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INTRODUCTION

WHAT FAST REACTORS CAN DO

Chain Reactions

Early in 1939 Meitner and Frisch suggested that the correct interpretation of the results observed when uranium is bombarded with neutrons is that the uranium nuclei undergo fission. Within a few months two very important things became clear: that fission releases a large amount of energy, and that fission of a nucleus by one neutron liberates usually two or three new neutrons. These discoveries immediately disclosed the possibility of a chain reaction that would produce power.

There was a difficulty, however, in making a chain reaction work. Natural uranium consists of two isotopes: 235 U (with an abundance of 0.7%) and 238 U (99.3%). Of the two only 235 U is "fissile", meaning that fission can be induced in it by neutrons of any energy. On the other hand 238 U undergoes fission only if the neutrons have an energy greater than about 1.5 MeV, and even then they are more likely to be captured or scattered inelastically.

Figure 1 shows the fission cross-sections of ²³⁵U and ²³⁸U and the capture cross-section of ²³⁸U as functions of neutron energy. Because ²³⁸U is so abundant in natural uranium capture in it dominates over fission in ²³⁵U except at energies below about 1 eV.

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Figure 1 Cross-sections of ²³⁵U and ²³⁸U.

Neutrons generated in fission have average energies of about 2 MeV and at that energy cannot sustain a chain reaction in natural uranium. If a neutron survives many scattering interactions, however, its kinetic energy decreases until it is in thermal equilibrium with the atoms by which it is being scattered. It is then known as a "thermal" neutron and its most probable energy is about 0.025 eV.

If a chain reaction is to take place, therefore, either the fission neutrons have to be reduced in energy to near the thermal level, in which case natural uranium can be used, or the proportion of ²³⁵U has to be increased substantially. Both of these routes were followed in the early work on nuclear reactors. The first led to the development of "thermal" reactors and the second to "fast" reactors, so called because the neutrons causing fission are fast as opposed to thermal.

Breeding and Consumption

It is possible to make use of the neutrons that are not needed to maintain the chain reaction in various ways. The most important is for Introduction

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breeding. When a neutron is captured in ²³⁸U the ²³⁹U that is formed decays in the following way:

$$^{239}\text{U}\frac{\beta}{23.5\,\text{min}} \to ^{239}\text{Np}\frac{\beta}{2.35\text{d}} \to ^{239}\text{Pu}\frac{\alpha}{24360\text{y}} \to ^{235}\text{U}$$
 etc.

The times shown are the half-lives for the decay processes. As far as reactor operation is concerned the long-lived plutonium isotope ²³⁹Pu is the end-product of the chain.

²³⁹Pu has nuclear properties quite similar to those of ²³⁵U and it can be fissioned by neutrons of all energies. Neutron capture thus provides a route for converting ²³⁸U into fissile material, so ²³⁸U is called a "fertile" isotope. ²³²Th, which is the only naturally occurring isotope of thorium, is also fertile. It behaves very similarly to ²³⁸U: the ²³³Th formed on capture of a neutron decays in a chain to ²³³U which is long-lived and fissile.

$$^{232}\text{Th}\frac{\beta-}{22.1\,\text{min}} \rightarrow ^{233}\text{Pa}\frac{\beta-}{27.4\text{d}} \rightarrow ^{233}\text{U}\frac{\alpha}{162000\text{y}} \rightarrow ^{229}\text{Th}$$
 etc.

Thus there are two naturally occurring fertile isotopes, ²³²Th and ²³⁸U, and three related fissile isotopes: ²³³U, ²³⁵U and ²³⁹Pu. There are other fissile and fertile isotopes but these five are the most important.

This ability to convert fertile isotopes to fissile raises the possibility of "breeding" new fissile material, but this can be done only if enough neutrons are available. The average number of neutrons liberated in a fission is denoted by $\overline{\nu}$. Its value depends on which isotope is being fissioned and on the energy of the neutron causing the fission, but in most cases it is about 2.5. We have seen that the fact that $\overline{\nu}$ is greater than 1 makes a chain reaction possible: the fact that it is greater than 2 is almost equally important. If we have a reactor in which on average one neutron from each fission causes another fission to maintain the chain reaction, and if in addition more than one of the other neutrons is captured in fertile material, then the total number of fissile nuclei will increase as the reactor operates. Such a reactor is called a "breeder".

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It is sometimes said that a breeder reactor generates more fuel than it consumes. This is rather misleading. The reactor produces more fissile material than it consumes, but to do this it depends on a supply of fuel in the form of fertile material.

Although $\overline{\nu} > 2$ suggests the possibility of a breeder reactor the requirement for breeding to take place is more complicated. When a neutron interacts with a fissile nucleus it does not necessarily cause fission. It may be captured, and if it is, it is effectively lost. The important quantity in determining whether breeding is possible is the average number of neutrons generated per neutron absorbed. This is denoted by η , where

$$\eta = \overline{\nu}\sigma_f / (\sigma_f + \sigma_c).$$

 η , which is sometimes called the "reproduction factor", is a function of the neutron energy *E*, and its variation with *E* for the three fissile isotopes is shown in Figure 2.

Of these η neutrons one is needed to maintain the chain reaction. Some of the remainder are lost either because they diffuse out of the reactor or because they are captured by some of the other materials present, such as the coolant or the reactor structure. The others are available to be captured by fertile nuclei to create fissile nuclei. If we denote the number of neutrons lost per neutron absorbed in fissile material by L and the number captured in fertile material by C, then C is the number of fissile nuclei produced per fissile nucleus destroyed and is given by

$$C \simeq \eta - 1 - L$$

(This is only a rough value because there are other things that may happen to neutrons).

If C is greater than one, as it must be if the reactor is to breed, it is known as the "breeding ratio". If it is less than one it is called the "conversion ratio". There is no logical reason for the existence of two names for C. The usage grew up because different words were used in the contexts of different reactor systems. Cambridge University Press & Assessment 978-1-107-03464-8 — An Introduction to the Engineering of Fast Nuclear Reactors Anthony M. Judd Excerpt More Information



Figure 2 The reproduction factor, η , for ²³³U, ²³⁵U and ²³⁹Pu.

In practice L cannot be reduced below about 0.2, so that breeding is possible only if η is greater than about 2.2. Figure 2 shows how this can be brought about. A fast reactor using any of the three fissile materials can be made to breed, although ²³⁹Pu gives the widest margin and ²³⁵U will allow breeding only if the energy of the neutrons causing fission is not allowed to fall much below 1 MeV. In all cases the higher the neutron energy the better the breeding ratio. A ²³³U-fuelled thermal reactor is just able to breed but the margin is very slender. The most widely favoured breeder system is based on the use of ²³⁸U and ²³⁹Pu in fast reactors, but there is also a certain amount of interest in the ²³²Th – ²³³U system, also in fast reactors.

A fast reactor does not necessarily have to be a breeder. The excess neutrons can be used in other ways. One such is to use them to consume radioactive waste materials by transmutation. This process can be applied to two classes of radioactive waste: fission products and

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"higher actinides" (i.e. nuclides with atomic numbers greater than 94). In both cases it is the most long-lived nuclides that are of interest because of a perceived difficulty in ensuring the integrity and safety of waste storage facilities over the very long periods, up to a million years, for which the waste remains dangerous. Fission products such as ⁹³Zr, ⁹⁹Tc, ¹²⁹I and ¹³⁵Cs have half-lives of the order of 10⁶ years, as does the actinide ²³⁷Np.

In most cases it is not possible to transmute any of these nuclides into stable isotopes. However, the fission products can be made less hazardous by transmuting them into other radioactive materials with shorter half-lives. Higher actinides can be eliminated by fissioning them. Some of the fissile higher actinides can be used in effect as nuclear fuel, and in all cases benefit can be taken of the energy released when they are fissioned.

It is also possible to envisage a fast reactor that, instead of breeding, acts to consume fissile material. In this way it may be possible to use a fast reactor to eliminate unwanted stocks of weapons-grade plutonium.

Energy Resources

Even if fast reactors are used to consume radioactive waste in due course their main function is likely to be to breed fissile material because in this way they can have a transforming effect on the world's energy resources.

Consider a uranium-fuelled reactor in which N atoms of ²³⁵U are fissioned. While this is happening CN new fissile atoms of ²³⁹Pu can be produced. If these in turn are fissioned in the same reactor and the conversion or breeding ratio C is unchanged (this is unlikely to be quite true because the fissile material has been changed, but the effect on the argument is not important), a further C^2N fissile atoms are produced. If these are fissioned, C^3N are produced, and so on indefinitely. The total number of atoms fissioned is therefore $N(1 + C + C^2 + ...)$. If C < 1, the series converges and its sum is N/(1-C). Introduction

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Conversion ratios for 235 U-fuelled thermal reactors are in the range 0.6 (for light-water reactors) to 0.8 (for heavy-water reactors and gascooled reactors). *L* is particularly large in light-water reactors because neutrons are readily absorbed by hydrogen.

If the fuel is natural uranium N cannot exceed 0.7% of the total number of uranium atoms supplied. If the reactor is a thermal reactor with a conversion ratio of 0.7 and the plutonium bred is recycled indefinitely the total number of atoms fissioned cannot exceed $0.7/(1-0.7) \approx 2.3\%$ of the number of uranium atoms supplied.

In a real system not even this number can be fissioned. When the fuel is reprocessed to remove the fission products and the excess ²³⁸U some ²³⁵U is inevitably lost. In addition some ²³⁹Pu is lost by conversion to higher isotopes of plutonium. As a result thermal converter reactors can make use of at most about 2% of natural uranium.

For a breeder reactor, however, with C > 1, the series diverges and in principle all the fertile atoms supplied can be fissioned. In practice, however, some are lost for the reasons mentioned earlier and the limit is around 60% of the fertile feed. Thus from a given quantity of natural uranium fast breeder reactors can fission about 30 times as many atoms as thermal converters and as a result can extract about 30 times as much energy.

To determine the importance of this difference we have to know how much uranium and thorium are available. The amount depends on the price, and a 2010 estimate by the World Energy Council suggests that, worldwide, about 230000 tonnes of uranium are recoverable at a price up to \$40/kg, but that if the price were to rise to \$260/kg ten times as much would be accessible. The extent of reserves of thorium is much less certain but seems to be comparable with those of uranium. Thorium can be made available as an energy resource only by means of breeder reactors.

Complete fission of a tonne of uranium, were that possible, would generate about 1 TWd, or 0.09 EJ, of energy in the form of heat. (An exajoule, EJ, is 10^{18} joules.) Thus if all the \$40/kg uranium in

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the world were used as fuel for thermal reactors that, with recycling, fissioned 2% of the feed, some 400 EJ thermal would be produced. If the same uranium were to be recycled to exhaustion in fast breeder reactors it would produce about 12000 EJ. But if the higher utilisation would allow the higher price of \$260/kg to be paid so that the greater resource became available the production would rise to 1.2×10^5 EJ. These quantities can be compared with about 9.0×10^{11} tonnes of "proved recoverable" coal reserves that could yield some 3000 EJ, or 1.6×10^{11} tonnes of "proved recoverable" oil that could yield about 800 EJ. In 2007 some 71 EJ of electricity was generated throughout the world.

There is considerable uncertainty about the true extent of mineral reserves in the earth's crust because new discoveries continue to be made. However, in spite of this the overall conclusion is that uranium used in thermal reactors has the potential to make a contribution to the world's energy consumption that is comparable with, but smaller than, that of oil, whereas uranium used in fast breeder reactors could contribute considerably more than, possibly 40 times as much as, all the world's fossil fuel. Thorium used in breeder reactors could probably make a similar contribution. Together they could provide the world with all the energy it needs for centuries to come. And they would do this without adding to the amount of carbon dioxide in the atmosphere.

HOW FAST REACTORS HAVE BEEN DEVELOPED

The Early Years

The history of fast breeder reactors is quite dissimilar from that of thermal reactors. From the earliest days after the Second World War the development of different types of thermal reactor was pursued in different countries: light-water reactors in the United States, heavy-water reactors in Canada and gas-cooled reactors in the United Introduction

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Kingdom, for example. Only towards the end of the 20th century did the various nationally based lines of development converge.

In contrast virtually the same path was followed in all the countries where work on fast reactors was done. The reason for this seems to have been that until the 1960s at least fast reactors were seen to be commercially valuable only well into the future, so that the advantages of cooperation appeared to outweigh the disadvantages of aiding possible competitors. Thermal reactors on the other hand were commercially important from the start and were developed in competition, which restricted the exchange of ideas and allowed different concepts to flourish.

International cooperation played a major role in fast reactor development for two main reasons. Firstly the nuclear data on which designs had to be based were inadequate until the 1960s. There was a lot to be gained from worldwide cooperation in measuring neutron crosssections to the required accuracy and exchanging and comparing the results. Secondly cooperation to ensure the safety of fast reactors was desirable even when there was competition in other areas.

This need to exchange information resulted in, among other things, a series of international conferences on fast reactors that were addressed mainly to the problems of reactor physics and safety and were held in the United States and various European countries throughout the 1960s and 1970s. These, together with the continual publication of information in the scientific press, kept the thinking in different countries from diverging and encouraged parallel development.

In one respect it is not altogether certain that this was an advantage. The use of liquid metals as coolants acquired a great deal of momentum, mainly because "everyone did it", and the search for alternatives was discouraged. Gas has certain advantages as a coolant, but at the time of writing no group has been able to develop a gascooled fast reactor to the point where it can be assessed fairly in comparison with a liquid-metal-cooled reactor.

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The Era of Metal Fuel

Before about 1960 it was thought that a high breeding ratio was the most important quality of a fast reactor. To achieve this the mean energy of the neutrons has to be kept high, and this requires that extraneous materials, especially moderators, should be excluded as far as possible from the reactor core. As a result the early reactors had metal fuel, the metal being either enriched uranium or plutonium, alloyed in some cases with molybdenum to stabilise it to allow operation at higher temperature.

The critical masses of these reactors were small and the cores were correspondingly small so that for high-power operation they had to be cooled by a high-density coolant (to avoid impossibly high coolant velocities). Hydrogenous substances were precluded because hydrogen is a moderator, so liquid metals were used. In most cases the coolant was sodium or sodium-potassium alloy. Some early experimental reactors were cooled with mercury but this fell out of favour because of its toxicity, cost, and low boiling point.

The many neutrons that leaked from the small cores of these reactors were absorbed in surrounding regions of natural or depleted uranium where the majority of the breeding took place. These were known as breeders, or blankets.

The first generation of low-power experimental fast reactors were built in the late 1940s and early 1950s to demonstrate the principle of breeding and to obtain nuclear data. They included CLEMENTINE and EBR-I in the United States, BR-1 and BR-2 in the Union of Soviet Socialist Republics, and ZEPHYR and ZEUS in the United Kingdom. CLEMENTINE, ZEPHYR, and BR-1 and 2 used plutonium fuel, which in the early years was more readily available than highly enriched uranium. Apart from ZEUS, which was a zero-power mockup of the later DFR, they all had very small cores, the largest being EBR-I (6 litres), which was a small power reactor with an output of 1.2 MW.