DENATURED FUEL CYCLES

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This paper traces the history of the denatured fuel concept and discusses the characteristics of fuel cycles based on the concept. The proliferation resistance of denatured fuel cycles, the reactor types they involve, and the limitations they place on energy generation potential are discussed. The paper concludes with some remarks on the outlook for such cycles.

The history of the concept of denaturing nuclear fuel goes all the way back to the early efforts that were made to find acceptable ways of handling nuclear energy in the postwar world. It first appears in the "Report on the International Control of Atomic Energy," commonly referred to as the Acheson-Lilienthal Report.[1] This report was released March 28, 1946. The report was the work of a board of consultants to the Department of State. This board of five members, chaired by Mr. David E. Lilienthal, chairman of the Tennessee Valley Authority, and including Dr. J. Robert Oppenheimer, were charged with considering "the problems arising as to the control of atomic energy" and "to study the subject of controls and safeguards necessary to protect this Government ..."

The report brings in the concept of denaturing with the following sentence, "^{235}U and plutonium can be denatured; such denatured materials do not readily lend themselves to the making of atomic explosives, but they can still be used with no essential loss of effectiveness for the peaceful applications of atomic energy." The report does not define the denaturant, and is in fact somewhat enigmatic on the whole subject. For it goes on to say that "... another case of an operation that we would regard as safe ... is the development of power from the fission of denatured ^{235}U and plutonium in high power-level reactors. Such power reactors might operate in the range from one hundred thousand to one million kW. If these fissionable materials are used in installations where there is no additional uranium or thorium they will not produce further fissionable material. The operation of the reactors will use up the material." The last two sentences particularly, leave the reader wondering about the nature of the denaturant contemplated.

A clarifying press release was issued April 9, 1946. The release first noted that after consultation with the Department of State, Major General L. R. Groves called together a group, representative of the outstanding scientists connected with the Manhattan Project during the development of the atomic bomb, and stated that the group had just completed a conference in which the measure of safety afforded by the use of denaturants was discussed. Their report read as follows:

"The possibility of denaturing atomic explosives has been brought to public attention in a recent report released by the State Department on the international control of atomic energy. Because, for security reasons, the technical facts could not be made public, there has been some public misunderstanding of what denaturing is, and of the degree of safety that it could afford. We have thought it desirable to add a few comments on these points. The report released by the State Department proposes that all dangerous activities in the field of atomic energy be carried out by an international authority, and that operations which by the nature of the plant, the materials, the ease of inspection control, are safe be licensed for private or national exploitation. The report points out the possibility of denaturing explosive materials so that they do not readily lend themselves to the making of atomic explosives may contribute to the range of licensable activities, and to the overall possibility of the proposed controls. The report does not contend, nor is in fact true, that a system of control based solely on denaturing could provide adequate safety."

"As the report states, all atomic explosives are based on the raw materials uranium and thorium. In every case the usefulness of the materials as an atomic explosive depends to some extent on different properties than those that determine its usefulness for peacetime applications. The existence of these differences makes denaturing possible. *In every case denaturing is accomplished by adding to the explosive an isotope, which has the same chemical properties.*+ These isotopes cannot be separated by ordinary chemical means ..."

"For the various atomic explosives the denaturing has a different effect on the explosive properties of the materials. In some cases denaturing will not completely preclude making atomic weapons, but will reduce their effectiveness by a large factor. The effect of the denaturant is also different in the peacetime applications of the materials. Further technical information will be required, as will also a much more complete experience of the peacetime uses of atomic energy and into economics, before precise estimates of the value of denaturing can be formulated. But it seems to us most probable that within the framework of proposals advanced in the State Department report denaturing will play a helpful part."

So it is clear that it was isotopic denaturing, the same as is being considered once again today, that the authors had in mind. With the failure of the internationalization proposals over the next year or two, however, the concept of denaturing seems to have faded into the background as nuclear energy development proceeded.

Some 30 years later, in 1976, the notion was picked up again and expanded upon by Theodore Taylor and others at Princeton. First in their influential articles in the bulletin of Atomic Scientists,[2] and later in a monograph entitled, "Nuclear Proliferation,"[3] they proposed an international reactor deployment system wherein the main fissile material would be ^{233}U, bred from thorium, and diluted with the naturally abundant isotope ^{238}U to such an extent as to make the mixture unsuitable for weapons without isotope separation. At maturity the system would consist of two classes of facilities: national reactors and a few international fuel cycle support centers. The national reactors would operate on a fresh fuel mix of something like one part ^{233}U, six parts ^{238}U and 10 to 60

*This work performed under the auspices of the U.S. Department of Energy.
+Italics mine.

parts thorium. The six to one ratio ^{238}U to ^{233}U would probably be sufficient for practical purposes to denature the ^{233}U, so that the denatured uranium, both in the fresh fuel assemblies and the spent fuel would not be suitable for weapons material. There would be some plutonium in the spent fuel produced by neutron capture in the ^{238}U diluent but only somewhere between a fifth and a tenth as much as present fuels, and less a tenth as much in plutonium breeder fuel, for the same amount of power.

The international fuel cycle support center would reprocess all spent fuel from national reactors and would undertake all fabrication and denaturing of fresh fuel assemblies to be sent to the national plants."

The authors also however go on to note, "Since it is unlikely that the current types of nuclear power plants could breed sufficient amounts of ^{233}U to allow them to be self-sustaining on a thorium cycle if the recycle ^{233}U is denatured with ^{238}U, the internationally controlled centers at which the fuel is reprocessed would also have to provide some additional source of ^{233}U." And that "one possibility is fast breeder reactors that use plutonium extracted from the converter reactor fuels in their cores and thorium as the fertile materials in the breeding blankets. Such reactors could produce more ^{233}U than the plutonium they consume. All recycled plutonium could in this fashion consumed on site at the centers, avoiding the need for any national access to plutonium free from fission products."

On April 7, 1977, the Presidential nuclear power policy statement was released. It had as its third point, "We will redirect funding of the U.S. Research and Development programs to accelerate our research into alternative nuclear fuel cycles which do not involve direct access to materials usable in nuclear weapons." The studies set under way by new policy, in effect defined "alternative nuc fuel cycles" by the presence of denaturing. Alternative cycles in these studies were of two types. One type were variants based on ^{233}U/thorium cycles, and the other, variants of the current ^{235}U once-through cycle.

The possibility of plutonium denaturing was also implied in the Acheson-Lilienthal report, and has been raised again from time to time over the past year or two. Plutonium denaturing though has not received the attention that fissile uranium denaturing has been given. The reason is quite simple. Reactor produced plutonium is always a mixture of the fissile isotopes, ^{239}Pu and ^{241}Pu, and the fertile isotopes, ^{240}Pu and ^{242}Pu. The proportions of each are set largely by the reactor performance and fueling requirements, and in practical situations the fissile fraction will predominate. While high concentrations of ^{240}Pu in particular make the material less desirable for weapons purposes, it appears to be impossible to rule out its usefulness on the basis of normal reactor ^{240}Pu concentrations. Further, increasing the amount of ^{240}Pu and ^{242}Pu by selective repetitive recycle sharply limits the amount of such fuel and thus its applicability. A variant of the idea is to decrease the usability by attempting to increase the ^{238}Pu content -- the latter is a minor constituent of reactor-produced plutonium that has the property of relatively high heat generation. However, it is not produced in appreciable fractions in fast breeder fuel cycles, and as the great incentive for using plutonium derives from its use in the breeder it is difficult to assign high importance to the plutonium denaturing ideas.

Uranium-denatured fuel cycles have been examined extensively in the studies of the past year or so. Their general characteristics can be divided into three general classes. First, their proliferation resistance, second, the reactor types they involve, and third, their energy generation potential.

A. Proliferation Resistance

The entire reason for considering denatured fuel cycles is based on the hope of improving proliferation resistance. It is important therefore that the relative effectiveness for many decades in the future of isotopic denaturing for this purpose be rigorously examined. The fact that the fissile uranium isotopes can be mixed with the fertile isotope ^{238}U such that isotopic separation techniques are required to separate fissile material from fresh reactor fuel is considered to be the key property. As isotope separation techniques, for the present at least, are considered to be more difficult than chemical reprocessing techniques, an increase in proliferation resistance may be assigned. The degree of the increase is the important point. Centrifuge techniques are coming on the scene now and more advanced isotopics separation technologies are probable in the future. Whether, in the face of this, a significant advantage should be assigned to denaturing is the real question. In any event, the situation differs somewhat for the two basic denatured cycles.

Once-Through Cycles: In the once-through cycle, ^{235}U contents are no more than a few percent of the ^{238}U content in the fresh fuel, so that the isotopic denaturing present is more than sufficient. Problems exist, however, at both the front and back ends of the cycle. For LWR's, or in fact for any converter reactor except the heavy water reactor, enrichment facilities are required (and even heavy water reactors would benefit from enrichment). The presence of the enrichment facility gives the capability for separation of pure ^{235}U. Also, at the other end of the cycle, spent fuel, containing plutonium, accumulates and the burden is placed on arrangements for satisfactory retrieval (if in a sensitive location), safeguarding and storage of continuously increasing amounts of such fuel. Although spent fuel initially is highly radioactive, its activity decreases as time passes and after a period of years it becomes fairly accessible.

^{233}U Based Cycles: In any cycle that uses artificially created fissile isotopes to fuel reactors, reprocessing remains an essential component of the cycle and the ^{233}U cycle is no exception. The denaturant concentrations discussed in most studies involve ^{238}U contents about seven times the ^{233}U content. The uranium is then mixed with thorium. For startup, where ^{235}U is used, the denaturant concentration is normally taken to be four parts ^{238}U to one part ^{235}U. In converter reactors these mixtures allow thorium to remain the main fertile species in the fuel. Reactor operation produces ^{233}U from thorium but also produces plutonium from the ^{238}U denaturant. In the case of ^{233}U, some small amount of additional proliferation resistance may be assigned due to the presence of radiation from the decay of the ^{232}U that is always present in such fuel. However, the important point is that the reprocessing step remains a requirement in this cycle.

Further, the combination of the increased mass difference between $^{233}U/^{238}U$ and $^{235}U/^{238}U$, the smaller critical mass of ^{233}U than ^{235}U, and the feed materials starting at a 12% fissile level instead of a 3 or 4% fissile level, makes the separation of equivalent amounts of weapon usable material in the ^{233}U

cycle is at least an order of magnitude easier than it would be starting from current LWR fuel.

B. Reactor Types

The types of reactor that can effectively use denatured cycles follow directly from the basic nuclear properties of ^{235}U and ^{233}/Th: Thermal reactors are implied. This is not to say that fast reactors could not operate on a ^{233}U/Th cycle. In principle, of course, they could, but the basic nuclear properties of ^{233}U and thorium sharply limit breeding properties in a fast reactor, and the incentive to undertake the effort to develop and deploy the fast breeder under these circumstances would be correspondingly limited. Further, a fast breeder ^{233}U/Th cycle would require reprocessing efforts of a very similar kind to those required for the plutonium/^{238}U cycle. In effect, the only real change would be the substitution of ^{233}U for plutonium, but still in a very similar cycle and any gain from the viewpoint of non-separability of fissile material would certainly be small, and in all likelihood it would be non-existent.

Once-through cycles are still less suited to any version of the fast reactor that has yet been shown to be feasible. The higher initial enrichments of the fast breeder, compared to thermal reactors, and the correspondingly higher fissile contents discharged in spent fuel give net fuel utilizations that are much worse than those for the present day LWR, much less for any improved version. Mixes of reactor types that include some fast breeders and some thermal reactors operating on alternative cycles, termed "symbiotic cycles," are possible and from one class of suggested means for balancing energy production requirements and non-proliferation goals. But if denatured cycles alone are considered, unless there are breakthroughs in fuel irradiation technology, it is almost certain that thermal reactors alone are involved.

In thermal reactors once-through cycles tend to rule out thorium use. A once-through cycle implies ^{235}U fissile fueling, with thorium as the main fertile element in place of ^{238}U. The higher neutron absorption of thorium, a fundamental nuclear property, requires higher initial enrichments to maintain reactor operation. In any calculation of the resulting fuel utilization in practical thermal reactor types that I am aware of, the resulting fuel utilization is invariably worse than for the corresponding $^{235}U/^{238}U$ once-through cycle. The reason is the same as in the fast reactor case, although to lesser degree. In both the fissile contents in the discharged fuel remain high and too much fissile material is discarded in the spent fuel.

The reference to breakthroughs in fuel technology was made for the following reason. The only way that the once-through cycle even in concept could overcome the limitations just described would be through breakthroughs in fuels and materials technology to allow extremely long fuel burnups. The same principle would hold for both thermal reactors using thorium as its fertile material or a fast reactor fueled with ^{235}U and using ^{238}U as its fertile material. Very long burnups could in principle convert substantial quantities of fertile material to fissile (thorium to ^{233}U to plutonium in the fast reactor case) and then burn a large amount of it in place. Enough would have to be burned in place to make up for the large amount of fissile that would be discarded in the spent fuel. In either a fast or a thermal reactor version the basic design problem would be similar. First, the conversion or breeding properties would need to be as high as possible (to efficiently convert fertile to fissile) and second, the burnups or reactor residence times would need to be as long as possible in order to extract sufficient energy from the bred fissile that the effects both of high initial inventory requirements and high fissile contents in the spent fuel would be overcome. These two design requirements tend to conflict. Further, in the fast reactor case at least, the irradiations that the fuel would have to undergo are far out of the range of any existing experimental data indicating feasibility.

Thus apart from reactor concepts based fundamentally on speculations of very long burnups, the only way that substantial gains in fuel utilization are possible is by reprocessing. This is specifically true for the ^{233}U/thorium cycle in thermal reactors.

With such reprocessing, substantial gains in fuel utilization over the current once-through cycle are possible. Net ^{235}U consumptions half of those of the current once-through cycle are probably easily achievable, and consumptions down to a quarter of the current once-through cycle are possible, at least in principle. In all these denatured fuel reactor designs there is a trade-off between fuel utilization, on one hand, and inventory and burnup on the other, and therefore in the amount of reprocessing required annually. In all, reprocessing is fundamental.

C. Energy Generation Potential

The most fundamental point of concern about the alternative cycles is the limitation they impose on the amount of energy that can be produced by nuclear power. Denatured cycles are net consumers of fissile material and their use alone implies a net drain on the world supply of fissile material. Put another way, the amount of nuclear power production possible is more or less directly dependent on the world supply of natural uranium. Thus in a fundamental sense, the alternatives to the reference uranium-plutonium cycle can be said to reflect alternative views of the necessary or desirable role of nuclear power. If a fairly limited role for nuclear power is envisaged denatured cycles may well suffice. On the other hand, if the magnitude of nuclear power production is not to be limited by the uranium resource base, true breeding is necessary and the uranium-plutonium cycle is almost undoubtedly required.

Another general point, useful to keep in mind, is that the three main reactor deployment possibilies -- the once-through cycle, the denatured ^{233}U/Th cycle, and the breeder reactor -- are, by their inherent characteristics, explicitly suited to one or other quite specific scenario for the future of nuclear power. Each reactor deployment strategy will suffer by comparison when applied to an energy scenario that is specifically suited to one of the other alternatives. It is important in assessing the meaning of comparisons of the resource utilization efficiency of various alternative reactor development strategies that this be understood. Simply, the breeder shows to best advantage in scenarios that contemplate exponential rises in nuclear electrical energy production and an essentially unlimited future; the denatured ^{233}U/Th cycles show to best advantage in scenarios that contemplate a leveling off in nuclear power production after a period that can include a relatively rapid rise, and once-through cycles are suited to scenarios that contemplate nuclear power production as a passing phase, possibly of considerable length, but nevertheless eventually phasing out.

The scenario that is seen in some recent policy-related papers is the second one, that is, the denatured cycle nuclear power leveling-off scenario. The characteristics of thermal reactors using denatured $^{233}U/Th$ cycles, i.e., rather high initial inventory requirements, but not as high as the fast breeder, and relatively small refueling requirements, are specifically suited to this scenario. Put another way, if one wishes to make a case for these cycles, the scenario for nuclear power that one should chose is such a plateau scenario. The breeder, when applied to this scenario, suffers greatly in the comparison, as a relatively rapid rise in nuclear capacity penalizes the breeder through its high inventory requirement and a short time period for rising capacity does not allow time for the breeding characteristics of the system to be of much help. Further, when sufficient time has passed that the doubling characteristics of the breeder are starting to become important such a scenario contemplates a leveling off of nuclear power production so the excess fissile material generated by the breeder is of little importance.

D. Outlook for Denatured Cycles

The rationale for denatured cycles rests on their possible application to increase in proliferation resistance. Most I think would agree that isotopic separation at the moment is more difficult, and may remain somewhat more difficult in the long run, than chemical reprocessing. For this reason the fissile content of denatured fuel is likely to be somewhat less accessible than plutonium contained in uranium. The magnitude of this difference and the importance to be assigned to it, however, is of course, the difficult point. I think most now would agree that insofar as the fresh fuel is concerned, standard LWR fresh fuel should be assigned somewhat more proliferation resistance than ^{233}U denatured fuel and it in turn somewhat more than plutonium/^{238}U. The differences, however, taking into account the entire fuel cycle, are probably not very significant when compared to alternative institutional arrangements for nuclear power deployment. This last statement would not be agreed to by everyone, but I think one would find nearly unanimous agreement to it in the reactor technical community.

Denatured cycles, if they are to lead to significant improvements in uranium resource usage over the present once-through cycle, must have reprocessing for any reactor types that have currently been shown to be feasible. Without reprocessing, fundamental breakthroughs in fuels and material technology to allow extremely long fuel burnups would be required for even conceptual feasibility. As long as reprocessing is present in any case, the denatured cycles offer little obvious advantage over Pu/^{238}U cycles.

The denatured cycles all impose a penalty on the amount of energy that can be produced from given uranium resources. The denatured cycles themselves are consumers of fissile material and their use alone implies a net drain on the world supply of fissile material. Thus they reduce the energy generation potential and to make up for this they must have a very substantial advantage in proliferation resistance. Because this does not appear to be the case, the future of the denatured cycle based on ^{233}U thorium does not appear to me to be very bright. My suspicion is that the thorium cycle, when and if it arrives, will come because of its technical merits and the wish to utilize the thorium resources rather than on the basis of superior proliferation resistance. The current once-through cycle, of course, will continue for some time. But even with postulated improvements in the LWR, and reduction in the isotopic tails assays, the period over which the once-through can satisfy world nuclear energy needs is likely to be of the order of the lifetime of existing reactors, unless large new supplies of uranium are discovered. Thus although it may still be too early to say for certain, there is little to point to that would suggest any gathering momentum in the direction of denatured cycles.

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