

immediate future, there seem to be no reasons to consider storage of plutonium for later use in FBRs as an economic alternative to prompt recycle of plutonium in LWRs.

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REPLY TO "COMMENTS ON 'ECONOMIC ALTERNATIVES FOR THE LONG-TERM USE OF PLUTONIUM PRODUCED IN LIGHT WATER REACTORS' "

My opinion about a possible limited stockpiling of plutonium is completely different from that of Ott.¹

Ott starts in his conclusions from an "intact nuclear scene." Unfortunately, this intact scene exists neither in Western Europe nor in the U.S. It is a matter of fact that today and for the near future in all countries of the western world, the fuel cycles of light water reactors (LWRs) and fast breeder reactors (FBRs) are not closed. Having this in mind, every speculation based only on mass balances and growth rates seems rather academic. It might be, and probably everybody working in the nuclear field hopes, that in the mid-1990's the situation for utilizing plutonium will be as simple as Ott has outlined, but presently and probably also for the near future (1976 until the end of 1980), the situation is rather complex and uncertain. Any utility that has to decide what to do with the forthcoming plutonium has to understand the following facts besides the economic demands:

1. As a result of the worldwide and long-term lack of reprocessing capacity for irradiated LWR fuel in Western Europe, only a fraction of the continually predicted (in literature) quantities of plutonium will be ready for utilization. In Western Europe, for example, we expect by 1985 4000 tons of irradiated fuel, but less than half of that can be reprocessed in the existing and planned reprocessing plants of the United Reprocessors GmbH, and this prediction seems in the face of the activities of the different powerful environmental groups more than optimistic. In the U.S. the situation is similar, if not worse.

2. As a consequence of this development, the different utilities in Western Europe and the U.S. build compact storage racks in their LWRs to enlarge the capacity in the existing and planned spent fuel pools. Presently they intend to store the irradiated fuel until the mid-1980's. This means that until this point in time, there is no plutonium available from this stored LWR fuel. Considering the stockpiling of plutonium, the enlarged capacity of spent fuel pools brings a new aspect into the

stockpiling scene, because in this case there are no additional charges for stockpiling the plutonium.

3. If utilities decide to recycle as much plutonium as they get (a) from the existing and working reprocessing plants and (b) from stores of partly unknown size, they are immediately faced with another problem. For instance, it is very difficult to get reprocessing agreements for the irradiated Pu/U fuel in the near future; the situation experienced in Europe so far is not very stimulating.

4. Utilities with FBRs have, because of reasons pointed out in items 1 and 2 above, difficulties in getting the necessary plutonium. [They need 2.6 tons Pu_{fiss} for one 1000-MW(e) FBR.] In addition, the first FBRs do not have the expected high breeding factors, and the large-scale reprocessing of FBR fuel is still unresolved.

5. As far as I know, there still exists in the U.S. much confusion and uncertainty about the licensing of plutonium recycling in LWRs. The major U.S. activity on the plutonium recycling field is "waiting" for a decision by the U.S. Nuclear Regulatory Commission on commercial recycling, to be made hopefully in 1977.

6. The tremendous increasing charges for U₃O₈ and separation work in the last few years have allowed the practically "worthless" plutonium to become a more and more "valuable" fission material. This fact is one of the present contradictions in connection with the use of plutonium, but plutonium gets its "value" only in closed fuel cycles.

And now the conclusion: In front of this background, there exist for several utilities the following alternatives:

- recycling the plutonium in LWRs and putting up with all the possible risks and uncertainties
- stockpiling the plutonium over a limited period for the later use in FBRs or so-called plutonium burners.

It was one of the purposes of the published work² in *Nuclear Technology* to show that stockpiling of plutonium over limited periods can be economically attractive, contrary to the usual published meaning of other authors in the past. Certainly I agree with these authors that stockpiling of plutonium is not the general solution, avoiding all the problems; in particular, the contrary is true, because the less plutonium that is recycled, the less experience is gained for fabrication and handling. But on the other hand, as practice shows, there are several utilities that are, for different reasons (essentially presented in the foregoing points 1 through 6), seriously interested in stockpiling plutonium. For these utilities, the general statement that stockpiling is economically unattractive is wrong, because in every single case the special situation of the utility has to be considered.

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A SHORT-DURATION LEACH TEST FOR RADIOACTIVE WASTE FORMS

The leach resistance of solid forms of radioactive waste is of major importance in determining the release rate of radioisotopes during long-term storage. Hespe¹ proposed the standard International Atomic Energy Agency leach test to provide a comparison of the leachability of the various forms of radioactive waste being developed. This test requires leaching for ~24 h (depending on pH of leachate) and analysis of the leachate for cations of interest by atomic absorption spectroscopy (AAS).

During the course of the Sandia program for solidification and consolidation of liquid radioactive waste by ion exchange with a hydrated titanate complex, a large number of samples were generated. The leach test proved to be the most time-consuming step in the evaluation of the various samples. By experimenting with various exposure times, it became apparent that in the case of crushed or powder samples, measurable leaching took place in a very short time. Those observations and the advantage of having a test of short duration led to the development of the instantaneous leach test.

The instantaneous leach test consisted of placing a crushed sample (not screened) into a 25-ml Royal Berlin Porcelain filter crucible with a porous bottom, with an average pore size of 7 μm , and pouring 100 ml of deionized water through with the aid of suction. Sample sizes of 1 g or less were used, and flow was adjusted to give water contact times of 3 to 4 min. Weight loss was determined on the sample, and AAS was used to analyze the leachate. The samples were dried for 1 h at 110°C and cooled in a desiccator for 0.5 h prior to weighing. A complete set of data can be obtained within 4 h depending on the number of elements analyzed for in the leachate. The results are typically expressed as grams of a given element leached per gram of sample, e.g., grams of cesium leached per gram of sample. The results can also be expressed as the fraction of an element leached from the sample.

The instantaneous leach test has proven to be surprisingly reproducible, considering that no attempt is made to control or determine the surface area of the sample. The results for three separately hot-pressed pellets (A1, A2, and A3) of the same material are shown in Table I. The scatter in the results are well within the acceptable error for measurements of this type. Comparison of A and B demonstrates the wide range of leachabilities encompassed by this test, while B1 and B2 demonstrate the reproducibility of the method for materials that are highly leachable. A total mass balance was not attempted because the number of elements of interest was limited. The apparent discrepancy in the mass balance is due in part to the ionic species that is

TABLE I
Short-Time Leach Test Data for Various
Crushed Pellet Waste Forms

Sample	Total Mass Loss (mg/g)	AAS Elemental Analysis (mg/g)		
		Na	Cs	Mo
A1	0.09	0.038	0.007	0.010
A2	0.14	0.016	0.006	0.005
A3	0.12	0.016	0.005	0.006
B1	15.4	1.7	4.7	5.3
B2	14.7	1.3	5.2	4.9
C	5.5	0.47	2.2	1.6
D (-Mo)	0.25	0.009	0.03	ND ^a
E	17.9	1.6	5.9	5.8
E+ 5% glass	6.2	1.0	0.82	2.5
E+ 30% glass	0.72	0.16	0.017	0.25

^aNot detected.

leached, i.e., molybdenum, which is present as molybdate compounds in the solid. Any other differences may be due to leaching of other anionic species, the loss of small particles through the filter, or experimental error.

The instantaneous leach test was also important for evaluating the effects of composition on the leaching behavior. Samples C and D in Table I demonstrate the effect of molybdenum on the leachability of cesium. Sample D is the same as sample C except that it contains no molybdenum. The E sample series demonstrated the effect of 5 and 30 wt% additions of borosilicate glass on sample leachability. The glass was added to serve as a consolidation aid.

It must be emphasized that the instantaneous leach test reveals nothing of the long-term or high-temperature behavior of a sample, which can be varied and complex. Also, due to the randomness of the sample and unknown surface areas, the data are not comparable to those of Hespe or any other standard leach test. However, the test has proven to be a highly reliable, self-consistent means for quickly screening and order ranking materials in a development program. Candidate materials can then be subjected to the more exhaustive, longer time leach tests.

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