

Nuclear Fission Energy Fuel Cycle Options

ISD-A
362

D. A. Freiwald

Refer requests for this report
to the Author

NOT CIRCULATE

PERMANENT RETENTION

REQUIRED BY CONTRACT

University of California



LOS ALAMOS NATL. LAB. LIBS.
3 9338 00191 0776

LOS ALAMOS SCIENTIFIC LABORATORY

Post Office Box 1663 Los Alamos, New Mexico 87545 505/667-5061

An Affirmative Action/Equal Opportunity Employer

UNITED STATES
DEPARTMENT OF ENERGY
CONTRACT W-7409-ENG. 36

Permission to reproduce this article is granted.

INTRODUCTION

As world reserves of conventional energy sources decrease (see Fig. 1), we must develop alternate sources. During the transition period until ultimate sources are developed, nuclear fission and coal are the most likely sources. The question marks in Fig. 1 indicate that it is not known to what extent coal and nuclear will be used to close the transition-period energy gap, because economic, environmental, and sociopolitical considerations are still being debated.

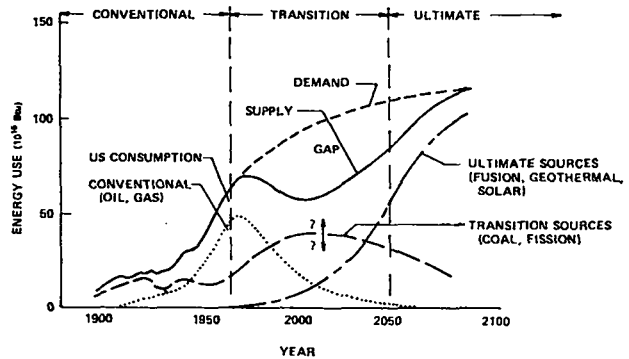


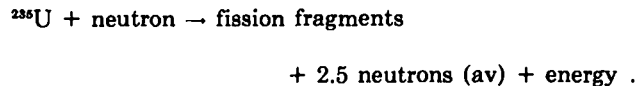
Fig. 1.

Probable changes in the energy source mix for the United States.

In this mini-review we focus on nuclear fission energy and discuss the present cycle as well as several options that have not been fully explored.

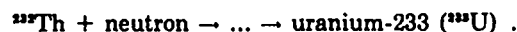
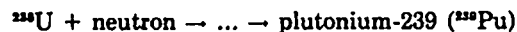
NUCLEAR FISSION

In the fission reaction, if certain kinds of heavy atoms (called *fissile fuels*) such as uranium-235 (^{235}U) capture a stray *neutron*, they split into lighter atoms (called *fission fragments*) and emit more than one neutron of their own in the process, along with considerable energy. Expressed as a formula, this reaction is



The emitted neutrons in turn can be captured by other ^{235}U atoms, thus causing additional splits and releasing more and more neutrons. Repeated capturing and splitting is called a *chain reaction*. It is sustained if there are enough fissile fuel atoms close enough together that the emitted neutrons will be recaptured. Having enough fissile atoms close together is called *having a critical mass*.

Sometimes the emitted neutrons are captured by different kinds of heavy atoms such as uranium-238 (^{238}U) and thorium-232 (^{232}Th), which aren't readily fissionable. These atoms, now altered, can be used as fissile fuel; the process of creating them is called *converting*, or *breeding*. Expressed as formulas, these reactions are



REACTORS

A reactor that simply "burns" fissile fuels such as ^{235}U is called a *burner*. A reactor that also has atoms like ^{238}U and ^{232}Th in or near its core, so that those atoms can be bred into fissile fuel, is called a *converter*. If the conversion proceeds so well that more fuel is produced than is consumed, the reactor is then called a *breeder*. At present, the 68 commercial reactors in the United States are only burners of ^{235}U . Some ^{239}Pu is produced in them, but the reactor designs aren't optimized for breeding, nor is the ^{239}Pu currently *recycled*.

TODAY'S NUCLEAR FUEL CYCLE

The cross-hatched pathway in Fig. 2 shows the present nuclear fuel cycle. Naturally occurring uranium is found in ores as a mixture of ^{238}U (99.3%) and ^{235}U (0.7%). At the beginning of the fuel cycle, the ore is mined, milled, leached with chemicals, and processed into a form called *yellow cake*. The waste products of this process, called mill tailings, (about 99% of the original ore mass) currently are disposed of in tailings ponds (called tailings piles in the industry).

The yellow cake (1 lb produces about the same amount of energy as 7.5 tons of coal) is shipped to *conversion plants* where it reacts chemically with fluorine to form the gas uranium hexafluoride (UF_6). The slight difference between the weight of the $^{238}\text{UF}_6$ molecules and the $^{235}\text{UF}_6$ molecules enables enhancement of the $^{235}\text{UF}_6$ concentration of the mixture in the next step of the process.

The historical technique for ^{235}U *enrichment* is called *gaseous diffusion*, but other techniques such as the *centrifuge* process are being developed, and new techniques using *lasers* are under research. The United States Government controls all domestic ^{235}U enrichment.

The amount of fuel enrichment done depends on the need or reactor type for which the $^{235}\text{U}/^{238}\text{U}$ will be used. In general, the higher the concentrations of ^{235}U (the fissile part) the more flexibility there is in reactor design, but most current reactors need fuel enriched to only about 3%. The fluorine in the enriched UF_6 mixture is then

chemically replaced with oxygen, and the enriched uranium oxide is fabricated into *fuel pins* clad in metal.

The byproducts of the enrichment process are called *uranium enrichment tails*, depleted in ^{235}U (conversely, enriched in ^{238}U). These are currently stored for possible use in a breeder reactor in the future. If the enrichment tails in storage now were bred into ^{239}Pu , the energy from burning the ^{239}Pu could supply this country's electrical needs for many decades.

At the reactor, the fuel pins are placed into rods, and enough fuel rods, along with neutron-absorbing control rods, are inserted into the reactor core, close enough together so that a critical but manageable mass of fissile fuel results. The nuclear chain reaction begins, liberating neutrons, fission fragments, and energy. The fission fragments remain trapped inside the fuel pins. The released energy is used to heat water, converting it into steam that turns turbines for generating electricity.

As the fuel "burns," it becomes depleted in ^{235}U , and eventually the ^{235}U concentration becomes too low for effective reactor operation. The fuel pins must be replaced about every three years, usually one-third of them every year.

The "spent" fuel pins still contain some ^{235}U , as well as fission fragments, ^{238}U , and some ^{239}Pu bred from ^{238}U . If the pins were *reprocessed*, the ^{235}U and ^{239}Pu could be reused as fuel, and the ^{238}U could be used in a breeder to breed more ^{239}Pu . However, reprocessing isn't currently done in the United States.

Instead, the spent fuel pins are sent to *interim storage*. They are called reactor nuclear waste,* but as mentioned above, that is a poor choice of words because of the valuable ^{235}U , ^{238}U , and ^{239}Pu content of the pins. Storage is a problem, however, because the pins are radioactive. The ^{239}Pu has a decay time of thousands of years. Fission fragments most dangerous to man, strontium-90 (^{90}Sr) and cesium-137 (^{137}Cs), have decay times of hundreds of years.

Research is underway on the suitability of deep underground sites for *permanent storage* of nuclear fuel cycle byproducts. They would likely be stored in solid form, for example melted with and solidified into glass—a process called *vittrification*.

The discussion above outlined the nuclear fuel "cycle" that is now used in this country (the cross-hatched pathway in Fig. 2). However, in fact it isn't a cycle—it is a one-pass system. It would be a cycle, though, if spent fuel pins were reprocessed.

OPTIONS

Figure 2 shows that there are other potential pathways for developing nuclear fission energy.

*In the broadest sense, nuclear waste also includes mill tailings, weapons program refuse, etc. Some of that waste is chemically treated before being put into interim storage.

LOS ALAMOS NATIONAL LABORATORY
 3 9338 00191 0776

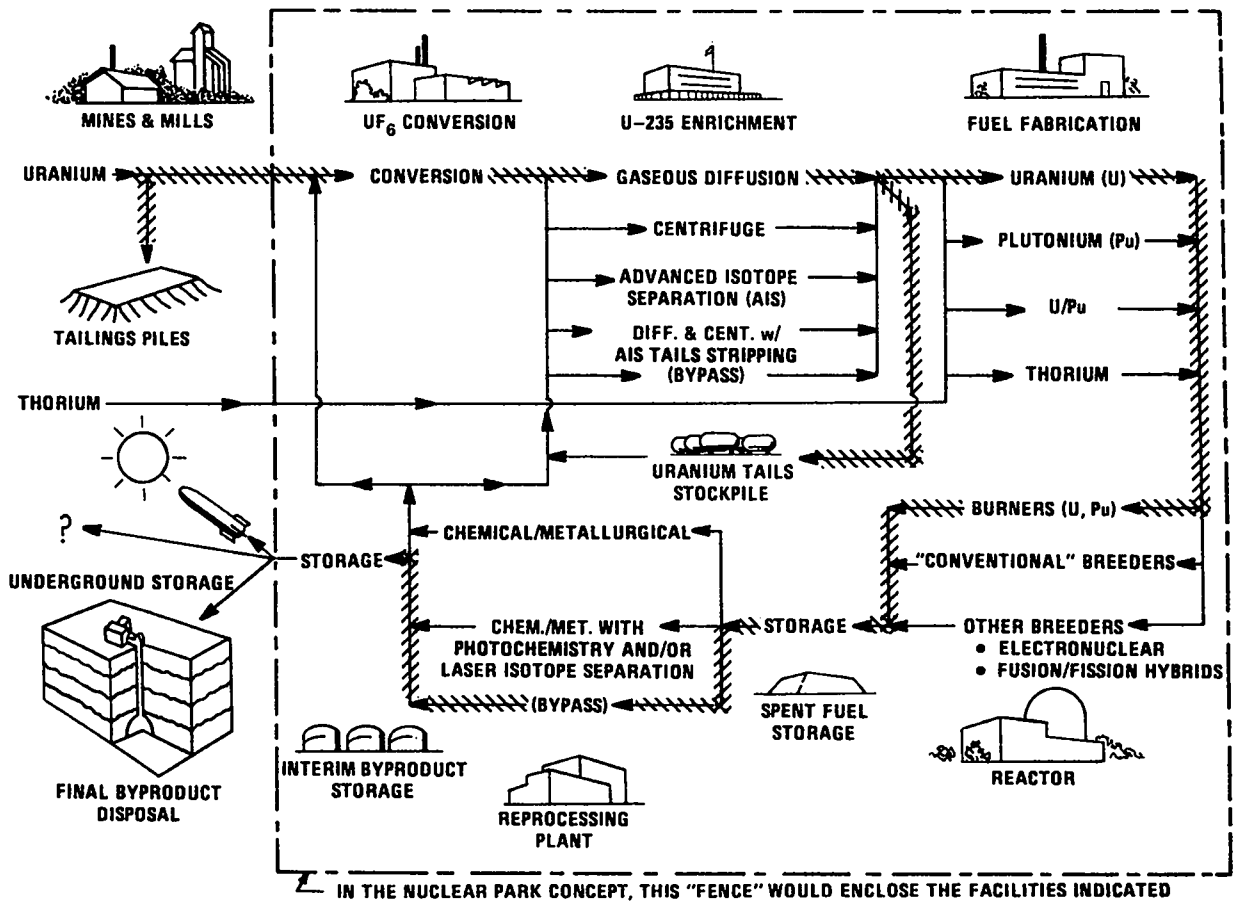


Fig. 2. Possible nuclear fission energy, fuel cycle pathways.

With continued mining of raw high-grade uranium ore, our supplies will be depleted in just a few decades. Even without breeding, one pathway is the use of reprocessed ^{235}U and ^{239}Pu , separately or combined as so-called *mixed oxide fuels* (MOX), in burners. Reprocessing would save between 20 and 40% on the amount of raw uranium that would need to be mined for a reactor. Reprocessing would also change the mixture of byproducts put into permanent storage, because most of the ^{235}U , ^{238}U , and ^{239}Pu would be removed for reuse. A chemical reprocessing method, called *PUREX*, recovers 99.9% of the uranium and plutonium, but it is now used only on nuclear weapons program byproducts, not commercial reactor byproducts.

Use of breeders to create ^{239}Pu from ^{238}U , or ^{233}U from ^{232}Th , is another option that could supply the United States with fission/electric power for centuries.

The term "conventional" breeder in Fig. 2 is a misnomer in that this country doesn't have a "ready to order" ^{238}U

breeder yet; further research and development is needed before breeders become a real commercial option.

Two other potential (but not commercially real) breeder options are shown in Fig. 2. Electronuclear breeding would use accelerated hydrogen atoms to knock neutrons out of targets, and the neutrons then would bombard ^{232}Th to breed ^{233}U . The conceptual fusion/fission hybrid would use neutrons from fusion reactions to cause fission reactions and additional energy release in a sub-critical fissile-fuel blanket surrounding the fusion core.

The kind of reactor used (burner, breeder, hybrid) would dictate the pathways in Fig. 2 up to the reactor—the so-called *front end* of the cycle—and would, in part, dictate the pathway used after the reactor—to so-called *back end* of the cycle.

Note the options currently bypassed in the back end of the cycle. As mentioned, chemical/metallurgical reprocessing would provide substantial fuel recovery. But

what about the other radioactive fission fragments? One option is *laser photochemical separation* (a technique that allows activation and separation of just one kind of atom or molecule in a mixture of several kinds), which may provide:

- Separation of uranium and plutonium from liquid waste for fuel recycling.
- Separation of long-lived fission products, such as iodine-129, zirconium-93, and technetium-99, that could be returned to a reactor where nuclear *transmutations* would reduce them to stable nonradioactive isotopes.
- Separation of heat-generating radioactive elements, such as ⁹⁰Sr and ¹³⁷Cs. Removal would reduce the heat load of the remaining byproducts. Such isolated elements might profitably be used to supply *process heat* (heat for an industrial process) during their decay (half-life about 30 years). Also, research is underway to use ¹³⁷Cs to irradiate municipal sewage sludge to make the sludge biologically inactive and thus suitable for fertilizer.
- Recovery of valuable metals such as rhodium and palladium that could be sold.

If laser photochemical separations were done, the remaining toxic wastes could be much more easily stored; after 700 years, less than one ten-millionth of the initial toxicity would remain.

Laser photochemistry is a new field, and little work has been done on the use of lasers at the back end of the cycle. This option needs further study.

In the meantime, these fuel cycle back-end wastes should be viewed as potentially very valuable, and perhaps a "permanent" storage facility should be designed for retrieval.

ISSUES

As we use more fission energy, we must consider these issues.

- **Mill Tailings.** Environmental problems and potential solutions have been identified, but more research and development is necessary.
- **Transportation.** Transport of nuclear material is currently necessary between each facility in Fig. 2. There is some concern about the material's susceptibility to terrorists and its environmental impact in an accident; but there has been much research on material transport in recent years, and solutions to potential problems are at hand.
- **Economics.** The economics of a fission system changes as the world energy-economic situation changes. It is under study and should be considered with desires for energy-economic independence for long-term national

- **Safeguards.** Safeguards is the protection and accountability of nuclear materials as they move through the cycle. The subject is under intensive research and development, with upgraded prototype systems now in use. The nuclear park concept (see Fig. 2) could also address safeguards and transportation concerns.
- **Safety.** The perceived (but not necessarily real) issue here is reactor safety. Accident deaths per year for the population of the entire United States are about 111,000. It has been estimated that with 100 nuclear power plants operating, there might be an additional two deaths per year from reactor accidents and two per year from routine emission of radiation. However, none have been reported yet even though we have 68 commercial reactors in operation.
- **Terrorism.** The transportation and safeguards aspects have been discussed. The main remaining concern is sabotage of a nuclear facility. Operating plants have increased security, including physical barriers and electronic detection.
- **Proliferation.** The underlying concern here is that development of the technology and possible subsequent sales to foreign countries may result in those countries being able to build a nuclear weapons program of their own, using reactors to breed weapon-grade material. However, they can develop the technology (albeit expensive) themselves or acquire it from countries other than the United States. Those countries could also pose an international threat using much less expensive chemical or biological weapons that are much more difficult to detect than nuclear weapons.
- **Fuel Cycle Byproducts.** Their processing, transporting, and storage have been discussed.

COMMENTS

Today's energy sources for the United States are the fission burner reactor, plus fossil energy supplies and a small amount of hydroelectric power. It will be several decades before ultimate sources such as fusion, solar, and geothermal are sufficiently developed that they have noticeable impact on our energy supplies (see Fig. 1).

To fill the energy gap in the transition period (which may last longer than indicated in Fig. 1) when oil and gas supplies are diminishing and ultimate sources are developing, this country has three options: coal, nuclear fission, and conservation. It is now generally agreed that coal alone can't fill the gap. We should conserve our energy no matter what its source, but the United States will still have base needs for high-grade energy.

LASER
CLASSIFIED
REPORT
JUN 16 1974
RECEIVED

Mini-Review
readers are encouraged
to correspond directly
with the author.