CHAPTER 1. Introduction

The invention of electricity has made a significant impact in the quality of human life and the growth of a country is often judged by its per capita consumption of electricity. With increasing concern of environmental degradation due to release of greenhouse gases, namely, NO\textsubscript{x}, SO\textsubscript{x} and CO\textsubscript{x} from indiscriminate burning of fossil fuels, nuclear fuels take the centre stage as a viable alternative source for electricity production. As this form of energy is also sustainable and contributes to the reduction in global warming, it is considered as an essential element in any nation’s energy policy.

In 1942, when the team led by Enrico Fermi, achieved the world’s first controlled nuclear chain reaction in the University of Chicago, United States Of America\textsuperscript{1}, it was envisioned that nuclear electricity will be economical. Though, this dream has not been realized, the fact that, the nuclear fission generates tremendous amount of energy with relatively small amounts of fuel material, makes it attractive for exploitation.

While the energy that could be obtained from coal, oil and gas is of the order of 22.3\times10^6 J/kg\textsuperscript{2}, nuclear fuel typically could provide energy of about 7.28\times10^{13} J/kg. Hence the amount of fuel needed to extract the same amount of energy is roughly of the order of 3\times10^6 times more in the case of coal, oil and gas when compared to that of nuclear route.

The nuclear fuel cannot be completely burnt at a single stretch in the reactor, either due to the fission products which inhibit the nuclear chain reaction or due to the degradation of the structural materials which contain the fuel. By recovering the unburnt uranium and the fissile materials produced in the reactor, more power could be tapped from this fuel. To realize this objective, reprocessing the spent fuel from the reactor is necessary.

1.1. The need for energy sources

Right from the early struggle for biological survival to the present technological world, the human life continues to exist due to the discovery and exploitation of various new sources of energy. Starting from learning to generate and make use of fire for their existence, humans have harnessed various forms of energies like wind using ships and windmills, hydropower using rivers and waterfalls, exploitation of chemical energy using coal, oil and natural gas. Only in the middle of 20\textsuperscript{th} century nuclear energy was discovered and used on a large scale.\textsuperscript{3} Fig. 1.1 shows the shares of the global primary energy sources as on 2014.
1.2. Disadvantages of fossil fuels

The long-standing impetus for the development of nuclear power has been the eventual need to replace the fossil fuels - oil (or petroleum), coal, and natural gas. Their supply is finite and eventually, at different rates for the different fuels, will be consumed. Natural gas resources may exceed those of oil, measured in terms of total energy content. Nonetheless, gas is also a limited resource. Coal resources are much more abundant than those of either oil or natural gas, but are the least desirable from the environmental safety point of view. The production of carbon dioxide in the combustion of fossil fuels presents a more difficult problem. Unless much of this carbon dioxide could be captured and sequestered, the resulting increase in the concentration of carbon dioxide in the atmosphere carries with it the possibility of significant global climate change.

Among the fossil fuels, natural gas has significant advantages. It is environmentally least damaging, in terms of both chemical pollutants and carbon dioxide production. If the projected supplies of “unconventional” natural gas live up to some of the estimates⁴, then supply difficulties may be postponed for many decades. Further, natural gas could be used in highly efficient combustion turbines operating in a combined cycle mode, in which much of the (otherwise) waste heat from the combustion turbine is used to drive a steam turbine. Nonetheless, reliance on natural gas as more than a short-term stop gap involves two significant issues. First, the gas supplies may be limited to the standard conventional
resources, advancing the time at which the availability of gas will become a problem and prices will rise substantially. Second, although preferable to coal in this regard, natural gas is still a source of greenhouse gases, primarily carbon dioxide from combustion and secondarily methane from leaks.

1.3. Why Nuclear Energy?\(^5\)
The predominant reason that the nuclear energy managers put forward for switching over to nuclear energy from the fossil fuels is the fact that, unlike the fossil fuels, the by-products of nuclear power are more benign to the environment. Also with appropriate choice of nuclear reactors, the nuclear fuel could be used inexhaustibly. This translates into the fact that with the same amount of fuel that was used to start a nuclear reactor, infinite amount of energy could be generated. Nuclear fission, the basic process for power production using nuclear reactors, emits huge amount of energy (~1 MWd per g of \(U^{235}\) atoms). Hence with a small amount of fuel, large amount of energy could be generated\(^6\).

In addition, some of the fission products generated in nuclear reactors possess many societal benefits like in medicine, industry, agriculture, food preservation, water resource management, environmental studies etc\(^7\). In the health care sector, the applications of radioisotopes for diagnostic and therapeutic purposes are innumerable. Today, nuclear medicine offers nearly one hundred procedures that are immensely helpful to a broad span of medical specialties ranging from oncology and cardiology to psychiatry due to which several millions of patients benefit annually, all over the world. Radiopharmaceuticals are preparations containing radioactivity used in medicine for either diagnostic or therapeutic purpose. The industrial applications of nuclear energy can be broadly categorized into four types, namely, (i) Non-destructive evaluation and testing, (ii) Nucleonic measurement and control systems (nucleonic gauges) which are essentially non-contact/non-intrusive mechanisms for measurement and control of process parameters such as thickness, density, level and composition, (iii) Radiotracer applications for industrial process troubleshooting and optimization and (iv) Radiation processing like development of commercial processes such as polymer modifications and sterilizations etc.

The nuclear energy route for power production though offers various advantages over the conventional fossil fuels as stated above, it is certain that great caution needs to be exercised in deploying it. The biggest concern of all is the environmental impact of the nuclear materials and the radioactive wastes generated in the process of nuclear fission.
which demands for a better way of disposition of the spent nuclear fuel and its associated radioactive wastes. The next big challenge is the probability of any nuclear accidents during any of the nuclear fuel cycle operations. These challenges could be overcome by engineering the process design in the most practically safest manner. The International Atomic Energy Agency (IAEA), Vienna and the Atomic Energy Regulatory Board (AERB), India have laid out various pre requisites for the safety of the operating personnel and the general public while carrying out any operation, be it industrial or R&D, which when followed meticulously ensures the management of the above challenges. Additionally, continuous use of nuclear energy also reduces the finite availability of the nuclear materials from the earth’s crust. But the concepts like breeder reactor wherein one can breed more fuel and end with excess fuel materials over and above the initial inventory, the issue of finite availability of nuclear material is already overcome by the world’s nuclear energy fraternity. Finally the most horrifying issue with nuclear energy is the deployment of nuclear energy in harmful ways by anti-social elements. To overcome this, the world nuclear bodies are setting up various treaties and unions which ensure the non-proliferation resistance of nuclear materials.

1.4. Nuclear Fuel Cycle

The nuclear fuel cycle refers to the series of operations which the nuclear fuel undergoes since its fabrication. The cycle starts with the mining of the nuclear fuel materials from the earth’s crust followed by its purification and fabrication to a suitable physical and chemical form as nuclear fuel. The various chemical forms include metals and alloys and also some ceramic forms like oxide, carbide and nitride. The usual physical forms include sheets of metal like in APSARA reactor at the Bhabha Atomic Research Centre (BARC) in Mumbai or rods for the rest of the chemical forms. Once the fuel is fabricated in the desired physical and chemical form, it is irradiated in the nuclear reactor for power production. When the burn up (amount of energy extracted from the given quantity of fuel) of the fuel reaches a certain set limit, the irradiated fuel is then unloaded from the reactor and is allowed to cool for certain period of time so that the radioactivity and the decay heat reduces to acceptable levels. Subsequently the cooled fuel is reprocessed wherein the unburnt and the newly bred fissile material are separated and recycled back to the fabrication plant for fresh fuel fabrication. The radioactive waste generated during the reprocessing operations is then sent to a waste management plant wherein the waste are
segregated and treated accordingly. The most important of that waste are the high level liquid waste (HLLW) which is finally fixed in glass matrix and then enclosed in SS canisters for short term storage in special shallow underground storage locations called tile holes. After surveillance during the short term storage period when the decay heat and radioactivity reduces considerably, they will be transferred into deep geological repositories for long term storage. Fig. 1.2 shows a typical nuclear fuel cycle.

![Schematic of Nuclear Fuel Cycle](image)

1.5. *Spent nuclear fuel*

The nuclear fuel once loaded into a nuclear reactor in an appropriate physical and chemical form, will be subjected to neutron induced fission for power production. For various reasons, this fuel could not be consumed completely or otherwise the burnup (the amount of energy extracted/extractable from the fuel) of the fuel is limited. In case of thermal nuclear reactors, the burnup is limited due to the accumulation of certain neutron absorbing fission products, like $^{83}$Kr, $^{95}$Mo, $^{143}$Nd, $^{147}$Pm, $^{149}$Sm, $^{157}$Gd etc., which also start competing with the fuel material in consuming the neutrons. These are called neutron poisons as the neutron absorbed in them does not lead to any fission. In the case of FBRs also these neutron absorbing fission products does get formed. But as their neutron absorption cross section is very low for the fast neutrons, neutron poisoning is never an issue in reaching higher burnup. But as the burnup increases, the physical integrity of the fuel clad and the reactor structural materials becomes an issue which limits further increasing the burnup. Hence the fuel has to be discharged. This discharged fuel from the
reactor after irradiation is termed as the **spent nuclear fuel**. In any given type of reactor, in addition to these factors, nature of the fissioning nuclide also plays a major role in determining the maximum amount of energy that could extracted from the given fuel. Thus in a FBR, $^{239}$Pu and $^{241}$Pu are the better fuel materials than other fissile materials as their fission cross section with fast neutrons are much better. This is explained more in detail in section 1.8 and 1.9. Also they tend to emit higher number of neutrons per fission in the fast region than the other fissile nuclides which would ensure sustained criticality with comparatively lesser fissile content than other fissile nuclides when all other parameters are maintained the same.

1.6. **Management of spent nuclear fuel**

The spent nuclear fuel, though could be managed in various different ways$^{10}$, they all fall into basically three different types$^{11}$. It could either be stored as such as a nuclear waste in an appropriate location and manner or it could be reprocessed for the recovery of unburnt and the newly bred fissile material present in it. The former case is called the **open or once through fuel cycle** and is normally practiced in countries where there is no dearth of nuclear fuel availability or any other alternate source of energy to meet their power requirements. Countries like USA, Canada, Czech Republic, Germany, Sweden, Finland, Spain, South Africa etc. follow this policy$^{12}$. These countries, have judged it to be more advantageous to implement the open fuel cycle, in which the spent fuel elements discharged from the reactor are stored and after a period of interim storage, the fuel will be conditioned and disposed of directly in a deep geological repository.

The latter case is referred as the **closed nuclear fuel cycle** where the spent nuclear fuel is reprocessed to recover the uranium and plutonium from the waste products for their recycle as new fuel elements for use in thermal and fast reactors for power production. In addition to uranium and plutonium, other nuclides such as $^{137}$Cs, $^{90}$Sr, $^{99}$Tc etc., will also be recovered separately for their use in the radiopharmaceutical and other societal beneficiary applications$^{7}$. The third way of managing the spent fuel is called the “**storage and postponed decision**” or “**wait and see option**”. This is essentially a strategy devised by many countries to postpone the decision for a certain period and to take a delayed decision based on the then current scenario.
1.7. *Three stage Indian Nuclear Power Program*

India's three-stage nuclear power programme (INPP) was judiciously formulated by Late Homi Bhabha, the founding father of the department of atomic energy (DAE), India, in the 1950s to secure the country’s long-term energy independence, through the use of uranium and thorium reserves found in the monazite sands of coastal regions of South India\textsuperscript{13}. The ultimate focus of the programme is on enabling the thorium reserves of India, which is the largest in the world, to be utilized in meeting the country's long-term energy requirements.

In the 1\textsuperscript{st} stage, nuclear power would be generated using pressurized heavy water reactors (PHWRs) driven by the available natural uranium fuel in the country. The spent fuel from these reactors would be reprocessed in-house for the recovery of newly bred Pu\textsuperscript{239} from U\textsuperscript{238}. At a strategic point in time, Fast Breeder Reactors (FBRs) would be set-up which forms the 2\textsuperscript{nd} stage of the program. The plutonium recovered by reprocessing the spent nuclear fuel from the 1\textsuperscript{st} stage would be used for fabricating U, plutonium mixed oxide fuel which will drive the FBRs in the second stage. FBRs due to their inherent design and the neutronics are potential producers of huge amount of energy from the given fuel.\textsuperscript{14} The breeding ability of FBRs will be carefully manipulated in multiplying the fuel and thereby making them self-reliant as well as fuel breeders to set-up more such FBRs subsequently.

At an appropriate juncture of time, the blankets of the FBRs will be loaded with the fertile element thorium which is available in abundance in our country to breed more U\textsuperscript{233} out of it. The U\textsuperscript{233} so obtained, will be separated, purified by reprocessing and would be refabricated as fresh fuel to drive the U-Th based Advanced Heavy Water Reactors (AHWR) in the 3\textsuperscript{rd} stage of INPP. A simplified schematic sketch of the three stage INPP is presented in Fig. 1.3.

Thorium is particularly attractive for India, since the country has only around 1–2\% of the global uranium reserves, but one of the largest shares of global thorium reserves, at about 25\% of the world's known thorium reserves. Closing the nuclear fuel cycle by reprocessing the spent fuel and to recycle the recovered fissile materials back to the reactor for power production is one of the key steps to the success of the entire INPP. Presently the INPP is in the transition between the 1\textsuperscript{st} and 2\textsuperscript{nd} stage wherein the plutonium produced in the first stage by breeding U\textsuperscript{238} using thermal neutrons in the natural UO\textsubscript{2} driven Pressurised Heavy Water Reactors (PHWRs) is reprocessed, recovered and used as driver fuel for the second stage. Thus reprocessing forms the necessary link between the first two stages and hence fast reactor fuel reprocessing plants are required to be set-up for the
successful implementation of the INPP.$^{15}$

![Three Stage Indian Nuclear Power Program](image)

**Fig. 1.3. Three Stage Indian Nuclear Power Program**

1.8. **UO$_2$ as nuclear fuel**

Uranium was discovered in 1789 by the German chemist Klaproth and named in honor of the planet Uranus, discovered five years earlier. It was not until 1841, half a century after the discovery of uranium that the French chemist Peligot succeeded in obtaining this element in a pure state.$^{16}$ Historically, 1941 marked the beginning of the Uranium era when investigation for deploying the atomic energy of uranium for military purposes begun.$^{17}$

Natural uranium refers to uranium with the isotopic ratio of 0.72% U$^{235}$, 99.27% U$^{238}$ and a trace of U$^{234}$ by weight (0.0055%). In terms of the amount of radioactivity, approximately 2.2% comes from U$^{235}$, 48.6% U$^{238}$ and 49.2% U$^{234}$. Among all these isotopes only U$^{235}$ is fissile that could be used as fuel in the nuclear reactors. The 0.72% U$^{235}$ is not sufficient to produce a self-sustaining critical chain reaction in light water reactors or nuclear weapons; these applications must use enriched uranium. Nuclear weapons require a concentration of 90% U$^{235}$ and light water reactors requires roughly 3% U$^{235}$. Natural uranium is an appropriate fuel for a heavy-water reactor, like the CANDU reactor.

In rare occasions, earlier in geologic history when U$^{235}$ was more abundant, uranium ore was found to have naturally engaged in fission, forming natural nuclear fission reactors.$^{19}$ U$^{235}$ decays at a faster rate (half-life of 700 million years) compared to U$^{238}$, which decays...
extremely slowly (half-life of 4.5 billion years). Therefore a billion years ago, there was more than double the $^{235}\text{U}$ compared to now.

Uranium though has major applications in military sector as penetrators owing to the high density of the metal. However its most common application is in the civilian sector as nuclear fuel. One g of $^{235}\text{U}$ on nuclear fission produces about 1 MWd of energy. This is equivalent to the energy produced from about 3.2 tonnes of coal\textsuperscript{20}. Commercial nuclear power plants use fuel that is typically enriched to around 3% $^{235}\text{U}$. The CANDU and Magnox designs are the only commercial reactors capable of using natural uranium as the driver fuel. In a breeder reactor, $^{238}\text{U}$ could also be converted into plutonium (referred to as breeding) through the following nuclear reaction\textsuperscript{21}:

\[
\frac{238}{92}\text{U} + n \rightarrow \frac{239}{92}\text{U} + \gamma \rightarrow \frac{239}{95}\text{Np} \rightarrow \frac{239}{94}\text{Pu}
\]  

(1.1)

Uranium is also an extremely effective fuel or explosive substance. Some isotopes of uranium and plutonium are capable of undergoing nuclear fission or disintegration by the action of neutrons and are referred as fissile isotopes: $^{235}\text{U}$, $^{233}\text{Pu}$, $^{239}\text{Pu}$ and $^{241}\text{Pu}$. Of these 4 isotopes, only $^{235}\text{U}$ occurs in nature while the other three are obtained in nuclear reactors by neutron bombardment of thorium or natural uranium or $^{240}\text{Pu}$ respectively. $^{233}\text{U}$ is formed from $^{232}\text{Th}$ according to the following reaction.

\[
\frac{232}{90}\text{Th} + n \rightarrow \frac{233}{90}\text{Th} + \gamma \rightarrow \frac{233}{91}\text{Pa} \rightarrow \frac{233}{92}\text{U}
\]  

(1.2)

The probability of nuclear fission reactions is usually expressed in terms of the effective cross-section ($\sigma$), in area dimensions, since the probability of reaction between the fuel and a bombarding particle is proportional to the area of the transverse cross-section of the target nucleus. The unit of cross-section is $10^{-24}$ cm$^2$, which is called a barn. This value is approximately the same as the geometric cross-section of an atomic nucleus, the radius of which is equal to $10^{-12}$-$10^{-13}$ cm. The value of $\sigma$ basically depends upon the nature of the bombarding particles and their energy.

In nuclear and particle physics, the concept of a neutron cross section\textsuperscript{22} is used to express the likelihood of interaction between an incident neutron and a target nucleus. In conjunction with the neutron flux, it enables the calculation of the reaction rate, for
example to derive the thermal power of a nuclear power plant. Larger the neutrons cross section, the more likely a neutron would react with the nucleus.

An isotope (or nuclide) could be classified according to its neutron cross section and how it reacts with an incident neutron. The various types of interaction between a neutron and a nucleus are pictorially depicted in Fig. 1.4.

**Fig. 1.4. Various types of interactions between a neutron and nucleus**

![Diagram of neutron-nucleus reactions](image)

Radionuclides that tend to absorb a neutron and either decay or keep the neutron in its nucleus are neutron absorbers. Corresponding to this reaction, the nuclide will possess a cross section designated as neutron capture cross section ($\sigma_c$). Isotopes that undergo fission are fissile and have a corresponding fission cross section ($\sigma_f$). When a fissile nuclide absorbs a neutron, there could be only two possible outcomes namely capture or fission. Hence for all fissile nuclides, the sum of $\sigma_c$ and $\sigma_f$ is called the absorption cross section designated by $\sigma_a$. Thus $\sigma_a = \sigma_c + \sigma_f$. The remaining isotopes will simply scatter the neutron, and have a scatter cross section ($\sigma_s$).

Some isotopes, like $^{238}_{\text{U}}$, have finite values for all three types of cross sections mentioned above. Isotopes with a large scatter cross section and a low mass are good neutron moderators ($^1\text{H}$, $^2\text{H}/\text{D}$ etc.). Nuclides which have a large absorption cross section are neutron poisons if they are neither fissile nor undergo decay (example $^{157}_{\text{Gd}}$, $^{10}_{\text{B}}$ etc.). A poison that is purposely inserted into a nuclear reactor for controlling its reactivity in the
long term and improves its shutdown margin is called a burnable poison.
The consolidated values of various cross sections for the fissile nuclides are provided in Table 1.1.

### Table 1.1. Neutron cross-sections of Fissile nuclides

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Neutron cross section (b)</th>
<th>Average over thermal spectrum</th>
<th>Slowing-down region resonance integrals</th>
<th>Average over fast spectrum$^\dagger$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\sigma_s$</td>
<td>$\sigma_c$</td>
<td>$\sigma_f$</td>
<td>$\eta$</td>
</tr>
<tr>
<td>$^{233}$U</td>
<td>12.19</td>
<td>42.20</td>
<td>468.20</td>
<td>2.495</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>15.98</td>
<td>86.70</td>
<td>504.81</td>
<td>2.433</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>7.90</td>
<td>274.32</td>
<td>699.34</td>
<td>2.882</td>
</tr>
<tr>
<td>$^{241}$Pu</td>
<td>12.19</td>
<td>334.11</td>
<td>936.65</td>
<td>2.946</td>
</tr>
</tbody>
</table>

Here $\eta$ is the regeneration factor and refers to the average number of neutrons emitted per fission. When $\eta$ is high, the number of neutrons available to sustain the chain reaction is higher and thus the probability of fission reaction is accordingly higher. Fig. 5 depicts pictorially the changes in the value of $\eta$ as a function of incident neutron energy for various fissile and fertile nuclides. These values provide a clear understanding of which is the best fuel for the given type of nuclear reactor in order to have maximum breeding. Thus as both $^{239}$Pu and $^{241}$Pu have relatively very high value of $\eta$ in the fast neutron region, they are the best fuel with respect to their breeding potential. Hence in principle they could be deployed in FBRs to extract infinite amount of energy from them, provided, they are sufficiently available initially and a closed fuel cycle is adopted. Similarly $^{233}$U and $^{235}$U are the best available fuel to be employed in the thermal neutron region. In this region the $\eta$ value of $^{239}$Pu is pretty low and hence is not considered a good fuel. It is also interesting to note from Fig. 1.5 that $^{233}$U could be considered as a universal nuclear fuel as its $\eta$ value is relatively pretty high and consistent under all neutron energy region barring the sharp dips in the resonance regions.

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* [http://www.kayelaby.npl.co.uk/](http://www.kayelaby.npl.co.uk/) (Kaye and Laby Online, National Physical Laboratory)
† These values are intended as a rough guide as they are strongly influenced by the construction details and the physical characteristics of the FR (size, geometry, coolant, structural material etc.)
1.9. MOX as nuclear fuel

Mixed oxide fuel, commonly referred to as the MOX fuel, is a nuclear fuel that contains oxide of more than one fissile material. It generally consists of plutonium oxide blended with the oxide of natural, reprocessed or depleted uranium. MOX usually consists of two phases, UO$_2$ and PuO$_2$, and/or a single phase solid solution of (U, Pu)O$_2$. The content of PuO$_2$ may vary from 1.5 wt.% to about 28–30 wt.% depending on the type of nuclear reactor where the fuel is intended to be used. Although MOX fuel could be used in thermal reactors to provide energy, efficient fission of plutonium in MOX could only be achieved in fast reactors as explained using the $\eta$ curve in the previous section.\textsuperscript{25}

The MOX fuel differs from UO$_2$ fuel as the fissile material is primarily $^{239}$Pu, and to a lesser extent $^{241}$Pu, rather than $^{235}$U. Plutonium and uranium have fundamentally different nuclear cross sections that result in the different performance of the materials in a nuclear reactor. Table 1.2 shows the comparison of various important nuclear properties of the fissile nuclides $^{235}$U and $^{239}$Pu.

\begin{table}[h]
\centering
\caption{Comparison of nuclear properties of $^{235}$U and $^{239}$Pu}\label{table:nuclides}
\begin{tabular}{|l|c|c|}
\hline
Parameter & $^{235}$U & $^{239}$Pu \\
\hline
Thermal neutron fission cross section ($\sigma_f$, barns) & 577 & 741 \\
Thermal neutron capture cross section ($\sigma_c$, barns) & 98 & 266 \\
Thermal neutron absorption cross section ($\sigma_a = \sigma_c + \sigma_f$, barns) & 676 & 1007 \\
\hline
\end{tabular}
\end{table}
<table>
<thead>
<tr>
<th>Parameter</th>
<th>$^{235}$U</th>
<th>$^{239}$Pu</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average number of neutrons per fission</td>
<td>2.43</td>
<td>2.87</td>
</tr>
<tr>
<td>Delayed neutron fraction</td>
<td>0.0065</td>
<td>0.0020</td>
</tr>
<tr>
<td>Energy per fission (MeV)</td>
<td>192.9</td>
<td>198.5</td>
</tr>
</tbody>
</table>

Plutonium in comparison with uranium, as tabulated above, has a higher thermal absorption cross section and fission cross section, more number of neutron emissions per fission, a larger energy per fission and a smaller delayed neutron fraction. These different nuclear properties have an impact on the neutron spectrum, reactivity coefficients and absorber effectiveness. The smaller delayed neutron fraction results in changes in the kinetic response of the reactor with the reactor responding more rapidly to reactivity changes. One of the major advantages of MOX fuel is that it facilitates utilizing surplus weapons-grade plutonium peacefully (proliferation resistance).

1.10. Aim and scope of the thesis

The primary mandate for the current research work is to determine the intrinsic dissolution kinetics of the plutonium rich FBR MOX fuels in nitric acid medium under typical PUREX process condition. In the course of fulfilling this mandate, the quantitative effect of various parameters like initial concentration of nitric acid, presence of nitrous acid, temperate, mixing intensity etc., on the dissolution kinetics of various fuel materials employed in this research work would also be studied. But as MOX fuel is precious, one couldn’t afford to carry out many experiments using them for various safety and economic reasons. Thus it was decided to carry out the initial studies with UO$_2$ fuel system for identifying the experimental conditions and methodology to determine the effect of various parameters influencing the rate of its dissolution in nitric acid and also to evaluate the quantitative effect of all these influencing parameters on the reaction rate. As the chemistry of UO$_2$ and MOX fuel systems does not vary much during the dissolution step, this will help in designing the proper dissolution system and also to identify the gross methods of carrying out the experiments. Subsequently based on the results obtained from UO$_2$ dissolution studies, more experiments would be performed using urania ceria simulated MOX fuel systems of various compositions with cerium used as the non-radioactive surrogate for plutonium. The results obtained in these studies will be extended for the (U, Pu) MOX fuel system. Followed by that, required number of limited
experiments will be carried out for the dissolution of (U, Pu) MOX fuel systems in nitric acid under typical PUREX process conditions to evaluate its dissolution kinetics. Finally, using the results of all the experimental studies, model equations will be developed to explain the dissolution behaviour of all the fuel materials in nitric acid medium. The model so developed for the UO$_2$ fuel system first, will be then gradually upgraded with the necessary changes to explain the dissolution behaviour of the MOX fuel material.

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