Reprocessing Alternatives: The Options Multiply

Reprocessing has for so long been a part of plans to complete the nuclear fuel cycle that little attention has been paid to ways in which a nuclear power system might operate without it.

The alternatives range from doing nothing (that is, considering spent fuel as waste) to doing something very similar to reprocessing, with the hope that its proliferation potential can be reduced by never quite separating the plutonium from the uranium. (Reprocessing was invented to separate pure plutonium for weapons and would accomplish much the same result in commercial plants.) Between these two poles, there are proposals for employing a variety of chemical and quasi-chemical processes in place of the Purex process, which virtually all the world's reprocessing plants now employ. There are also proposals to modify or replace the popular light water reactors—which produce 99 percent of this country's nuclear power and about 80 percent of the world's—because they are more wasteful of uranium than most other types of reactor. The issue of uranium utilization arises because reprocessing the plutonium from light water reactors would reduce the amount of uranium required for their operation by about 30 percent. The purpose of the alternatives is to keep pure plutonium from appearing at any stage of a civilian nuclear power cycle.

As hints from the White House suggest that the new Administration will take a strong position against any move to deploy reprocessing at the present time, options that have long been overlooked, forgotten, or debunked because they were out of line with past policies are being dusted off and reanalyzed. New studies are surfacing rapidly, but the various alternatives differ considerably in the degree of protection againstproliferation that they would afford.

The first study to be put on the table is the so-called tandem fuel cycle that was proposed last summer by the Arms Control and Disarmament Agency (ACDA) during an interdepartmental survey of the proliferation potential of the extant nuclear technologies. The tandem proposal would have taken spent fuel from light water reactors and burned it further in Canadian CANDU reactors to extract additional energy without reprocessing. The technical merit of the tandem proposal is being vigorously debated, but it marked a significant turn in the nuclear debate. "Relatively less proliferation had never before been a design objective of nuclear technology," says Thomas D. Davies of ACDA.

Although the rationale for most of the new studies is to explore ways to reduce proliferation, the exercise could in fact turn into a wider review of the premises of nuclear policy—not just the type of reprocessing chemistry, but also the choices that have been made about fuel cycles, reactor concepts, and the associated problems of waste disposal. The analysis is far from complete, but what is emerging is a rediscovery of a fact that the nuclear engineers of the 1950's were quite familiar with: a nuclear power system can be configured in many different ways.

Less Proliferation Is the Rationale

The simplest alternative to reprocessing is to dispose of the spent fuel in the form in which it comes out of the reactor—as durable ceramic pellets of uranium and plutonium dioxide sealed in noncorrosive tubes of zirconium alloy. Countries that have light water reactors have largely ignored this option, but Canada has not because the concentration of plutonium in CANDU fuels is much lower than in other reactor fuels. Methods that have been developed in Canada (see box) appear to be safe, reliable, and relatively inexpensive ways of storing spent fuels for 50 to 100 years.

Another approach is to try to find other chemical processes that would be safer against proliferation than the standard Purex process. The list of possibilities is a long one—Mark Goldstein at the Brookhaven National Laboratory has found that 32 different reprocessing techniques have been proposed in the scientific literature since 1942. Some of them proved less than satisfactory, of course, but others were dropped when the focus of nuclear research narrowed in the early 1960's. Apart from solvent refining techniques such as Purex, the processes that appear to have the most promise, according to Goldstein, are pyrometallurgical techniques (similar to the reprocessing scheme used in conjunction with the first U.S. experimental breeder, EBR-I), pyrochemical methods, and electrochemical methods—which Russian researchers are currently emphasizing. The basic problem with trying to make any reprocessing system proliferation-proof, however, is that most processes were designed to produce plutonium (or plutonium derivatives) in a relatively pure state. So a step backward is often required to make the plutonium less accessible.

The reprocessing alternative favored by the Energy Research and Development Administration (ERDA) is a minor modification of the well-established Purex process called coprocessing because the end product would be a mixture of uranium and plutonium. The modification would be accomplished by altering the partitioning stage of the Purex process, where the fission products have already been removed from dissolved spent fuel and uranium and plutonium are about to be separated. In one coprocessing scheme for light water fuel, no separation at all would be made. The mixture coming out of the reprocessing plant would be about 1 percent plutonium and 1 percent fissionable ^{235}U. In that case extra enriched uranium would have to be added to the coprocessing product to bring the fuel up to 3 percent fissionable content, as needed for light water reactors.

If the uranium-plutonium mixture produced by coprocessing were diverted for weapons, a chemical separation would be needed to get pure plutonium. However, the chemical separation would be simpler than reprocessing because the uranium-plutonium mixture would be largely decontaminated. "The fuel could be fairly easy to pull the plutonium out of," says Marvin Moss of ACDA, "but it would indeed have some safeguard advantages over normal reprocessing."

The coprocessing concept could also be applied to the breeder reactor, and such a possibility is being investigated by ERDA. Some form of reprocessing is essential for the breeder, which by definition is a reactor that produces more fissionable material than it consumes, because it must have the capability to recycle fuel to keep operating. Coprocessing, as applied to the breeder, would almost certainly require a plant with the capability to do some separation of uranium and plutonium, so the relevant safeguards question would be whether such a plant could not be surreptitiously altered to do complete separation. The breeder reactor needs a fuel with 20 percent fissionable content to operate, but the result of coprocessing would give a mixture with only about 8 percent plutonium. By drawing off some uranium at the partitioning stage, the plutonium concentration could be raised to the desired amount. According to researchers who have looked at the idea, it is clear that a plant can be built so that the results of the partitioning stage are variable (the half-finished plant in Barnwell, South Carolina, is built in just that way) but more work is needed to determine whether a design can be built that could not be easily altered. Breeder coprocessing could possibly be done without any uranium-plutonium separation, but this would require that some of the recycled breeder fuel be subsequently burned in light water reactors.

A variation of coprocessing being studied at Battelle Memorial Institute in Columbus, Ohio, is intended to minimize waste as well as proliferation. Through careful control of the oxidation states in the Purex process, Meyer Pobereskin hopes to remove the long-lived actinides from the waste stream and put them in the uranium-plutonium mixture. Although the chemistry necessary for this has not been tested yet, it could reduce the time scale of the waste disposal problem from many thousand to many hundred years and also make the uranium-plutonium mixture so radioactive (with actinides plus some fission products) that it could be handled only by remote control.

A "master blend" coprocessing scheme, proposed by R. Widrig at Battelle Pacific Northwest Laboratories, would produce a 10 to 30 percent plutonium mixture, which could be used in either breeders or light water reactors, by dilution with the proper amount of natural uranium.

If the scope of reprocessing alternatives is broadened to include reactors other than the present light water reactors, the number of options becomes surprisingly large. Modified light water reactors, CANDU reactors, and high-temperature gas reactors could each stretch uranium resources just about as far as reprocessing with the present light water reactors would. In addition, the neutron economics of light water and CANDU reactors could be significantly improved with thorium fuels, making further uranium savings and creating a category of "near breeder" reactors. Finally, a category of very advanced, conceptually unique reactors known as homogeneous reactors could greatly conserve resources and drastically change present perceptions of how proliferation-resistant a nuclear technology might be.

Table 1. Approximate natural uranium requirements for various reactors and fuel cycles, taken from documents published by Atomic Energy of Canada, Ltd. Items marked with an asterisk are interpolated from estimates by the U.S. Energy Research and Development Administration.

Reactor type	Fuel cycle	Natural uranium consumption (g kwe^{-1}year^{-1})
Light water (LWR)	Enriched uranium, once through	195*
	+ Uranium recycle	160
	+ Plutonium recycle	114
	Thorium burning (^{235}U makeup)	65
CANDU	Natural uranium, once through	130
	+ Plutonium recycle	60
	Thorium burning (^{235}U makeup)	20
High-Temperature gas (HTGR)	Highly enriched uranium + thorium	100*
Fast breeder	Uranium + plutonium recycle	~2

Perhaps the most thoroughly tested alternative to the light water reactor is the CANDU, of which there are more than a dozen large commercial units under construction or operating around the world. The CANDU reactor core is cooled and moderated with heavy water, and it can therefore operate with natural uranium fuel (which is composed of 0.7 percent fissionable ^{235}U) rather than the enriched uranium (3 percent ^{235}U) used by light water reactors.

A look at what happens to the burnable ^{235}U gives an indication why the CANDU achieves substantially better fuel efficiency. At the end of one pass through the CANDU, the ^{235}U content of the fuel is burned down to 0.2 or 0.25 percent, just as low as the concentration of ^{235}U left behind in the "tails" at the end of the enrichment process. It is also considerably lower than the ^{235}U concentration in light water reactor spent fuel (about 1 percent). The light water reactor would have to burn virtually all the ^{235}U in its fuel to match the CANDU's efficiency. As it happens, the spent light water reactor fuel is more than rich enough to start up a CANDU—the fact which basically explains why the tandem cycle proposed by the arms control agency is feasible. The details of the fuel history are actually more complicated because both reactors continually produce and consume plutonium, but the end result is that the CANDU uses about 30 percent less natural uranium to produce the same amount of power (Table 1).

A new study just completed by ERDA suggests that the same resource-saving benefits of the CANDU (or the tandem cycle) could be obtained by replacing some of the light water in the present U.S. reactors with heavy water.

Called a spectral shift reactor, the latest ERDA suggestion would burn the present U.S. reactor fuels longer by gradually changing the concentration of heavy water in the reactor during the life of each load of fuel. The concept could be "a possible modification of the standard pressurized water reactor," according to Saul Strauch at ERDA, who is heading a program to evaluate a number of ways to recover the energy value of used nuclear fuel without separating plutonium. The capital cost of the modification could be as low as $20 million (mostly the cost of heavy water) plus $1 million per year for extra operating costs, according to some estimates. Such a cost increment would be a minor influence on the price of electricity from a $1 billion reactor.

When a new load of fuel is installed, the spectral shift reactor would operate with a mixture of 60 percent heavy water and 40 percent light water, which gives a slightly faster spectrum of neutrons and promotes the production of plutonium in the fuel rods. As the fuel burns, heavy water is bled out of the reactor until the mixture at the end is about 90 percent light water. This shift counters the effect of the buildup of fission products, which tend to poison the reaction and stop it. By the end, most of the plutonium and ^{235}U would be burned. If many reactors were operated in the spectral shift mode, the United States might have to build a number of large plants to separate heavy water from natural sources—as the Canadians have done. But for a modest test of the principle, the present heavy water separation facilities at ERDA's Savannah River Laboratory would probably be sufficient.

The ERDA program is also studying two other ways to improve the fuel utilization of light water reactors—either using the present uranium oxide fuel with a slightly higher enrichment of ^{235}U or by improving on the past experience with metallic uranium fuel at lower enrichment.

Another presently available technology that can stretch uranium reserves as much as the CANDU or tandem is the high-temperature gas reactor. Whereas a light water reactor would use about 6000 tons of natural uranium (U_3O_8) over its 30-year lifetime, Strauch estimates that the high-temperature gas reactor would require only 3000 tons. But as presently designed, the reactor uses a fuel of thorium mixed with 90 percent enriched uranium, which is an ideal weapons material. Some engineers suggest that it could operate on lower enriched uranium with an economic penalty. But at lower enrichments, the resource saving benefits would be quickly reduced.

The thorium-uranium cycle could also achieve considerable fuel savings in water cooled reactors. In particular, the thorium-uranium cycle could make the CANDU into a near breeder. This fuel cycle would burn a small amount of natural uranium, but most of its energy would come from ^{233}U, a fissionable isotope of uranium bred from thorium. Since ^{233}U is a potential weapons material, most versions of this fuel cycle would dilute or "denature" the ^{233}U with natural uranium. The composition of fresh denatured fuel for a heavy water reactor might be 2 percent ^{233}U, 14 percent ^{238}U, and 84 percent thorium.

Most versions of the denatured fuel cycle would require reprocessing and would inevitably produce some plutonium from the ^{238}U left in the fuel to denature it. But the amount of plutonium would be one-tenth or less the amount produced in the present reactor system. The denatured thorium-uranium cycle is one of the more complicated alternatives suggested to reduce proliferation, and it would probably require at least one tightly safeguarded reprocessing plant. But it would have the advantage that fresh reactor fuel would be completely free of plutonium.

All the alternatives described above suggest ways to extract additional energy from a nuclear fuel cycle while making it more proliferation-resistant, but they implicitly use the present practice of the nuclear industry as the norm. Yet spent fuel from light water reactors contains plenty of plutonium (one reactor discharges 250 kg of plutonium per year) which could be extracted with chemical techniques, even though it might be a very messy job. The idea of a homogeneous reactor would—if it works—make possible a nuclear power system in which no fissionable material ever left the reactor once it started up and any diversion of material from the reactor would quickly shut it down. Such a reactor would straddle the line between the present "burner" reactors and the projected breeders, producing exactly as much fissile material in the core as it needed to keep itself going.

The input for the homogeneous reactor would be fertile material (natural thorium or uranium), which would only be made fissile inside the reactor's containment structure. The fuel would not be encased in fuel rods—the core would be a homogeneous medium such as gas, liquid metal, or molten salt so it could be circulated through a purifying system to continuously remove fission products. In

Canadian Disposal of Spent Fuel

For about a year, Canada has been testing a unique plan to store spent reactor fuel in concrete silos above ground to isolate the fuel's radioactive waste from the environment for 50 years or more.

With only four silos constructed so far, the program is not yet large enough to handle 1 year's fuel from one reactor. But what makes it remarkable is that it appears to be the sole test in any country of the simplest thing to do with spent fuel in lieu of reprocessing—to store it away for an indefinite period.

Like most countries, Canada puts its fuel into water-filled pools when it is discharged from the reactor, but after 5 years—when the decay heat of the fuel is considerably reduced—the fuel bundles could be sealed into steel cans and the cans would be sealed in 5-meter-high concrete silos. The thick silo walls would shield the environment from gamma rays, and the three layers of containment (fuel cladding, steel can, and concrete) would act as multiple protective barriers. Each silo is designed to hold 4.4 tons of spent fuel, which generates about 2 kilowatts of heat, raising the temperature at the center of the silo to about 135°C. The tests, which are being conducted at the Whiteshell Nuclear Research Establishment at Pinawa, Manitoba, show that the concept is working well so far.

Canadian workers lowering spent fuel into a 50-year storage silo.

Interim storage for 50 to 100 years, rather than long-term storage in a geologic formation, is being pursued in Canada because the Canadian nuclear policy is to defer any decision on whether to reprocess spent fuel or write it off as waste. The cost of using silos for interim storage would only be $10 per kilogram (in 1972 Canadian dollars), according to W. W. Morgan at Whiteshell. This is as much as 20 times cheaper than some U.S. estimates and would add only 0.1 mill/kwh to the cost of electricity. As ten silos would be needed for each CANDU reactor each year, the number required by a mature Canadian nuclear industry would not be small—but Morgan estimates it would cover only a few hundred acres at most by 2000.

American nuclear experts see no reason why the concept (or an alternative Canadian proposal for storage in air-cooled vaults) could not be used for light water fuels, although some modifications would be necessary because light water fuel stays in the reactor longer and has about three times as much decay heat per ton. The fuel assemblies are also longer, so they would probably require a taller silo. Anything other than short-term storage of spent fuel has been antithetical to the U.S. nuclear plan for so long that it has hardly been studied, although the Atlantic Richfield Hanford Company proposed some waste disposal concepts that could be applied to spent fuel. So far the Canadian plan appears to work, and as one U.S. official said, "it is so simple—no pumps, no fans, no filters."—W.D.M.

the gas version—which is generically related to the Rover program that developed a nuclear rocket engine for which there was no apparent practical use—the total amount of fissile material in the core might barely be enough to make one bomb. Such a reactor scheme, according to William Kitterman at ERDA, might be the only way to make the benefits of nuclear power available to any nation, regardless of its intentions, without also transferring a sufficient quantity of material with which to develop a nuclear arsenal. The homogeneous reactor proposal is a radical one—it would mean throwing out virtually all the nuclear technology that is now used.

The set of alternatives that have come to the surface during the last year of heightened public debate over proliferation is no doubt incomplete, and much of the analysis of the benefits of various proposals is in flux. Not the least of the problems is to decide where the proliferation line should be drawn, since there are now four or five different sets of criteria being used to distinguish where the risk of proliferation becomes too great. But the fact that reactors have never before been designed with proliferation considerations in mind is beginning to permeate the consciousness of many people, and "the technical questions are so fascinating that they are getting the community hooked," says Ted Taylor at Princeton University. What is becoming clear, says another well-respected nuclear engineer, is that there are a great many alternatives between the light water reactor, which "wastes uranium like crazy," and the liquid metal fast breeder, which is probably the most proliferation-prone technology yet conceived.

—WILLIAM D. METZ

This is the second of two articles.

Catastrophe Theory: The Emperor Has No Clothes

Catastrophe theory is one of the few areas of mathematics research that have surfaced from the mathematics community and caught the fancy of the press and the general public. The theory and its application were the subject of the first article on mathematics published in *Newsweek* in at least 7 years, were subsequently the subject of a *Scientific American* article, and have been praised by a number of mathematicians as well as by many investigators in other fields of science, including the social sciences. This attention, however, may have been premature, according to a number of eminent mathematicians who are harshly critical of catastrophe theory models.

The originator of catastrophe theory, René Thom of the Institut des Hautes Etudes Scientifiques in France, says in *New Scientist*, "Catastrophe theory is not a 'scientific theory'; it is a language and as with ordinary language every author will use it to his own taste and with his own 'style.' " But mathematicians stress that models based on catastrophe theory make use of a well-established body of mathematics, that was itself developed by Thom and others. This mathematics constitutes a study of how solutions to a system of equations vary when certain parameters that appear in the equations are perturbed. When the parameters vary, the solutions can be pictured as jumping from one value to the next. These jumps, or discontinuities, are said to be "catastrophes."

Catastrophe theorists believe that many discontinuous phenomena, such as the crash of a stock market or the sudden attack of an angry and frightened dog, lend themselves to descriptions in terms of these models. Catastrophe theory has been applied, during the past 6 years, in an enormous number of disciplines, including physics, biology, sociology, political science, economics, linguistics, and psychology. A number of investigators are now trying to use these models to study practical problems. For example, some psychologists in England are using a catastrophe theory model of the behavior of prisoners to advise prison authorities and to make decisions about ways to prevent prison riots.

Perhaps most disturbing to the critics are the claims made by the proponents of catastrophe theory models. Thom, for example, says that these models will provide such insight that, in the future "only mathematicians will have the right to be intelligent." The theory has been lauded as "an intellectual revolution in mathematics—the most important development since calculus." E. Christopher Zeeman of the University of Warwick in England, one of the chief publicists for catastrophe theory models, writes in *Scientific American*: "A mathematical method [catastrophe theory] for dealing with discontinuous phenomena has only recently been developed." In the opinion of many mathematicians, this statement ignores numerous developments, such as quantum mechanics, bifurcation theory, shock wave theory, and thresholds. Proponents of catastrophe theory models constantly stress their applicability to biology and the social sciences—fields in which other mathematical models have so far been only minimally successful.

Partly in response to the extraordinary publicity these models have generated and the extravagant claims made for their applicability to practical problems, some mathematicians have been examining the theory more closely. They have concluded that many of the statements and claims about the models are exaggerated, even irresponsible, and that the mathematical reasoning behind them is often sloppy or blatantly wrong.

Models Are Criticized

The current attack on catastrophe theory models is being led by Héctor Sussmann of Rutgers University in New Brunswick, New Jersey, and his associate Raphael Zahler (who made a very harsh criticism of catastrophe theory models of the propagation of nerve impulses). Sussmann, Zahler, and their supporters stress that they are criticizing not the mathematics underlying catastrophe theory but rather the models themselves. The heart of the criticism, according to John Guckenheimer of the University of California at Santa Cruz, is that Zeeman and Thom, the principal developers of these models, have a "real reluctance to get their hands dirty with the scientific details of the applications. Even more amazing to some is their cavalier attitude toward mathematics. Thom is hardly interested in proving theorems and Zeeman has used the work 'theorem' more loosely than any other mathematician I know."

Sussmann, focusing on the models of Zeeman, makes the feelings articulated by Guckenheimer more specific. Zeeman's models are the most numerous, the most widely known, and the most extensively criticized. Zeeman, contacted about Sussmann's harsh criticisms, said he was unfamiliar with the details of the criticisms; when they were described, he gave no direct or specific rebuttals to any of them.

One persistent problem in these models, Sussmann says, is that Zeeman plays on the propensity of readers to define

(Continued on page 350)