocessing (U_{rec}), if recycled, is about \$60 per kilogram U_{rec}.[89] This compares with a cost of reprocessing of \$1,000–1,800 per kilogram heavy metal (i.e., the total weight of uranium, plutonium and other transuranic isotopes in the spent fuel). Therefore, the value of the recovered uranium offsets only a few percent of the cost of reprocessing. Since low-enriched uranium fuel contains about 0.9 percent plutonium, of which 70 percent is fissile isotopes, the remaining uncompensated cost of reprocessing amounts to between \$150 and \$280 per gram of fissile plutonium. Since a kilogram of MOX fuel contains 35 to 50 grams of fissile plutonium, the cost of the plutonium recovery, if charged to the MOX fuel, would contribute between about \$5,000 and \$14,000 per kilogram to the price of MOX fuel—or roughly 4 to 13 times the current price of low-enriched uranium fuel with the same energy value (see below).

Even when the costs of reprocessing are considered sunk (i.e., the plutonium is considered a "free good"), the high costs associated with the extra health and safeguards protections that must be taken in any facility processing plutonium make the *fabrication cost* of MOX fuel by itself higher than the *total* cost of low-enriched uranium fuel at today's prices for natural uranium and separative work. A simple comparison between the costs of LEU and MOX is illustrated by figure 2.[90] Assuming as a base case a natural uranium price of \$40 per kilogram of uranium (somewhat higher than the current long-term contract price)[91] and a price for enrichment of \$100 per separative-work unit (SWU),[92] the cost of LEU fuel (burnup 43 MWd kg^{-1}) can be read from the graph as about \$1,100 per kilogram HM. This converts to an undiscounted cost of 3.2 mills per KWh_e.[93] A realistic price for MOX fuel fabrication and delivery today is \$1,500 per kilogram MOX (4.4 mills per KWh_e).[94] At these prices, MOX would therefore be about one-third more expensive than LEU fuel, even disregarding the cost of separating the plutonium from spent fuel and the extra security costs for MOX recycling.

The price of uranium, which has been low for a number of years, is likely to remain so. Large reserves of uranium ore exist in many countries, while uranium from the CIS has also recently come onto world markets. The current spot-market price for uranium has stood at about \$20 per kilogram of uranium for over a year, while the secondary market price for separative work has fallen below \$70 per SWU.[95]

Siemens, which has just built a large (100 tonne heavy-metal per year throughput) MOX fuel fabrication plant in Germany, claims that it could build a duplicate plant for a capital cost of DM700 million (\$450 million, at the late 1992 exchange rate) that could produce competitively priced MOX fuel, (about \$1,000 per kilogram HM). Siemens has proposed that the West fund the construction of such a plant in Russia to process surplus CIS weapons plutonium.[96] On the other hand, the prices for MOX could be significantly greater than the charges currently quoted in Europe. For example, fuel fabrication prices are very sensitive to throughput. Contract charges of up to

$3,000 per kilogram MOX have been projected for the Siemens German plant if it operates significantly below capacity.[97] At this level, MOX would cost almost three times as much as the LEU described above.

Given the various uncertainties, we take a central range of the MOX-fuel LEU-fuel cost differences $200–500 per kilogram. With this cost difference and for MOX fuel designed for 43 megawatt-days burnup per kilogram heavy metal (MWd $kgHM^{-1}$) and containing about 45 grams of weapons-grade plutonium per kilogram[98] the cost of recycling 100 tonnes of weapons-grade plutonium in MOX (i.e., the premium to be paid for fuelling reactors with MOX rather than LEU) would range from $0.44 to $1.1 billion.

Appendix C: Plutonium: How Soluble in Glass Is It?

Very little information is available on the solubility of plutonium in glass, defined as the maximum concentration that can be loaded before the glass undergoes a transition from a homogeneous phase to a heterogeneous phase.[99] There are two main phenomena which can result from such a transition: devitrification, in which the formation of a crystalline precipitate occurs; and glass-glass phase separation (immiscibility), in which the melted glass separates into regions containing glasses of different compositions. The latter effect can cause heterogeneity on a wide range of different scales, so it sometimes can only be observed using high-resolution microscopy.

For the purposes of long-term storage of radioactive wastes, both types of transition must be avoided. Crystallization is the more immediate problem, causing the glass matrix to be degraded at once, but phase separation, even at small scales, is a sign of potential future instability of the glass. For instance, the presence of silica-depleted glass regions would provide sites where leaching by water would be facilitated. Thus typically the necessity of preventing crystallization places one constraint on the concentration of non-glass forming ions like plutonium, and avoidance of phase separation imposes additional requirements.

These phenomena are sensitive to a wide range of different factors, such as the absolute and relative concentrations of the various ions that form the glass, the melting temperature, the cooling rate, and the temperature at final disposal. By suitable manipulation of these parameters, glass technologists can optimize the conditions for solubility of a desired component.

The properties of a cation (positive ion) that determine to a significant extent its behavior in glass are surprisingly few in number: these include its size and its charge (which in turn fix the strength with which it interacts electrostatically with other ions). For this reason, the solubility of plutonium can be estimated by comparison with the solubilities of less exotic cations which have similar characteristics. The properties affecting solubility, while rather difficult to measure directly in glass, can be inferred from the structure of the crystals formed by the oxide compounds in which the cations can be found. This is because the bond lengths which occur in glasses are usually very similar to those that occur in the corresponding crystals, the former being disordered, but nearly as dense, versions of the latter.

For example, the dissolution of uranium in borosilicate glass has been studied by Schreiber et al.[101] Uranium is an actinide element like plutonium, and even though the metallic forms (and therefore the chemical properties) of the two elements are quite different, the oxides UO_2 and PuO_2 have very similar crystal structures, indicating that their behavior in glasses should be homologous. Uranium has been found to have a solubility (with respect to crystallization) as high as 40 percent by weight when in 6+ oxidation state with an optimized glass composition; thus it is reasonable to predict that a quantity of plutonium on the same order could be dissolved in glass before the onset of crystallization, assuming a sufficiently oxidizing environment.

The above experiments with uranium, however, did not include a detailed study of the microstructure, and thus cannot exclude the possibility of phase separation. To estimate the upper limit on plutonium concentration with respect to phase separation, one may turn to a study of the waste glass ABS-39, which was experimentally determined to be homogeneous over the observed temperature range (>600°C).[102] (At lower temperatures, phase separation would not be experimentally observable, for even if it were thermodynamically favored, it would be kinetically inhibited due to the high melt viscosity.) This glass contains both UO_2 (1.66 percent by weight) and ZrO_2 (1.26 percent by weight). The zirconium content is important in this context because the zirconium cation is tetravalent, like Pu^{4+}, and the compound ZrO_2 is composed of crystals with bond lengths and coordinations (numbers of nearest neighbors) similar to those of PuO_2. Although zirconium is known to promote phase separation, the concentration contained in ABS-39 is apparently below the critical value. If plutonium were substituted for zirconium and uranium, this glass could accommodate at least 4.4 percent by weight plutonium before the occurrence of phase separation. This estimate is consistent with the German experiment with plutonium solubility in borosilicate glass cited in the text.[47]

Changes in the composition of the base glass can have a significant impact on the quantity of plutonium that can be dissolved in the glass. For example, one study has shown that the zirconium saturation limit in aluminosilicate melts increases sharply as the number of alkali cations in excess of aluminum increases, implying that alkalis assist in the stabilization of soluble complexes formed by tetravalent cations of large ionic radius.[103] It is likely that this mechanism is largely responsible for the higher solubility found in the German glass relative to the Russian glass cited in the text,[48] since the composition of the latter contained a substantially smaller number of excess alkalis than that of the former. Future solubility studies should recognize the importance of this and similar mechanisms.

Due to the complexity of multicomponent glass systems, however, it is possible that specific chemical processes could occur which might lead to discrepancies with the above estimates. Thus the solubility of plutonium in HLW glass can only be conclusively established by performing a comprehensive series of experiments involving a wide range of glass compositions.

NOTES AND REFERENCES

1. J. Carson Mark, "Explosive Properties of Reactor-Grade Plutonium," (Washington DC: Nuclear Control Institute, August 1990).

2. Thomas B. Cochran, William M. Arkin and Milton M. Hoenig, *US Nuclear Forces and Capabilities* (Cambridge, MA: Ballinger, 1984), p. 31.

3. Three kilograms of plutonium per warhead is the average obtained by dividing the estimated production of weapons plutonium by the US and USSR by the estimated total number of warheads in their arsenals in the mid 1980s and subtracting about 25 percent to take account of losses during fabrication. See Frank von Hippel, David H. Albright and Barbara G. Levi, *Quantities of Fissile Materials in US and Soviet Nuclear Arsenals* (Princeton University, Center for Energy and Environmental Studies Report No. 168, July 1986).

4. The US nuclear weapons arsenal contains about 500,000 kilograms of highly enriched uranium. (See F. von Hippel, David H. Albright and Barbara G. Levi [op. cit.] 1986; and D. Albright, F. Berkhout and W. Walker, *World Inventory of Plutonium and Highly-Enriched Uranium* [Oxford: Oxford University Press, 1993] chapter 4). The size of the CIS weapons HEU stockpile is still uncertain, with estimates ranging from 500 tonnes up to 1,100 tonnes.

5. James D. Watkins, US Secretary of Energy, letter to Senator Carl Levin, 24 February 1992, enclosure 1, quoted in *Rethinking Plutonium: A Review of Plutonium Operations in the US Nuclear Weapons Complex* (Columbia, SC: Energy Research Foundation, April 1992), p. 13. The rate of 2,000 per year is based on the facility operating one shift per day, five days per week.

6. The igloos at Pantex are described in George T. West, *United States Nuclear Warhead Assembly Facilities (1945–1990)* (Pantex Plant, Amarillo, TX: Mason & Hanger - Silas Mason Co., 1990) as being "nominally 12 meters deep, 7.6 meters wide and 4.5 meters high at most" for a floor area of about 90 m^2. See also Robert S. Norris and William M. Arkin, "Pantex Lays Nukes to Rest," *Bulletin of the Atomic Scientists*, October 1992, pp. 48–49. John Fleck, in "DoE Eyes Los Alamos Lab for Plutonium Work," *Albuquerque Journal*, 17 August 1992 quotes the report of a Department of Energy task force meeting on the US weapons plutonium problem as to the effect that the new Nuclear Materials Storage Facility at the Los Alamos National Laboratory in New Mexico has sufficient capacity to store 60 tons of plutonium, an "aging vault complex" at the former plutonium-production site at Hanford, Washington has a capacity of about 20 tons and that the storage capacity at the other former US plutonium-production site, near the Savannah River in South Carolina, is only a little more than half a ton. See also *Storage of Plutonium in the Naval Fuel Material Facility* (Aiken, SC: Westinghouse Savannah River Company Safety Analysis Report No. DPSTSA-200-16, Addendum 1, October 1991 review draft).

7. Victor Mikhailov, Minister, Russian Ministry of Atomic Energy, private communication and interview in *Kosmolskaya Pravda*, 22 July 1992. The four sites are: Nizhnya Tura and Zlatoust in the Urals; and Penza and Arzamas-16 south of Gorky. The maximum disassembly rate is reportedly slightly lower than the maximum assembly rate (7,000 warheads per year).

8. Viktor Slipchenko, Department of Non-proliferation, "Arms Export Control and Conversion," Russian Foreign Ministry, presentation at the International Workshop on the Disposal of Plutonium, Bonn, 16 June 1992.

9. "Soviet Nuclear Threat Reduction Act," *Congressional Record*, 27 November 1991, S18798.

10. See, for example, the testimony by a panel of Bush Administration officials in the Senate Armed Services Committee Hearings, *The Disposition of US and CIS Strategic Nuclear Warheads under the START I Treaty and the June 17, 1992 US/Russian Joint Understanding on Further Reductions in Strategic Offensive Arms (START II)*, 4 August 1992.

11. Evgeny Mikerin, Director, Department of Uranium Enrichment, Plutonium and Isotope Separation, Russian Ministry of Atomic Energy (MinAtom), presentation at the International Workshop on Disposal of Separated Plutonium, King's College, London, 18 June 1992.

12. "US Agrees To Buy Russian HEU, But Details Must Be Worked Out," *Nuclear Fuel*, Special Issue, 2 September 1992. Also see Thomas L. Neff, "Integrating Uranium from Weapons into the Civil Fuel Cycle," *Science & Global Security* 3 (3–4) 1992.

13. "It's Official: US Stops Making Material for Nuclear Weapons," *New York Times*, 14 July 1992, pp. 1, 18. The decision had already been made unofficially somewhat earlier (see *Nuclear Weapons Complex Reconfiguration Study* [Washington DC: US DOE, DOE/DP-0083, January 1991], p. 49).

14. Thomas B. Cochran and Robert Standish Norris, *Russian/Soviet Nuclear Warhead Production* (Washington DC: Natural Resources Defense Council, 15 May 1992); and "Russia," *Nucleonics Week*, 9 July 1992, p. 19.

15. Production of fresh weapons plutonium also continues because older plutonium contains on the order of a tenth of a percent of Am-241. In the US (at the Rocky Flats facility [now closed]) Am-241 was separated out of recycle plutonium using a pyrochemical process. The Russian approach has apparently been to dilute the recycle plutonium with fresh plutonium. However, an americium removal facility is reportedly under construction in Russia. If the demand for fresh weapons plutonium ends while there is still a need for the heat and electricity from the remaining reactors, their fuel demand could be reduced by an order of magnitude by going to higher fuel burnup. However, the fuel would probably still have to be reprocessed eventually, because it contains uranium in metal form and is therefore not suitable for direct disposal.

16. Russian President Boris Yeltsin, television and radio address on arms control, 29 January 1992.

17. William Dircks, "Nuclear Fuel Recycling—The IAEA Perspective," paper given to the 25th Japan Atomic Industrial Forum Annual Conference, Tokyo, April 1992.

18. After about five years, and for more than 100 years thereafter, the gamma dose from spent fuel will be dominated by the 0.66-MeV gamma ray that is emitted in 85 percent of the decays of 30-year half-life Cs-137 (*Table of Isotopes, 7th edition*, C.M. Lederer and V.S. Shirley, editors, [New York: John Wiley & Sons, 1978]).

Consider a freshly discharged fuel assembly of uranium with a burnup of 33 MWd $kgHM^{-1}$. The fuel will contain about 3.3 curies of Cs-137 per MWd (see note 53) or 96 curies per kilogram UO_2. The "dose rate" at which energy from the Cs-137 gamma rays is being generated in the fuel is D_A = 1,150 joules per kg-hour. The gamma energy-absorption coefficient in UO_2 at 0.66 MeV is $\mu_U = 0.0718\ cm^2\ gm^{-1}$.

In the approximation that the mean length for gamma energy absorption is small relative to the radius of the fuel assembly, the dose in tissue at the surface is $D_S = 0.5D_A\mu_w/\mu_U$ = 260 Grays per hour (joules per kg-hour), where $\mu_w = 0.033\ cm^2\ gm^{-1}$ is

the energy-absorption coefficient in tissue (approximated as water) for 0.66-MeV gamma rays.

At distances r perpendicular to the center of the fuel assembly that are short in comparison to its length but long in comparison to its radius, R, the dose rate is approximately $D = 0.5D_S R/r = 15/r$ Grays per hour (r in meters). At $r = 1$ meter and after one Cs-137 30-year half-life D will still be eight Grays per hour. Given that a lethal dose is 2.5–4.5 Grays, a dose rate of eight Grays per hour would become lethal in about 30 minutes. (Gamma energy attenuation coefficients from J.H. Hubbell, *Photon Cross Sections, Attenuation Coefficients, and Energy Absorption Coefficients from 10 keV to 100 GeV* [National Bureau of Standards, NSRDS-NBS 29, August 1969], Table 1-7.)

19. After a few years, the gamma dose from reactor-grade plutonium will be dominated by the 0.06-MeV gamma ray emitted in 36 percent of Am-241 decays (*Table of the Isotopes*). For a near-worst case, consider reactor-grade PuO_2 that originally contained 9.1 percent Pu-241 (see table 3) after all that Pu-241 had decayed into 430-year half-life Am-241. The average rate at which the 0.06-MeV gamma energy would be emitted in the PuO_2 would then be $D_{Pu} = 144$ joules per kilogram per hour.

Assuming a mass density of the PuO_2 of 10 gm cm^{-2} (87 percent of crystal density), the radius of a PuO_2 sphere containing four kilograms of plutonium would be $R = 4.6$ centimeters and the energy-absorption length for the 0.06-MeV gamma rays would be 0.02 centimeters. As in the previous note, in the approximation that the radius of the sphere is large in comparison with the energy-absorption length, we may calculate the surface dose as $D_S = 0.5D_{Pu}\mu_w/\mu_{Pu} = 0.46$ Grays per hour, about 1/500 of surface dose for the spent fuel assembly in the previous note, where the gamma energy-absorption coefficients in tissue (water) and PuO_2 at 0.06 MeV are $\mu_w = 0.032$ cm^2 gm^{-1} and $\mu_{Pu} = 5$ cm^2 gm^{-1}. (We have approximated μ_{Pu} by the value for UO_2).

At a distance r large in comparison to R, the dose rate will be $D = 0.5D_S(R/r)^2$. For $R = 1$ meter we obtain $D = 0.5$ milliGrays per hour, about 1/10,000 of the dose rate at this distance from a relatively fresh spent fuel assembly. We would like to acknowledge Steve Fetter's suggestion of the simple approach used in these calculations.)

20. Lawrence Scheinman and David A.V. Fischer, "Managing the Coming Glut of Nuclear Weapon Materials," *Arms Control Today*, March 1992, pp. 7–12.

21. For a discussion of portal monitors, see Paul E. Fehlau, *An Applications Guide to Pedestrial SNM* [Special Nuclear Material] *Monitors* (Los Alamos, NM: Los Alamos National Laboratory report # LA-10633-MS, 1986); and David Albright, "Portal Monitoring for Detecting Fissile Materials and Chemical Explosives," in *Reversing the Arms Race: How to Achieve and Verify Deep Reductions in the Nuclear Arsenals*, Frank von Hippel and Roald Z. Sagdeev, eds. (New York: Gordon & Breach Science Publishers, 1990) p. 239.

22. Thomas E. Shea, "On the Application of IAEA Safeguards to Plutonium and Highly Enriched Uranium from Military Inventories," *Science & Global Security* **3** (3–4). See also P. Chare, R. Schenkel, B.G.R. Smith, H. Wagner, S. Kaiser, J-C. Saglini, "Safeguarding Large Plutonium Stores," *13th Annual Symposium on Safeguards and Nuclear Material Management (Proceedings)*, Avignon, May 1991, pp. 71–76.

23. *Plutonium Fuel: An Assessment*, (op. cit.) p. 64.

24. C.H. Bloomster, et al.,*Options and Regulatory Issues Related to Disposition of Fissile Materials from Arms Reductions* (Richmond, WA: Pacific Northwest Laboratory, PNL-SA-18728, 1990) p. 12, based on US Nuclear Regulatory Commission, *Final Generic Environmental Statement on the Use of Recycle Plutonium in Mixed-Oxide*

Fuel in Light Water Cooled Reactors (GESMO), NUREG-0002, IV-I-8, August 1976.

25. Assuming a four percent real interest rate and an operating life of 30 years, the annual capital charge is $14 million. Bloomster et al. assume that there would be a total staff of 200 (160 guards) at a "fully-burdened" cost of $110,000 per person per year for an annual salary cost of $22 million. If $6 million is included for annual operating and maintenance costs, total annual costs come to $42 million.

26. Operating costs are estimated at $44 million per year during loading and removal of plutonium at the facility, and $27 million during the storage period. See E.R. Johnson, "Alternatives for Disposal of Plutonium from Nuclear Weapons Disarmament Activities," paper given at the Annual Meeting of the Institute of Nuclear Materials Management, Orlando, Florida, July 1992.

27. *Plutonium Fuel: An Assessment* (Paris: OECD/NEA, 1989) Table 14. More recently, Cogema has announced a "de-americiation" plant to be commissioned at La Hague by 1995. The cost of this procedure, which relies on dissolving the plutonium oxide, using an oxidation agent based on silver, followed by an ion-exchange process, is said to be "relatively modest" (A. MacLachlan, "Cogema Inaugurates UP3 With Promise of More Upgrades," *NuclearFuel*, 27 April 1992).

28. For each gram of fissile plutonium contained in fresh MOX fuel, approximately one megawatt-day of fission energy is generated. (One gram of fission releases approximately one megawatt-day of energy. Although the fissile plutonium is not be completely fissioned, some of the U-238 in the fuel is converted into plutonium and fissioned. These two effects approximately cancel each other in MOX fuel.) A world LWR capacity of 300 GWe operating at a capacity factor of 70 percent with heat-to-electricity conversion ratio of one-third, would release 230 million MWd (thermal) of fission energy per year, equivalent to 230 tonnes of fission. If by the year 2000 there is 200 tonnes of surplus weapons-grade plutonium (94 percent fissile) and 200 tonnes of reactor-grade plutonium (70 percent fissile), this material would, in principle, be sufficient to power world LWR capacity for about $(0.94 \times 200 + 0.7 \times 200)/230 = 1.4$ years.

29. According to OECD estimates, world uranium resources are 5.4 million tonnes at extraction costs of less than $130 per kilogram (1989 prices) in "reasonably-assured reserves" and "estimated additional resources" (extensions of known deposits) *(Uranium: Resources, Production and Demand 1989* [Paris: OECD/NEA, 1990] pp. 20, 24, 26, 28, 30). At a burnup of 50.7 MWd $kgHM^{-1}$ (a reasonable assumption for LWRs by the year 2000) about 140 tonnes of natural uranium is required per operating GWe-year (5.2 percent enrichment of LEU and 0.225 percent enrichment of uranium tails assumed). (In addition, 685 tonnes of uranium per GWe—about 5 years annual use—would be required for the initial cores.)

30. The OECD best estimate is 19.1–23.5 million tonnes for potentially recoverable uranium from conventional resources, including "speculative resources" recoverable at costs less than $130 per kilogram, see *Uranium: Resources, Production and Demand, 1989* (Paris: OECD/NEA, 1990).

31. The annual discharge of *fissile* plutonium per GWe-yr (70 percent capacity factor) for an LWR operating with 50.7 MWd kg^{-1} burnup is 0.12 tonnes per year (*Nuclear Energy and Its Fuel Cycle* [Paris: OECD/NEA, 1982], p. 149).

32. *Plutonium Fuel: An Assessment*, (Paris: OECD/NEA, 1989) Table 12B (for 43 MWd $kgHM^{-1}$ burnup).

33. W. Gmelin, "Plutonium Safeguards within the European Community," *1990 Insti-*

tute for Nuclear Materials Management Proceedings (annual meeting), pp. 8–13.

34. Marvin Miller, *Are IAEA Safeguards on Plutonium Bulk-handling Facilities Effective?*, (Washington DC: Nuclear Control Institute, August 1990); and Frans Berkhout and William Walker, "Safeguards at Nuclear Bulk Handling Facilities," in J. Poole (ed), *Verification Report 1992* (London: VERTIC, 1992).

35. Discount factors due to fuel cycle leads and lags are ignored (see *Plutonium Fuel: An Assessment* [op. cit.] pp. 125–125).

36. *Plutonium Fuel: An Assessment*, (op. cit.) p. 50. Assuming a heat-to-electricity conversion ratio of one-third and an annual capacity factor of 70 percent. For a burnup of 33 MWd $kgHM^{-1}$, the feed is 0.28 tonnes of fissile plutonium per year. For a burnup of 53 MWd $kgHM^{-1}$, it is 0.24 tonnes per year.

37. Of current world nuclear capacity of about 330 GWe, some 85 percent is LWR capacity (about 280 GWe). "Electricity Generated by Nuclear Power in 1991", *Nuclear Engineering International*, May 1992, p. 4.

38. Darrell F. Newman and Andrew W. Pritchard, "Non-Fertile Fuel for Nuclear Reactors," Invention Report, Battelle Pacific Northwest Laboratories, 19 February 1992; and *Plutonium Fuel: An Assessment*, p. 50. In principal, the isotopic mix of plutonium could be improved by softening the neutron spectrum to increase the capture-to-fission cross section of the fuel. This could be accomplished by substituting beryllium metal for the UO_2 in the fuel. However, besides serving as a fertile material for the creation of new plutonium, the U-238 oxide also provides a prompt negative reactivity feedback due to the increased absorption of neutrons in the U-238 if the fuel temperature increases. This is an important safety feature. An equivalent effect could be obtained by adding a small amount of an isotope like Rh-103. (Darrell F. Newman, Pacific Northwest Laboratories, private communication, June 1992).

39. The original rationale for fast reactors was that they could extract roughly 100 times more energy out of a kilogram of uranium than conventional LWRs by converting the non-fissile U-238 that makes up 99.3 percent of natural uranium into fissile plutonium isotopes. A breeder would be fueled by plutonium but would, in effect, sustain itself on U-238 by producing more plutonium than it consumed. The main economic problem of fast reactors is that their higher capital costs are not offset by lower fuel cycle costs at foreseeable uranium prices.

40. Naoaki Usui, "Japan Prepares for Forum on Nuclear Warhead Disposal," *Defense News*, 2 March 1992, p. 8. The costs quoted by Japan are roughly comparable to costs incurred for the Superphénix fast reactor—$5.3 billion for the period 1986–1992, including evidently fuel and operating costs as well as capital costs. (A. Machachlan, "Prime Minister Orders More Work, Public Inquiry for Superphénix," *Nucleonics Week*, 2 July 1992, p. 13).

41. For a plant life of 30 years and a real discount rate of four percent, the capital recovery factor is 0.057; so that the annual capital charge for a $4 billion plant would be $229 million. In addition, the fabrication costs for two tonnes of plutonium per year would be about $15 million, assuming that the cost of fabrication of one kilogram of MOX fuel is $1,500 (roughly the same as for LWR-MOX fuel) and a plutonium content of 20 percent in the MOX.

42. If the *incremental* cost of power from the fast reactor relative to that of an LWR fueled by LEU was attributed solely to plutonium burnup, the cost of irradiating 60 tonnes of plutonium over a 30-year period would be about $3.5 billion. This assumes an

LWR capital cost about half that assumed above for a fast reactor. The cost of a 1-GWe LWR in Japan is now about $2.4 billion (at an exchange rate of 125 yen per dollar). (Tatsu Suzuki, Massachusetts Institute of Technology, private communication, 20 November 1992.) This is less than half the capital cost quoted by the Japanese for the plutonium burner. Assuming a source of free plutonium and a once-through fuel cycle, the fuel costs for a fast reactor and LWR would be roughly comparable; but the capital costs of nuclear power dominate the generation cost of electricity.

43. *Plutonium Fuel: An Assessment*, (Paris: OECD/NEA, 1989) table 12.

44. J.A.C. Marples, "The Preparation, Properties, and Disposal of Vitrified High Level Waste from Nuclear Fuel Reprocessing," *Glass Technology* **29** (6) December 1988, p. 235.

45. C.H. Bloomster, et al. (op. cit.).

46. C.H. Bloomster et al., p. 32; and private communications with Bloomster and with John Mandel, Pacific Northwest Laboratories (ret.), June 1992.

47. C.T. Walker and U. Riege, "Compatibility of Actinides with HLW Borosilicate Glass: Solubility and Phase Formation," in *Ceramics in Nuclear Waste Management*, T.D. Chikalla and J.E. Mendel, eds. (National Technical Information Center: DOE CONF-790420, 1979), pp. 198–202.

48. E.S. Prokin, O.A. Alekseev, T.N. Ananina, and E.E. Ermoleav, "Behaviour of Plutonium Dioxide in a Molten Phosphate Glass," *Radiokhimiya* **31** (1) pp. 140–144, January/February 1989; A.S. Nikiforov, A.S. Polyakov, V.V. Kulichenko and Y. Matyunin, "Problem of localization of transplutonium elements," *Radiokhimiya* **32** (2) March/April 1990; V.V. Kushnikov, Yu.I. Matyunin and N.V. Krylova, "The Behaviour of Alpha-emitting Radionuclides in the Solidification of High-activity Waste," *Atomnaya Energiya* **70** (4) April 1991, pp. 239–243.

49. The saturation level is about 5×10^{18} alpha disintegrations per gram. R.P. Turcotte, "Radiation Effects in High-level Radioactive Waste Forms," *Radioactive Waste Management 2*, December 1981, pp. 169-177.

50. J.A.C. Marples (op. cit.).

51. The specific heat of waste glass is about 0.7 joules $(\text{gram-}^{\circ}\text{C})^{-1}$ (J.A.C. Marples [op. cit.]).

52. The slowest rate of cooling discussed by Marples (op. cit.) is from 1,000°C to 70°C in 80 hours at the Pamela plant in Belgium. Assuming an ambient temperature of 20°C, this corresponds to an exponential cool-down time constant of 27 hours. At the 450–500°C temperature at which solidification occurs, the cooling rate would then be about (400°C)/(27 hours) = 15°C per hour.

53. Fission of a gram of U-235 by thermal neutrons produces a total of 6.17 curies (about 3 curies each) of Sr-90 and Cs-137. Fission by thermal neutrons of a gram of Pu-239 produces about 3.34 curies of Cs-137 and 1.09 curies of Sr-90 for a total of 4.43 curies gm^{-1}. (D.F. Rider, *Compilation of Fission Product Yields* [Vallecitos Nuclear Center, Pleasanton, CA: General Electric Report No. NEDO-12154-3C, 1981)].

54. US DOE, *Integrated Data Base for 1991*, DOE/RW-0006, Rev. 7, October 1991, Tables 2.12, 2.17. The HLW in the tanks takes four forms: liquid, sludge, salt cake and slurry. In addition, the numbers for Hanford include Sr-90 and Cs-137 that has been isolated in capsules that will also be mixed into the HLW glass. The published activities of the stored wastes are about twice as large as quoted here because they include

short-lived decay products of Sr-90 and Cs-137 (2.7-day half-life Y-90 and 2.55-minute half-life Ba-137m, respectively).

55. US General Accounting Office (GAO), *Nuclear Waste: Defense Waste Processing Facility—Cost, Schedule, and Technical Issues* (Washington DC: GAO/RCED-92-183, June 1992), chapter 2.

56. US DOE, *Integrated Data Base for 1991,* Table 2.12. See also the US Congressional Office of Technology Assessment report, *Long-lived Legacy: Managing High-Level and Transuranic Waste at the DoE Weapons Complex* (Washington DC: OTA background paper, May 1991).

57. For a burnup of 33 MWd $gmHM^{-1}$. About two-thirds of the fission is of U-235 (6 curies of Sr-90 plus Cs-137 per gram of fission) and about one-third of Pu-239 (four curies gm^{-1}).

58. US GAO, *Nuclear Waste: Defense Waste Processing Facility—Cost, Schedule, and Technical Issues.*

59. Based on 1990 inventories. 12,000 cannisters are due to be filled with glassified liquid, sludge and salt cake, 2,000 cannisters with glassified slurry and 2,000 cannisters with Sr-90 and Cs-137 capsules. Each cannister is to have a volume of 625 liters. (Steve Burnham, Department of Energy, Hanford, private communication, 27 May 1992.) We assume a glass density of 2.5 gm cm^{-3}.

60. Thomas Cochran and Robert Norris, *Russian / Soviet Nuclear Warhead Production,* (Washington DC: Natural Resources Defense Council, NWD 92-1, 2 April 1992), p. 19.

61. There is a lot of confusion about this number. The *Material of the Commission to Investigate the Ecological Situation in the Chelyabinsk Region* quoted in Cochran and Norris, NWD92-1, p. 41, gives 500 megacuries for the total activity. Another report gives a figure of 976 megacuries (*Conclusion of the Environmental Experts of the State Expert Committee of the USSR State Planning Commission* [GOSPLAN] *and of the Expert Group of the Supreme Soviet of the USSR* [Chelyabinsk: South Urals Publishers, 1991], pp. 23, 25.) These numbers should be approximately halved if they include the activity of the short-lived decay products of Sr-90 and Cs-137. The report of the environmental experts also indicates that approximately 153 megacuries have been released into the environment (including 120 megacuries into the waste pond Lake Karachay). However, in this case, apparently only the Sr-90 and Cs-137 are counted.

Prior to 1978 the Mayak complex reprocessed fuel from the six reactors on the site (five military graphite-moderated production reactors and one heavy-water-moderated reactor). These reactors (now shutdown) are estimated to have fissioned about 30 tonnes of U-235 prior to 1978 (Cochran and Norris, 1992, pp. 72–73). This would have been associated with the production of about 180 megacuries of Sr-90 plus Cs-137, which would have decayed to about 100 megacuries by 1992. In 1978, the plant was converted to the reprocessing of civilian fuel and since then, about 30 tonnes of civilian plutonium have been separated. (Evgeny Mikerin, private communication, June 1992). Assuming that the fuel burnup averaged 20–30 GWd t^{-1}, this would be associated with the fission of 58-64 tonnes of U-235, generating about 480 megacuries of Sr-90 plus Cs-137—about 400 megacuries after decay to 1992. On this basis, the total inventory of HLW at Chelyabinsk-40 should be 490–560 megacuries of Sr-90 plus Cs-137. Subtracting the 153 megacuries reported to be in the environment and the 45 megacuries already glassified, would leave 295–360 megacuries in the HLW tanks.

62. Private communication, Russian Ministry of Atomic Energy (MinAtom) official, June 1992.

63. At 33 MWd $kgHM^{-1}$ and one gram of fission per MWd, 250 tonnes of spent fuel would contain 8.25×10^6 grams of fission (one MWd corresponds to within a few percent to one gram of fission). About two-thirds of this fission would have been of U-235 (6.17 curies of Sr-90 plus Cs-137 per gram of fission) and one-third of Pu-239 (4.43 curies gm^{-1}) for a total of 46 MCi.

64. Evgeny Mikerin, private communication, June 1992.

65. Britain's first generation of power reactors were scaled up versions of its graphite-moderated, CO_2-cooled, plutonium-production reactors. The fuel for these reactors is natural uranium metal clad in a magnesium-oxide alloy. Hence the name "Magnox."

66. For data prior to 1984, see *Quantities of Fissile Materials in US and Soviet Nuclear Weapons Arsenals*, Table 5-11. From 1984–1992, 7,600 metric tonnes of Magnox fuel with an average burnup of approximately 4,400 MWd t^{-1} were reprocessed (D. Albright, F. Berkhout and W. Walker, *World Inventory of Plutonium and Highly-Enriched Uranium* [Oxford: Oxford University Press, 1993]).

67. J.A.C. Marples (op. cit.) indicates about one curie gm^{-1} "including activities of daughters." *Nuclear News*, April 1991, p. 71 cites an activity of 0.3 megacuries per 400 kilograms of glass. We assume that both numbers include the activities of short-lived Y-90 and Ba-137m decay products and therefore divide by two to get the Sr-90 plus Cs-137 activity (see note 56).

68. *Nuclear News* (op. cit.) April 1991, p. 71.

69. D. Albright, F. Berkhout and W. Walker (op. cit.).

70. For data prior to 1984 see *Quantities of Fissile Materials in US and Soviet Nuclear Weapons Arsenals*, Table 5-13. For subsequent years, see H. Hirsch and M. Schneider, *Wackersdorf ist Tot—Es Lebe La Hague* in *Rest-risko*, no. 6, (Hamburg: Greenpeace, April 1990), p. 10 and D. Albright, F. Berkhout and W. Walker (op. cit.), Table 6.6.

71. D. Albright, F. Berkhout and W. Walker (op. cit.).

72. Charles Forsberg, Oak Ridge National Laboratory, private communication, June 1992.

73. C.H. Bloomster et al. (op. cit.), p. 35. The following costs are included: $7 million for conversion into oxide, and $8 million for processing into glass.

74. Assuming a useful plant life of 30 years, and a real discount rate of four percent, the additional annual capital charge would be $6 million. Additional operating and maintenance costs are estimated at $24 million per year. The annual increment due to plutonium handling, would therefore be $30 million. Source: John Plodinek, Senior Technology Advisor, Savannah River Technology, private communication, December 1992.

75. C.H. Bloomster, et al. (op. cit.) estimate vitrification costs of $130,000 per tonne of glass. E.R. Johnson (see reference in note 26) estimates vitrification costs in a dedicated facility as $170,000 per tonne of glass—or $34 million per tonne of contained plutonium at the assumed concentration of 0.5 percent.

76. I.A. Andryushin, Yu.A. Trutnev, and A.K. Tchernyshev, "Problems Related to the Development of Nuclear Explosive Technologies for Elimination of Toxic Waste," paper presented at the Fourth International Workshop on Nuclear Warhead Elimination, Washington DC, 26–7 February 1992, Washington DC (translation available from the Natural Resources Defense Council, Washington DC), p. 31.

77. A US study of a similar approach to the destruction of chemical weapons claims great economies of scale if a program of many explosions were carried out and mining were done hydraulically—about $6 million dollars for each of 36 100-kiloton explosions [Alexandria, VA: Defense Nuclear Agency, *The Feasibility of Chemical Munitions Disposal Using Nuclear Explosions*, 1982], p. 81.

78. It would require the fission of about six kilograms of fissile material to produce a yield of 100 kilotons, assuming no significant thermonuclear contribution to the yield. The fast fission of one gram of U-235 yields 6.03 curies of Sr-90 plus Cs-137 and the fast fission of one gram of Pu-239 4.28 curies (D.F. Rider, *Compilation of Fission Product Yields*, 1981).

79. Theodore B. Taylor, presentation at the Fourth International Workshop on Nuclear Warhead Elimination, Washington DC, 26–27 February 1992, Washington DC.

80. See A.G. Croff and G.E. Michaels, "An Overview of Partitioning-Transmutation," proceedings of the American Nuclear Society conference, *LMR* [Liquid Metal Reactors]: *A Decade of LMR Progress and Promise*, Washington DC, 11-15 November 1990; and "Trying Transmutation," *Scientific American*, May 1992, pp. 36-37.

81. In one scheme analyzed by LANL, spallation neutrons would be thermalized by a heavy-water moderator to produce a dense neutron flux. These neutrons would then be incident on a slightly subcritical liquid target containing small spheres of plutonium oxide suspended in heavy water. (Edward Arthur, Los Alamos National Laboratory, private communication, June 1992). While the physics of accelerator production of neutrons seems straightforward, major engineering problems would have to be solved to make this a practical scheme.

82. See T. Inoue, M. Sakata, H. Miyashiro, T. Matsumura, A. Sasahara and N. Yoshiki, "Development of Partitioning and Transmutation Technology for Long-lived Nuclides," *Nuclear Technology* **93**, February 1991, pp. 206–219.

83. *Impacts of New Developments in Partitioning and Transmutation on the Disposal of High-level Nuclear Wasted in a Mined Geological Repository*, Lawrence Livermore National Laboratory, mimeo (Lawrence D. Ramspott, *et al.* (LLNL); Thomas Cotton and John Burns (JK Research Associates); Amy McCabe and William Colglazier (University of Tennessee) and William W.-L. Lee (Lawrence Berkeley Laboratory) February 1992.

84. See J.K. Bates, J.P. Bradley, A. Teetsov, C.R. Bradley, and M. Bucholtz ten Brink, "Colloid Formation During Waste Form Reaction: Implications for Nuclear Waste Disposal," *Science 256*, 1 May 1992, pp. 649-651.

85. See H. Shaw (LLNL), "Is Borosilicate Glass a Better Waste Form than Spent LWR Fuel?" in Appendix C.

86. All figures used here are derived from: D. Albright, F. Berkhout and W. Walker, *World Inventory of Plutonium and Highly-Enriched Uranium* (Oxford: Oxford University Press, 1993) SIPRI, chapters 5–7.

87. Such an assessment is given in chapter 12 of D. Albright et al. (op. cit.).

88. These assumptions correspond to annual MOX fuel reloads of 96 tonnes per year in France, 108 tonnes per year in Germany and 72 tonnes per year in Japan. Furthermore, we assume that depleted uranium is used as the matrix in all cases, and that the plutonium has been separated from fuel with a burn-up of 33 GWd t^{-1} stored for 10

years. Total plutonium enrichment of the MOX is therefore 5.6 percent. Source: M. Rome, et al., 1990 (op. cit.) in Table A4, note a.

89. We assume that the U-235 enrichment in recovered uranium is 0.92 percent, equivalent to 0.2 SWUs of separative work per kilogram assuming a 0.3 percent U-235 in the tails assay. About 1.5 kilograms of natural uranium (U_{nat}) would be required to produce one kilogram of 0.92 percent enriched uranium at this tails assay. Thus the value of recovered uranium U_{rec} is approximately (1.5 · U_{nat} price) + (0.2 · SWU price). At current nat U prices ($40 per kilogram) and SWU prices ($100 per SWU) the value of U_{rec} comes to $80 per kilogram. However, account has to be taken of the neutron poisoning effect of U-236 and higher fabrication costs due to enhanced gamma radiation in U_{rec}. This reduces the value by about 30 percent. See *Plutonium Fuel: An Assessment*, (op. cit.) Annex H, section 2, pp. 124-131.

90. The fixed cost of low-enriched uranium fuel is the sum of the costs of converting natural uranium to UF_6 (assumed to be $7 per kilogram U) and of fuel fabrication ($200 per kilogram U for low-enriched uranium fuel). (*Plutonium Fuel: An Assessment*, [op. cit.] p. 69.) Enrichment and uranium prices are treated as variable costs. The enrichment assumed here was 3.7 percent U-235. This corresponds to 4.71 kilogram separative work units (SWU) and 8.3 kilograms of natural uranium (0.711 percent U-235) per kilogram of enriched uranium in the fuel assuming that the associated depleted uranium contains 0.3 percent U-235 (the approximate optimum for the current ratio of prices for natural uranium and enrichment work).

91. The average delivered price of natural uranium to US utilities was $34.50 per kilogram U in 1990 (*Domestic Uranium Mining and Milling Industry 1990* [US DOE Energy Information Administration: Washington DC, 1991], Appendix E, Table E10).

92. In mid October 1992 the average spot price of enrichment was $65–70 per SWU. US DOE Long-Term Contract Prices were $125 per SWU in October 1992. We take the mean price paid by utilities to be $100 per SWU. Source: "Current Uranium Pricing Indicators," *NuclearFuel* 12 October 1992, p. 2; and *Nukem Market Report*, August 1992, p. 32.

93. We assume an electrical efficiency of the reactor of 33 percent. At a burnup of 43 MWd kg^{-1}, the electric energy produced from a kilogram of fuel is then 34 million MWh kg^{-1}. A mill is $0.001.

94. Price-setting in MOX fabrication is still in the pre-competitive stage since most currently operating plants are upscaled pilot facilities. Typical price ranges now quoted are $1,300–1,600 per kilogram MOX. The lower figure is the base price charged by Siemens for PWR-MOX at its new Hanau plant. Source: M. Hibbs, "German Utilities Bracing for MOX Fuel Cost Increases," *NuclearFuel*, 6 January 1992.

95. *NuclearFuel*, 14 September 1992, p 3.

96. Peter Schmiedel, Siemens, presentation to the International Workshop on the Disposal of Plutonium, Bonn, 16 June 1992.

97. M. Hibbs, *NuclearFuel*, 6 January 1992.

98. 4.2 percent *fissile* plutonium per kilogram (*Plutonium Fuel: An Assessment*, p. 50).

99. See, *e.g.*, Horst Sholtze, *Glass: Nature, Structure and Properties*, (New York: Springer-Verlag, 1991); A.J.G. Ellison and A. Navrotsky, *Thermochemistry and Structure of Model Waste Glass Compositions*, in *Proceedings of the Radwaste Symposium*, (Materials Research Society: Pittsburgh, 1989).

100.W. Lutze and R.C. Ewing, eds., *Radioactive Waste Forms for the Future*, (Amsterdam: NorthHolland, 1988).

101.H.D. Schreiber, et al., "The Chemistry of Uranium in Borosilicate Glasses, Part 2. Base Compositions Containing Titanium Relevant to the Immobilization of Nuclear Waste," *Phys. Chem. Glasses* **23** (1982) pp. 147–153.

102.J.E. Engell and G. Roed, "Metastable Liquid Immiscibility in Nuclear Waste Glasses," in *Scientific Basis for Nuclear Waste Management*, Vol.6 (S.V. Topp, ed.) (New York: Elsevier, 1982) p. 609.

103.E. Bruce Watson, "Zircon Saturation in Felsic Liquids: Experimental Results and Applications to Trace Element Geochemistry," *Contrib. Mineral. Petrol.* **70** (1979) p. 407.