W.B. LEWIS MEMORIAL LECTURE

THE FUTURE NUCLEAR VISION

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Abstract

We are on the threshold of unprecedented changes in the global nuclear community. The various factors affecting these changes can be classified into the following main drivers: 1) environmental concerns 2) economics, 3) population growth, and 4) energy security. A major consequence will be the expansion of operating nuclear power plants from the few hundred we have today to a few thousand by the end of this century. This expansion will present challenges and opportunities in every area of the nuclear industry, including design and development, construction, supply, operations, maintenance, regulation and safety, decommissioning, and the entire nuclear fuel cycle. The creative innovation that characterized the birth of the nuclear power industry is needed in all these areas to address fully the challenges and opportunities. Some aspects of thorium fuel cycles are used to illustrate this point.

1. Introduction

W.B. Lewis and his colleagues established the technical foundation of the Canadian nuclear industry and the CANDU[©] reactor during a period of intense creativity and innovation¹. Indeed, it can be argued that almost every major concept we have today was originally considered or developed during Lewis' era. This initial outburst of underlying innovation has been followed by an important period of development that has seen enormous advancements in safety, design, delivery, supply, and operation of CANDU reactors.

But in addition to this, it is my belief that the time has come to re-examine on a more fundamental basis all aspects of our technology and in so-doing, perhaps challenge some of the conventional approaches. It is opportune to do so since our industry's growth will attract new generations of talented people who will undoubtedly bring new perspectives to technology directions.

This lecture is divided into 3 parts. First, we will examine the drivers that are likely to shape our industry over the next few years and the consequences of these drivers. Second, thorium fuel deployment will be used as an example of an important technology that requires advancement to ensure sustainability for the very long term. Third, we will consider the fission process itself to see if a more fundamental look at the various components of fission could lead to some speculative ideas on what could be done in the much longer-term future to enhance fuel and fuel cycles. The intent of the latter topic is to stimulate some out-of-the-box thinking on how we might advance well beyond where we are today since I believe we still have a long way to go to realize the full potential of nuclear power technology.

¹ CANDU is a registered trademark of Atomic Energy of Canada Limited.

2. Drivers shaping the nuclear industry

The drivers for growth of the nuclear industry can be classified into four main topics: environment, economics, population growth, and security of energy supply. In characterizing the key drivers, I have drawn extensively on information from the OECD/NEA Nuclear Energy Outlook (NEO) published late last year [1].

2.1 Environmental concerns -- global warming

Global warming has become an increasingly urgent international issue since the 2007 update report by the Intergovernmental Panel on Climate Change (IPCC) [2]. The IPCC states in this report that as the result of additional research, the probability that anthropogenic emissions are affecting climate change is now greater than 90%. At the recent 2009 March Copenhagen meeting to update the 2007 report, the climate researchers issued the following press release: "The worst-case IPCC scenario trajectories (or even worse) are being realized ... There is a significant risk that many of the trends will accelerate, leading to an increasing risk of abrupt or irreversible climatic shifts" [3].

Power generation is not only the largest source of CO_2 emissions, but it is also the most rapidly growing source, as indicated in Figure 1. In addition, the recent rate of the increase appears to be accelerating.



Intergovernmental Panel on Climate Change (2007)

Figure 1 Sources of anthropogenic carbon dioxide emissions

Although other emissions sources sometimes receive considerable publicity (such as the oil sands industry in Alberta), the data show clearly that electricity generation is the critical area to focus the main effort to reduce emissions. Also, while all approaches to CO_2 emission reductions such as conservation have a role, the reality is that the only way to seriously address emissions is to include in the various measures an aggressive new-build nuclear reactor program. Nuclear power is the only large-scale energy generation technology that can be widely deployed before the end of the century to address the emissions issue.

2.2 Economics

The second major driver is economics. The OECD/NEA NEO considered the cost of generation in OECD countries, and the results for nuclear, coal, and natural gas in Canada are shown in Figure 2. Since Canada is one of the most competitive countries in the world with respect to electricity production, one would expect that if nuclear energy is competitive here then it can be competitive everywhere.



Figure 2 Levelized unit energy cost in US\$ for electricity generation in Canada. Source: OECD/NEA/IEA (2005), reported in [1]

Contrary to some perceptions, the actual data show that nuclear power in Canada is the least expensive choice. If additional charges are levied for the environmental cost of carbon dioxide emissions, this cost advantage will increase significantly. Moreover, the cost of nuclear power is relatively insensitive to the cost of fuel. The OECD/NEA NEO states that a doubling in the cost of fuel would raise electricity costs by 75% for natural gas, 40% for coal, and only 4% for nuclear [1]. Therefore, costs are both lower for nuclear and are relatively stable even with the inevitable fluctuations in fuel costs. Such long-term steady costs will benefit competitiveness and strategic planning by industry.

It is also of interest to consider how Canadian nuclear plants compare with other OECD countries. These data are presented in Figure 3. It is perhaps remarkable that Canadian plants produce electricity at costs below those of many other countries including those that operate many more plants with the benefit of economies of scale.

2.3 Population growth

The world's population growth will undoubtedly have a major impact on the deployment of new nuclear power plants. The United Nations most recent medium scenario is that the world's population will increase from under 7 billion people today to more than 9 billion by 2050. Also,

there is the continuing aspiration for human development throughout the world. This will create a demand for electricity that will exceed that due to the population increase alone since electricity is the engine of growth, particularly for the new knowledge-based economies.

In addition, we will see new pressures on the basic requirements for human survival, such as fresh water. The annual increase in the consumption of fresh water is 64 billion cubic meters, which, to put this increase into context, is about the annual volume used by Egypt [4]. Even in developed countries, fresh water is becoming an important issue. As a result, desalination has increased from almost nothing in the early 1970s to more than 3.5 million cubic meters per day in 2004 [1].



Figure 3 Nuclear power levelized unit energy costs for OECD countries. Data from OECD/NEA NEO (2008)

2.4 Energy security

Energy security has three main considerations: 1) the world supply of uranium, how long that supply can be accessed economically, and the prospects for extending uranium resources; 2) securing a domestic source of fissile material for those countries that do not want to depend on the continuing reliability of imports; and 3) the ability to store fuel locally to overcome any short term disruptions in the supply chain.

As discussed later in this lecture, uranium supply appears to be secure for the next few decades depending on the rate of nuclear power growth. Even when conventional supplies become scarce or expensive, advanced fuel cycles could extend fuel resources indefinitely. Domestic supply of fuel can be secured by developing fuel cycles that extend uranium supplies or that breed sufficient new fuel for a closed cycle. Owing to the very small amount of fuel required to operate a nuclear power plant and the fuel's high stability, fuel can be stored locally for relatively long periods of time. Therefore, there is considerable flexibility when developing nuclear fuel security policies that does not exist for other fuels.

3. Effect on nuclear power growth

The World Nuclear Association, the International Atomic Energy Agency, and the OECD/Nuclear Energy Agency have all assessed the prospects for the future expansion of nuclear power and have come to similar conclusions. In what follows, for consistency I will draw once again on the OECD/NEA NEO [1].

Figure 4 shows the low and high NEA scenarios. The low scenario assumes there will be little growth in the number of nuclear plants but that older smaller plants will be replaced by larger 1000 MWe plants. The high scenario appears to have a slope similar to the rapid increase in capacity in the 1970s and early 1980s. The NEA has determined that the nuclear industry is capable of meeting this demand. Owing to the drivers discussed above, I believe we need to plan for the high scenario and even for scenarios that go beyond these estimates. The implications are that by the end of this century the world will be operating a few thousand nuclear plants in contrast to the few hundred plants today.



Figure 4 OECD/NEA predictions for nuclear growth to 2050.

4. Fuel supply

The rapid increase in nuclear power raises the question of long-term fuel supply. Figure 5 shows the overlay of the NEA growth projections on the supply of uranium. The overlay suggests that uranium supplies are quite adequate to meet demand until at least 2030 and beyond. It is unusual for a mineral resource to have a secured supply for such a long period of time. Also, from past experience we can expect the uranium mining industry to respond to the increase in demand and that the high scenario fuel supply requirements will be achieved.

However, the rapid expansion of nuclear power will put additional emphasis on the supply issue over longer periods of time. To commit to a 10-fold increase or more in nuclear power, decision-makers will have to be confident that the large upfront investments that are required will not be undermined by fuel supply over the likely 60+ year lifetime of the plants. In addition, a sharp increase in the use of nuclear fuel will likely raise concerns about the eventual disposition of nuclear waste in some countries, particularly where progress on deep geologic disposal has been slow. Finally, even though world nuclear fuel resources may be plentiful, some nations will want to

develop local supplies by exploiting their indigenous resources of thorium. For all these reasons, fuel cycles will receive considerably more interest in the coming years.



Figure 5 NEA NEO assessment of world uranium supplies [1]

5. Reactors and fuel cycles

Two key questions are the type of reactors and the types of fuel cycles the world will adopt. The answer to these questions will depend on the complex interplay between economics and policy. While policy will drive the initial efforts, as in all things, economics is likely to be the main consideration for longer-term sustainability.

5.1 The dominance of water-cooled reactors

Currently, the only commercially successful reactor type that is still being built today for power production is the water-cooled reactor. We have considerable experience with the design, safety, and operation of these reactors, which are major factors for managing the risk associated with large capital investments. Moreover, the major reactor vendors have just invested considerable resources to develop state-of-the-art Generation III technology and it is likely that they will need to recover these investments over the next few decades when the major expansion of nuclear power will occur.

For all these reasons, the rapid large-scale deployment of nuclear power using water-cooled reactors will likely predominate until other reactor types prove to be more compelling. But the full commercialization of other reactor types may take several decades. Therefore, to meet the urgent needs created by the drivers discussed above, it seems to me that we will need to focus on thermal reactors and not wait for new technology that will be available at some uncertain time in the future. This is not to say that such development is unnecessary; it simply accepts the economic reality that most if not all new commercial nuclear plants over the next few decades will be water-cooled thermal reactors. Therefore, in what follows I will concentrate on the deployment of water-cooled reactors in future fuel cycles².

² The Generation IV Super Critical Water Reactor is the only Generation IV reactor that is based on water cooling. The NRCan program builds on the CANDU reactor and, therefore, is an advanced evolution of the water cooled reactor.

5.2 Fuel cycles

There are many considerations for the deployment of future fuel cycles: economics, waste management, proliferation resistance, policies, lead times, and technology.

Fuel cycles can be classified into three underlying strategies. The first strategy is the once-through cycle where the fuel is used in a reactor and is then treated as a waste. This is the category currently followed by most nuclear power nations. The second strategy is the recycling of spent fuel to extract the remaining fissile material before disposing of the waste. This strategy extends the supply of uranium but requires the implementation of relatively complex reprocessing technology, the full benefit of which may not be realized if this is considered the end point for fuel cycle development. The third stage is to develop self-sufficient cycles which are largely independent of an external supply of uranium. This stage requires a fuel and reactor that can breed new fissile material.

From a purely economics point of view, the once-through fuel cycle is very compelling. The entire cost of the fuel cycle, including the safe permanent disposal or retrievable storage in deep geological structures, is included in the cost of power generation. This has proved to be a sustainable economic model and as we saw earlier, the price of nuclear electricity is very competitive with all other forms of electricity production even though nuclear electricity prices include the costs of waste disposal.

The second strategy is aimed primarily at spent LWR fuel with a fissile Pu content of about 0.6% and a fissile U-235 content of 0.9%, depending on burnup and initial enrichment. For this approach, the Pu is extracted from the fuel and burned in LWRs. The wastes include fission products and the minor actinides, but the major by-product is the recovered uranium (RU) containing 0.9% U-235. The Pu from LWRs would eventually be used to fuel fast reactors, which would in turn breed additional Pu from U-238 for self-sufficiency.

The alternative to breeding Pu-239 from U-238 using fast reactors is to breed U-233 from Th-232 using thermal reactors. The U-233 could be used to extend uranium resources (strategy 2) or even to achieve a self-sufficient closed cycle (strategy 3).

Proponents of advanced fuel cycles will argue that we need to make better use of existing resources, since in the once-through cycle we are currently leaving more than 95% of the energy generation potential in the used fuel. Advanced fuel cycles are required to ensure the sustaining of nuclear power well beyond the current supply of uranium that is relatively inexpensive to exploit. Some proponents will also argue that we should endeavour to reduce nuclear wastes, particularly long-lived minor actinides, in the spent fuel by devising strategies to burn the waste in current and future reactors. This will become more urgent as fuel wastes increase in proportion to the increase in nuclear capacity. Finally, there is the policy driver mentioned above for fuel supply security in some countries that do not have sufficient indigenous uranium supplies to support their nuclear program.

However, if new technologies such as fast reactors and conventional reprocessing are established and integrated into the cost structure, there is a question of whether the cost of generation will still be competitive. Therefore, I believe there is considerable incentive to look at the existing thermal nuclear plants and their role in future fuel cycles. In particular, the wide-spread belief that the fast reactor/reprocessing route is the only way to sustain nuclear power in the longer term needs to be examined carefully. It is prudent to consider an alternative since a single approach to future sustainability is not without risks.

6. Thorium fuel

The various thorium fuel cycles have been well-documented [5]. I consider only three topics in this lecture – the use of U-233 for thermal breeding, a comparison of uranium and thorium fuel, and recycling using dry decontamination of spent thorium fuel.

6.1 Why U-233 is used for thermal breeding

The starting point for all advanced fuel cycles is the availability of neutrons. In particular, we want

the smallest possible (n, γ) absorption by fissile isotopes, which has the dual negative effect of removing both fissile fuel and neutrons. For breeding, a minimum average of two neutrons per fission is required, one to sustain fission and the other to produce more fissile material. However, since there are inevitable neutron losses in the core due to structural materials and leakage, the net average neutron generation per fission needs to be larger than two.

Figure 6 shows the average fission neutron properties for the main fissile species, U-235, Pu-239, and U-233 as a function of neutron energy, determined from ENDF/B VII cross section data [6]. The red solid line is the average number of neutrons produced per fission event, usually designated v. As might be expected, Pu-239 has a higher v as it is a larger nucleus. However, the most important neutron parameter, designated η , is the green dotted line which is the average number of neutrons remaining after correcting for the self-absorption of neutrons in the fissile nucleus.

Fission cross sections for fast neutrons are more than two orders of magnitude smaller than for thermal neutrons. However, the number of neutrons produced per fission increases sharply with

incident neutron energy in the fast region. As well, the (n, γ) reaction cross sections are decreasing more rapidly with energy than the fission cross sections. The fission cross sections then level off and more reaction channels open up, such as (n, n f) and (n, 2n f). The net result is that there are more neutrons available per fission event for sustaining fission and for breeding, notwithstanding the smaller cross sections.

Figure 6 also shows why a breeding cycle based on U-235 in a thermal reactor is not possible. The η values are 2.05 in the thermal region, which is not sufficient to sustain breeding. Pu-239 is also not suitable for thermal breeding since its η value in the thermal region is only 2.18 and in a harder spectrum actually dips well below 2. On the other hand, Pu-239 is an excellent fuel for breeding in the fast neutron region above 100 keV, with η values approaching 3 at 1 MeV. But only U-233 is suitable for thermal breeding with $\eta = 2.31$ and is, therefore, the only fissile isotope that can form the basis for a thermal breeding cycle. The CANDU reactor, with its high neutron efficiency, is particularly well-suited for this fuel cycle.



Figure 6 Neutron v (average neutrons/fission, red line) and η (average neutrons available per neutron absorbed, dashed green line) values for the three main fissile isotopes.

It is interesting to note that even in the resonance absorption region the U-233 η values are ≥ 1 . This means that U-233 might also be suitable for a homogeneous core reactor where the moderator and fuel are mixed. With other fissile material, a homogeneous core would quickly lose its neutrons in the resonance region.

6.2 Actinides in thorium and uranium fuel

Figure 7 shows the build up of actinides when a cubic centimetre of uranium and a cubic centimetre of thorium are subject to the same thermal flux over the same length of time. (Note that we are ignoring fast neutron reactions in this simple assessment.)

At steady state, where we are burning U-233 as fast as we are producing it, the U-233 concentration in thorium is quite high -1.5%. If thorium fuel pins are introduced into fuel bundles with uranium driver fuel, then at first some additional U-235 is required to breed U-233. However, the U-233 builds up over time with the net effect that the amount of U-235 needed in the core is reduced, thus improving uranium utilization [5]. If a direct recycling scheme is employed, whereby thorium fuel pins are recycled without any reprocessing so they can achieve high burnup, then uranium requirements could be reduced by 33% from the already highly efficient natural uranium cycle. In addition, the irradiated thorium pins could be stored for future extraction of the U-233, which would further improve uranium utilization.



Figure 7 Long-lived actinide isotope build-up in uranium and thorium at a thermal neutron flux of 4.3e13 for one year. The original target material concentrations (U-238, U-235, and Th-232) and shorter-lived transient species (e.g., Pa-233) are not included. The major fissile isotopes have a shaded background.

If a recycling scheme is developed where the fission products are removed from the spent thorium fuel, then the U-235 requirements can be reduced considerably further, by a factor of 4. Thorium fuel recycling technology is also essential for the ultimate development of a self-sufficient equilibrium cycle where the reactor breeds the same amount of U-233 as is present in fresh fuel.

From Figure 7, one of the key advantages of thorium is immediately apparent. Uranium in a thermal neutron flux produces a relatively large number of long-lived actinide isotopes. Thorium, on the other hand, produces much less long-lived minor actinide material and, therefore, is considered to be a much cleaner fuel.

It is also noted that considerable U-236 builds up in irradiated uranium. This isotope is a neutron poison in LWRs but has little detrimental effect in the softer CANDU neutron spectrum. For this reason, the recycling of the recovered uranium from spent LWR fuel in CANDU reactors is very attractive, particularly since RU, as mentioned previously, is the most abundant by-product from reprocessing LWR fuel. In addition, the high thermal flux in CANDU reactors is an effective way to destroy the minor actinides³. Therefore, there is considerable synergism between LWRs and CANDU PHWRs that I believe will be increasingly exploited as the requirements for waste destruction and higher uranium utilization grow [7].

³ The thermal flux in a CANDU reactor fuelled with actinides in an inert matrix approaches the fast flux values in an FBR.

6.3 Dry processing

6.3.1 <u>DUPIC</u>

An alternative approach to conventional uranium fuel reprocessing is the DUPIC cycle (DUPIC = Direct Use of PWR Fuel in CANDU reactors) [8]. This approach makes use of the fact that there is sufficient reactivity in spent LWR fuel even loaded with fission products to operate a CANDU reactor. DUPIC has the potential to increase the energy from existing LWR spent fuel by up to 50%, which would result in a considerable savings in uranium. I will discuss DUPIC in a bit more detail since it has some relevance to a "dry processing" approach that could be used for thorium fuel cycles.

The process ultimately selected for DUPIC involves oxidation/reduction steps between UO_2 and U_3O_8 to condition the fuel for sintering into CANDU fuel pellets. The oxidation/reduction has the side benefit of removing some volatile fission products, thus increasing fuel burnup and reducing radiation fields. This can be understood by considering how the chemical species for fission products change in fuel under different temperature and oxidizing conditions, summarized in Table 1.

Table 1 includes about 75% (atomic percent) of the fission products in spent fuel as well as the broad range of chemical behaviour likely to be encountered. Under UO_2 fuel conditions, the fission products exist as oxides and oxy-compounds. On oxidizing UO_2 to U_3O_8 , the fuel disintegrates to a powder, thus exposing the fission products to the added oxygen. Of the 10 most abundant fission products, half of them are expected to be volatile at temperatures below 2000 C under oxidizing conditions. At this point, I will note that an obvious but important aspect of DUPIC is that only a very small quantity of material is actually evaporated from the fuel and most of the fuel materials remain in the solid state. This means that the off-gas system is small and relatively simple.

Of course, there are many other important fission product species removed from the fuel (such as iodine), which are present in smaller quantities. In addition, there are fairly benign fission products, such as zirconium and barium that have relatively small neutron absorption cross sections and could remain in the fuel with little reactivity penalty. However, there are also some very high absorption cross section species such as neodymium that, if removed, would result in much improved fuel performance.

Table 1 also compares the chemical species in ThO_2 fuel to those in UO_2 . ThO_2 is much more stable than UO_2 and the oxidation potential in the fuel is, therefore, correspondingly lower. As a result, thorium fuel contains many fission product species in their elemental form. In particular, Mo is not oxidized in thorium fuel, which eliminates its propensity to react with other fission products.

We can also consider whether a DUPIC oxidation/reduction process can be developed to remove species that are volatile under reducing conditions. Figure 8 is an Ellingham diagram for three different environments surrounding the fuel (dashed lines) and for two fission products: ruthenium, which is volatile in air, and strontium, which is volatile under reducing conditions.

From Figure 8, we can see that air should start to oxidize RuO_2 to $RuO_3(g)$ at temperatures above about 1500C. This is consistent with air sweep gas experiments at CRL where 100% of the ruthenium was released from spent fuel at about 1600 C. Of course, it is not necessary to have all the ruthenium in the gas phase since the equilibrium constantly shifts to vaporize more ruthenium as the $RuO_3(g)$ is swept away.

The Ten Most	UO ₂	U ₃ O ₈ [O ₂ /U=10]		ThO₂
Abundant Stable and Long-Lived Fission Products in Fuel*	1000C	1000C	2000C	1000C
Zr	ZrO ₂	ZrO ₂	ZrO ₂	ZrO ₂
Мо	Cs₂O·MoO₃	Cs₂O·MoO₃	MoO ₃ (g)	Мо
Nd	Nd_2O_3	Nd_2O_3	Nd_2O_3	Nd_2O_3
Cs	Cs₂O·MoO₃	Cs₂O·MoO₃	Cs(g)	Cs
Ce	CeO ₂	CeO ₂	CeO _{2-x}	CeO _{2-x}
Ru	RuO ₂	RuO ₂	RuO ₃ (g)	Ru
Sr	SrMoO ₄	SrMoO ₄	SrMoO ₄	SrZrO ₃
Ва	BaMoO ₄	BaMoO ₄	BaMoO ₄	BaO∙ZrO ₂
La	La ₂ O ₃	La ₂ O ₃	LaO ₂ (g)	La ₂ O ₃
Тс	TcO ₃	Tc ₂ O ₇ (g)	TcO ₃	Тс

Table 1 Predominant chemical species for the 10 most abundant solid fission product elements in fuel. The noble gas isotopes are not included. The listed elements comprise about 75% (atomic percent) of the fission products in spent fuel.

It is also apparent that using hydrogen to drive off strontium as Sr(g) will only work at relatively high temperatures, at least when the partial pressure of water is maintained at 10^{-3} atm. Another approach would be to use the C/CO system at low CO partial pressures to create stronger reducing conditions, a standard procedure in metallurgy. In that case, strontium would be removed at much lower temperatures.

But oxidation/reduction will not work for some important species – for example, neodymium. For this, we would need to consider more advanced DUPIC processes where other reagents are employed, such as fluorine and chlorine. Such a discussion is beyond the scope of this paper for DUPIC; however, we shall return to this concept in the next section on thorium fuel recycling.

6.3.2 <u>Th-232/U-233 recycling</u>

The full potential of the thorium cycle can only be realized by separating the fission products from the irradiated fuel, so we will discuss this more detail. A DUPIC-type process would only work if the fuel is subject to an oxidation/reduction process that also breaks up the fuel lattice and allows the fission products to be exposed to the surrounding gaseous environment. Figure 8 shows that the C/CO system could be used to cycle between ThO₂ and Th, but it is not clear that this would break up the lattice. Therefore, the spent fuel may have to be broken up mechanically. In addition, we need to consider how to remove some of the high absorption cross section fission products such as Nd₂O₃, which is not volatile in either oxidizing or reducing conditions.

One approach is to treat the fuel with fluorine or chlorine to convert most of the fuel species to the halide form. This takes advantage of the fact that many fluorides and chlorides are volatile. Figure 9 shows the effect of fluorine on the thorium, uranium, and neodymium in the fuel. The U-233 in the fuel is converted to volatile UF_6 , which can be separated from the rest of the species at very low

temperatures. This would appear to be an effective way to "mine" the uranium from the fuel with little or no contamination from other materials. The uranium contains U-232, which would render the fuel unsuitable for weapons applications due to the high radiation fields associated with the U-232 decay chain. However, by going to higher temperatures, it would also be possible to evaporate some of the ThF₄ with the UF₆ as well; for example, at just above 1000 C the evaporated material would be 50% uranium and 50% thorium.



Figure 8 Ellingham diagram for ruthenium and strontium in UO₂ fuel and for ThO₂. It is noted that the fission product species activities were set equal to the mole fractions in the fuel, and the equilibrium lines for strontium and ruthenium assume that 1% of the fission products are in the gas phase.

The extracted uranium could be blended with fresh thorium (or with depleted or natural uranium) to provide new fuel and the fission products could be retained in the irradiated thorium. This would leave a large amount of thorium waste and would not be a particularly sustainable use of a valuable breeding resource.

To reuse the irradiated thorium removal of the fission products would also be required. Figure 9 shows that at temperatures above about 1400 C, NdF₃ becomes volatile but so does ThF₄, so there would be no separation. However, by adjusting the chemistry we have considerable flexibility to optimize the system to get the desired separations. For example, Figure 10 shows what happens in oxidizing conditions and low fluorine concentrations. This combination was selected to stabilize the ThO₂ with excess oxygen and reduce the amount of ThF₄, while providing sufficient fluorine to convert Nd₂O₃ to the volatile NF₃. While the system needs to be optimized, Figure 10 shows that it should be possible to achieve acceptable separations. This mixture of oxygen and fluorine also retains the uranium in the solid state, so it could be used as a DUPIC-like process to remove only the fission products, thus increasing the proliferation resistance of the fuel cycle even further.



Figure 9 Species resulting from the treatment of irradiated thorium fuel with fluorine.



Figure 10 Thorium oxide fuel (1 mol) with 10 mol O_2 and 0.01 mol F_2 .

Figure 10 includes the behaviour of ruthenium under these oxidizing conditions to illustrate that other fission products would also be removed from the fuel. Obviously, these assessments are quite rudimentary and much more detailed analysis is required. Nevertheless, they illustrate the flexibility we have in developing a dry processing approach to exploit the full potential of the thorium fuel cycle.

Once again, as for DUPIC, it is emphasized that for this type of dry separation we are volatizing relatively small amounts of material from the fuel and leaving the thorium (and uranium if desired) in the solid state. This approach should be the least-cost option for dry processing and might be considered before reverting to dry processes that use bulk vaporization of all the fuel materials.

In summary, some of the key advantages of thorium are that we do not need a new reactor type to establish the fuel cycle, thorium is about three times more abundant than uranium in nature, thorium is a relatively clean fuel as far as the minor actinides are concerned, and if we focus on the underlying chemistry we should be able to develop a relatively simple dry process for recycling. This would extend uranium resources in the short term and would lead progressively and logically to the development of a self-sufficient cycle.

7. Fission – can we do more than heat water?

As a final topic, I would like to pose the question "are we getting everything we can from fission?" I am reminded that nuclear power is still a very young technology and that to date we have only used the remarkable process of fission to heat up water. The conversion of fission energy to heat is very efficient, but more than 60% of the energy is lost when we convert the heat to electricity. I would be disappointed if, by the end of this century, we are still thinking of nuclear fuel as only a heat source. So, the final part of this lecture is admittedly highly speculative but it is meant to stimulate the imaginations of future generations of nuclear experts who will undoubtedly come up with their own ideas.



Figure 11 The fission process.

Figure 11 summarizes the fission process. When a fissile nucleus absorbs a thermal neutron that causes fission, energy is carried away by a number of processes. Here, I will consider the two processes that create energetic particles – fission neutrons and fission fragments. In these processes, the energy is concentrated in just a few particles. Ultimately, we make use of the concentrated energy by allowing it to dissipate throughout the fuel as low grade heat.

Could we make better use of this concentrated energy before it is dissipated? I believe this to be the case, but we are going to have to expand considerably our conceptual thinking about fuel to include highly advanced fuel designs that go far beyond our current approach. Fortunately, on-power

refuelling and the simple CANDU fuel bundle design provide considerable flexibility, which is a good starting point.

7.1 Fission neutrons and "fusion-enhanced fission"

Fission neutrons carry off about 5 MeV of kinetic energy, with an average energy per neutron of about 2 MeV⁴. The fission neutrons initially interact with fuel atoms and lose energy until they escape to the moderator with remaining energies around 100 keV. The energy loss, on average, is much smaller for collisions with heavy nuclei such as U and Th (~0.8 %) compared to collisions with O (~11%), so most of the energy is lost in collisions with the lighter nuclei.

Let's suppose that an advanced fuel design allows the fast neutrons to interact with deuterium (D) and tritium (T) nuclei. For example, the fuel could be a hydride of D and T, or the fuel design could be more sophisticated with zones of high density D and T to absorb the energy. In any event, most of the neutron energy would be transferred to these nuclei. The average energy loss per collision is 44.4% for D and 37.5% for T. Perhaps 5 D/T recoils would result from each neutron before it escapes from the fuel, or about 12 recoils per fission. The D/T recoils would have kinetic energies ranging from 900 to 50 keV and would collide with stationary D/T nuclei in the fuel. The peak in the cross section for the fusion reaction D + T = He(3.6 MeV) + n(14 MeV) occurs at ~110 keV for a D particle on a stationary T target and at ~190 keV for T on a stationary D target. Therefore, we are in the right energy range, particularly since the higher energy to the target atoms. However, these are sub-coulomb barrier reactions, so the peak cross section is only 5 barns. Nevertheless, with a dense target of D and T, the reaction rate could possibly be optimized.

There are many potential advantages to this approach. We are using a small amount of the neutron's energy to drive an exothermic reaction to gain an additional 17.6 MeV. Those D and T recoils that do not undergo fusion will simply transfer their kinetic energy to the fuel so no energy is lost. But more importantly, we are gaining extra neutrons that produce fast fissions with very high neutron multiplicities and η values. Thus, we increase the number of neutrons available for breeding and also gain additional energy from the fission process. In addition, since the neutron energy is well above the threshold for U-238 and Th-232 fission, we effectively increase the amount of fissile material in the fuel. The fast fission factor, which is already high for CANDU reactors, is increased. At the same time, a by-product of CANDU reactor operation that may be considered a waste – tritium -- is turned into energy.

Of course, there are also very many challenges – for example, how we would handle the generated He and the increase in U-232 from fast neutron reactions. Therefore, we can only speculate whether such an approach could be engineered to work. But it is an example of how we might reconsider the fundamentals to enhance fuel performance and to move closer to a self-sufficient thermal breeding cycle using thorium.

7.2 Fission fragments

Fission fragments carry off 85% of the energy arising from fission. Again, we take an intense source of energy and allow it to dissipate as low grade heat to eventually heat up water. If we could devise a way to more directly tap into the fission fragment energy to create electricity, then we

⁴ Note that the most probable fission neutron energy is about 1 MeV, with a distribution tail extending out to higher energies, such that the average neutron energy is closer to 2 MeV.

could increase the efficiency of nuclear power by avoiding the losses associated with generating electricity from steam.

We can use the SRIM Monte Carlo code to determine how the fission fragments lose energy to the material in which they are travelling [9]. The fission fragments transfer about 96% of their kinetic energy to ionization in the fission fragment track⁵. The electrons from the ionization in turn produce electron cascades surrounding the track. This intense displacement of charge eventually dissipates as the electrons become thermalised and recombine with the positive ions, transferring their energy to the fuel. The question we might ask is whether there are better ways to make use of this dense electron/ion region before it degrades to low grade heat. For example, a fuel design where the fission fragments recoil into a gas could facilitate the collection of charge or a more sophisticated approach would couple directly electromagnetically. However, I will not attempt to address further this complex but intriguing question in this lecture, but will leave it as a challenge for others to ponder for the long-term future.

8. Concluding remark

A key point I have tried to make in this lecture is that given the inevitable rapid future expansion of nuclear energy, now is the time to think about how we are going to advance our technology over the coming few decades and even over the rest of the century. If we do this properly with an appropriate balance between shorter term development and longer term advanced innovation, then the future is going to be as exciting as we choose to make it. And I think that is exactly what W.B. Lewis would have expected.

9. References

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⁵ The 4% energy loss not accounted for by transfer to electrons goes into nuclear stopping. This would also create D and T recoils in a mixed hydride fuel, some of which have energies sufficient to cause fusion. A preliminary assessment indicates that about 2.5 recoils capable of undergoing fusion would occur per fission.

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