B. S. Tomar · P. R. Vasudeva Rao ·
S. B. Roy · Jose P. Panakkal ·
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Editors

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Foreword

With the inevitable global thrust towards achieving net zero GHG emissions and the imperative to meet growing energy needs of most of the world as it seeks to improve quality of life of people, a rapid scale up of nuclear energy seems inevitable. Looking at the magnitude of this challenge in run up to clean energy transition, particularly in growing economies as also the unresolved nature of the issue of permanent disposal of spent nuclear fuel that is being used in once through mode, closing the nuclear fuel cycle has become a matter of paramount importance. Leveraging energy from thorium in addition to energy from uranium also necessitates recourse to closed fuel cycle. Thanks to the vision of Dr. Bhabha, consistent and autonomous pursuit of the well thought off three stage nuclear power program and emphasis on self-reliance, India today is among the very few most advanced countries in the world in this sensitive high technology area. Further advances in this area have also opened the possibility of gross reduction in repository requirements for disposal of high-level radioactive waste, reduction in radiotoxicity of such waste to levels comparable with naturally occurring uranium and thorium deposits within a period consistent with institutional life time (say ~ 300 years). As a byproduct of such efforts, several applications of radio-isotopes derived from nuclear waste in the field of health care, industry, agriculture, food technology, etc. have opened up and have been rising steadily. Developments in nuclear fuel cycle are thus destined to cater to global needs in energy, food, healthcare and environmental management.

Activities in nuclear fuel cycle cover a very wide spectrum. The front-end part covers exploration, mining, milling, refining and nuclear fuel fabrication. While the back end comprises of the spent fuel storage, reprocessing, fuel refabrication, byproducts for other applications and radioactive waste management. Both front and back-end activities involve stringent quality as well as nuclear material accounting and control. Although India has a very comprehensive program relating to nuclear fuel cycle it remains an evolving endeavor with many dimensions unfolding progressively. We have large teams of competent and qualified professionals as well as researchers working with synergy in this very specialized area.

In this context, the present book on Nuclear Fuel Cycle will provide the impetus to the professionals working in the field of nuclear science and engineering in updating

their knowledge in the field of nuclear fuel cycle. In addition, it will encourage the students in pursuing nuclear science and engineering by providing basic knowledge on various concepts and strategies employed in nuclear fuel cycle. As the book is written by practicing nuclear specialists, it provides an insider's view on various processes in the nuclear fuel cycle. The introductory chapter provides the general information about the nuclear fuel cycle, its basic framework and strategies being followed world-wide. The chapters on exploration, mining, milling and processing of uranium, nuclear fuel fabrication, quality control of nuclear fuels, post irradiation examination, spent fuel reprocessing and radioactive waste management provide the core knowledge on the various stages of the nuclear fuel cycle. The chapter on thermodynamic aspects of fuel behaviour in the reactor provides basic understanding of the interaction between fuel and clad, swelling behaviour of fuel, fuel—fission products compatibility and thereby helps in predicting long term safety aspects of the reactor. The chapters on accounting and control of nuclear materials, and safety in storage and transport of nuclear materials are equally important in terms of the ascertaining the basic standards in safety, security and safeguards in handling nuclear materials.

I am sure the book will generate wide spread interest among the professionals, engaged in nuclear fuel cycle activities world-wide. Further, it will be useful as a reference book for the budding researchers and professionals who are going to make a career in the field of nuclear science and engineering. I compliment Prof. Vasudeva Rao and his team of editors for bringing out this book on Nuclear Fuel Cycle, which probably is the first of its kind in the world encompassing all aspects of the nuclear fuel cycle including safety, security and safeguards.

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Preface

During the last seven decades, the activities connected with nuclear fuel cycle have grown and spread to several countries. The number of students studying nuclear science and technology and personnel engaged in development of fuel cycle technologies has also increased. A textbook covering all the fundamental aspects of the nuclear fuel cycle for the postgraduate students and researchers in the field of nuclear science and technology has been a long-felt need. A limited number of books are available on nuclear fuel cycle, and these do not have a comprehensive and in-depth coverage. Moreover, they are written more than a decade ago and do not cover the latest developments.

The present book is written keeping in mind the students coming out of universities after their graduation. This book can be prescribed as a textbook for a course on nuclear fuel cycle for postgraduate students. The chapters are designed in such a way that the reader is taken from the fundamentals to the advanced level in a gradual manner. The Chap. 1 explains the nuclear fuel cycle, gives basic information about nuclear fuels and reactors and briefly describes the various aspects of the nuclear fuel cycle and the activities under front end and back end. The Chap. 2 explains the starting point of the fuel cycle covering exploration, mining, milling and processing of uranium, production of metal and advanced fuel materials. Enrichment of uranium using different techniques is also covered in this chapter. The chapter on fabrication of nuclear fuel elements introduces different fuel materials for reactors of various types and describes various types of conventional and advanced fuel elements for commercial (thermal and fast) reactors and research reactors. Quality control techniques, both physical and chemical, used in nuclear manufacturing facilities are explained in the next chapter. The next chapter deals with the phase equilibria, thermophysical and thermochemical properties of nuclear materials with special emphasis on actinide oxides, carbides, nitrides and metal alloys. Chapter 6 explains the various techniques used for postirradiation examination and the data generated useful for the entire nuclear fuel cycle. The chapter on nuclear fuel reprocessing explains the techniques for extraction of valuable plutonium and removal of fission products and minor actinides from the spent (irradiated) fuel.

Radioactive waste is generated in the nuclear fuel cycle by different agencies, and the nature and content depend on various factors. The philosophy and the technology of radioactive waste management are explained in Chap. 8. The philosophy and the techniques used for Nuclear Material Accounting and Control (NUMAC) at various stages of nuclear fuel cycle are explained starting from the fundamentals in Chap. 9. The statistical aspects in nuclear accounting are also covered in this chapter. The transport and storage of nuclear material are governed by international laws stipulated by IAEA, and the safety standards are explained in the next chapter. The various terms used and the classification of radioactive materials and the design criteria for the packages for transport are well explained in Chap. 10. Lastly, Chap. 11 deals with the basic principles of radiation protection, definitions and the effects of exposure to radiation exposure. The safety considerations applicable to mines and mills, fuel fabrication facilities, radiological laboratories, hot cells and waste disposal facilities are also discussed in this chapter.

The authors have also made all efforts to include photographs, schematic drawings and other illustrations for easy understanding of the subject. References, suggestions for additional reading, and wherever possible, exercises are given at the end of the chapters. This book will be useful for any institution (both academic and research) conducting courses on nuclear science and technology and conducting research on related topics. We hope that the international community of academicians and researchers will find this book very useful for updating their knowledge on fuel cycle.

We are grateful to Homi Bhabha National Institute (an institution of Department of Atomic Energy, India, with the status of university) for conceiving this book and bringing together a number of experts both serving DAE and retired from service. We are also thankful to all the authors for recording their valuable knowledge and practical experience in writing this book, and to the reviewers who have gone through the chapters and given their critical inputs to ensure that the book provides a balanced and at the same time adequate details of various aspects of fuel cycle.

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Acknowledgements

The book on nuclear fuel cycle was conceived by Homi Bhabha National Institute (HBNI) to prepare a textbook for postgraduate students, researchers and practicing professionals. The need for such a book has been felt for quite some time, as such a comprehensive description of the principles and processes involved in the entire nuclear fuel cycle has not been covered in any other textbook on this topic so far. We are grateful to Sri K.N. Vyas, Secretary, Department of Atomic Energy and Chairman, Council of Management, HBNI, for his encouragement for writing such books. We are grateful to Dr. Anil Kakodkar, Chancellor, HBNI and Former Chairman, Atomic Energy Commission of India for readily consenting to write the foreword for the book. We have also received encouragement and support from several senior colleagues from DAE; we are particularly thankful to the Directors of Bhabha Atomic Research Centre (BARC) and Indira Gandhi Centre for Atomic Research (IGCAR).

The authors of individual chapters have made the most valuable contributions towards bringing out the book in the present form. The authors drawn from the different R&D institutions of the Department of Atomic Energy are experts in their fields, and their experience in teaching and research is reflected in the respective chapters. We gratefully acknowledge the valuable contributions of all the authors in bringing out this book.

Several experts, from the Department of Atomic Energy, have provided important inputs towards the preparation of various chapters of this book. We would like to acknowledge in particular, Mr. Rajkumar, Mr. Sandeep Sharma, Dr. A. Rao, Dr. Amit Patel, Dr. Mohammed Sirajuddin, Mr. P. M. Khot, Mr. D. P. Rath, Mr. Chetan Baghra, Mr. Nagendra Kumar, Dr. Chiranjit Nandi, Mr. Somesh Bhattacharya and Mr. Mukesh Choudhary, from BARC and Dr. Sekhar Kumar from IGCAR.

The draft of the book was subjected to internal review by experts from India, namely Dr. D. K. Sinha, Director, Atomic Minerals Directorate, Hyderabad (Chap. 2), Dr. C. Ganguly, Former Chief Executive, Nuclear Fuel Complex, Hyderabad and Dr. S. Majumdar, Former Head, Radiometallurgy Division, BARC, (Chap. 3), Shri H. S. Kamath, Former Director, Nuclear Fuels Group, BARC (Chap. 4), Dr. Mrs. Renu Agarwal, Former Scientific Officer, Fuel Chemistry Division, BARC (Chap. 5), Shri S. Anantharaman, Former Head, Post Irradiation Examination Division, BARC

(Chap. 6), Dr. A. Ramanujam, Former Head, Fuel Reprocessing Division, BARC (Chap. 7), Dr. P. K. Sinha, Former Head, Waste Management Division, BARC (Chap. 8), Dr. K. L. Ramakumar, Former Director, Radiochemistry and Isotope Group, BARC and Head, Nuclear Control and Planning Wing, DAE (Chap. 9) and Dr. M. R. Iyer, Former Head, Division of Radiological Physics, BARC (Chap. 10). We gratefully acknowledge the contribution of all the reviewers which provided critical inputs that helped in improving the content and presentation of the book.

All aspects starting from the planning of the book, finalization of contents, review process, etc., were extensively debated by the core team of editors along with the chapter authors through a number of meetings (even during the pandemic period), for which the support provided by HBNI was invaluable. In this regard, we would like to place on record our deep appreciation for the support and cooperation rendered by Dean and Vice Chancellor, Homi Bhabha National Institute, in bringing out this book. Thanks are also due to Dr. Mrs. P. M. Aiswarya, Research Associate, HBNI, for her editorial assistance by way of improving the quality of figures and tables as well as checking and formatting the references.

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About the Editors

Prof. B. S. Tomar is presently Institute Chair Professor at Homi Bhabha National Institute (HBNI), Mumbai. He joined the Radiochemistry Division of Bhabha Atomic Research Centre (BARC), Mumbai, in 1982. He is an expert in nuclear and radiochemistry, radiation detection and measurement, and non-destructive assay of nuclear materials and has published more than 220 research papers in international journals. He has taught the subject of nuclear and radiochemistry at postgraduate level for over 25 years. He superannuated as Director Radiochemistry and Isotope Group, BARC, in December 2017 and later served as Raja Ramanna Fellow, DAE, during 2018–2021. He was Visiting Professor to Technical University, Delft, The Netherlands, during 2007–2008 and Member of the Standing Advisory Group on Safeguards Implementation (SAGSI), IAEA, during 2016–2017.

Prof. P. R. Vasudeva Rao was the Vice Chancellor of Homi Bhabha National Institute (HBNI), a Deemed-to-be University under the Department of Atomic Energy (DAE), India till recently. He earlier served as Scientific Officer with Indira Gandhi Centre for Atomic Research (IGCAR) at Kalpakkam and was its Director, at the time of his superannuation. He obtained his Ph.D. degree in Chemistry from University of Bombay in 1979 for his work on chemistry of actinide elements. Prof. Rao is an expert in Fuel Cycle Chemistry and especially chemistry of fast reactor fuel cycle and is well known for his research on actinide separations. He was responsible for the development of several facilities for R&D on fast reactor fuel cycle at IGCAR. He has over 300 publications in peer-reviewed international journals. He is a Fellow of the Indian National Academy of Engineering as well as the National Academy of Sciences, India.

Dr. S. B. Roy pursued her B.Tech. in Chemical Engineering from Calcutta University in 1981 and obtained doctorate degree in Chemical Engineering from Indian Institute of Technology (IIT) Bombay in 2001. She joined the 25th batch of BARC

training school in 1981 and later joined Uranium Extraction Division of BARC in 1982. She was instrumental in development of uranium fuel of different grades for special applications, process for recovery of uranium from secondary sources like monazite, research, facility development and production management of specific quality nuclear fuel materials for various Indian Research Reactors. Post superannuation, she served as Raja Ramanna Fellow, DAE, and later as Visiting Scientist till November 2020, wherein she contributed in the field of Safety Review and Human Resource Development.

Dr. Jose P. Panakkal joined Bhabha Atomic Research Centre from 16th batch of training school and had been working at BARC in various capacities for 40 years. He superannuated as Head, Advanced Fuel Fabrication Facility, Nuclear Fuels Group, BARC, and was awarded Raja Ramanna Fellowship by DAE on superannuation. He also had worked as Guest Scientist at Fraunhofer Institute of NDT, Saarbrucken, Germany (1987–1989). He was recognized as Ph.D. Guide by Mumbai University and Homi Bhabha National Institute (HBNI). He is specialized in the fabrication of nuclear fuels particularly containing plutonium for both thermal and fast reactors and was in charge of an industrial scale MOX (Mixed Oxide) fuel fabrication facility which has made MOX fuels for various types of reactors (BWRs, PHWRs, and fast reactors). He has also developed new techniques for fabrication, quality control, and non-destructive evaluation of nuclear fuels and material characterization as evident from his 280 publications including 130 publications in international journals and conferences.

Shri. Kanwar Raj graduated with honors in Chemical Engineering in 1972 from Indian Institute of Technology Roorkee, India. Since then, he has worked in the various capacities in the field of design, research & development, operations, and safety analysis of radioactive waste management plants/facilities. Shri Raj has extensive expertise in design, commissioning, and operation of high-level radioactive vitrification systems as well as in evaluation of long-term performance of vitrified waste product under geological disposal conditions. As Chief Scientific Investigator, he has led two BARC teams for studies in this area in collaboration with Kfk/INE Karlsruhe and as a part of coordinated research program of IAEA, Vienna. As Head, Waste Management Division, BARC, Mr. Raj supervised operations of Waste Management Facilities at Kalpakkam, Tarapur, and Trombay Centres of the Department of Atomic Energy, India. He has gained hands-on experience in management of radioactive waste generations from the entire nuclear fuel cycle, viz. radiochemical laboratories, fuel fabrication facilities, research/power reactors, reprocessing plants, etc. His expertise also encompasses management of spent radiation sources from various industries, medical centers, and research institutions. Mr. Raj has experience in preparation of National and International Waste Safety Standards for Indian Atomic Energy Regulatory Board and IAEA. He is the author/co-author of about 80 research papers published in various journals.

A. N. Nandakumar a Ph.D. in Physics from Mumbai University, has 34 years of R&D experience in the Bhabha Atomic Research Centre, Mumbai, India, in the fields of safe transport of radioactive material, calculation of radiation shielding, and calculation of radiation dose under normal and emergency conditions involving radioactive material. He was Chief Scientific Investigator from India for the IAEA Coordinated Research Projects on Development of PSA techniques relating to the Safe of Radioactive Material and on Collection of Accident Data for Quantification of Risk in Transport of Radioactive Material. Subsequently, he was appointed Head, Radiological Safety Division, Atomic Energy Regulatory Board, Mumbai. He worked as Transport Safety Specialist and as Consultant in the Radiation, Transport and Waste Safety Division, IAEA, Vienna. He has published over 60 scientific and technical papers in national/international scientific journals and conference proceedings.

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Chapter 1
The Nuclear Fuel Cycle: Introduction

P. K. Mohapatra and P. R. Vasudeva Rao

1.1 What Is a Fuel Cycle?

With the ever-increasing energy demand due to the rising global population and a growing emphasis on technologies those have minimal carbon footprint, nuclear energy has emerged as an important element of any sustainable solution for meeting energy requirements. Production of nuclear energy is mainly through controlled nuclear fission in a reactor where fissile isotopes of actinides such as ^{235}U, ^{239}Pu, and in some cases ^{233}U are used in the form of a suitable compound (or metal alloy) as the fuel, even though fusion is also a long-term candidate as an energy resource.

The amount of energy extracted from nuclear fuel in a nuclear reactor is expressed in gigawatt-days per metric ton of heavy metal (GWd/tHM). "Burn-up" is a measure of fuel depletion and is also measured as the ratio of the fuel atoms fissioned to the number of initial heavy metal atoms, expressed as atom %.

The life cycle of the material used as the nuclear fuel, starting from the mining of the ore to the fabrication of the fuel, its irradiation in reactor, and subsequent processing of the spent nuclear fuel, is termed as the nuclear fuel cycle. Figure 1.1 provides a depiction of various stages of the nuclear fuel cycle.

The nuclear fuel cycle can be of different types, depending on several factors, of which the following are most important:

(a) fissile nuclide being exploited for the generation of energy through fission (^{235}U, ^{239}Pu, or ^{233}U),
(b) the form of the fuel (chemical as well as physical),
(c) the type of reactor in which the fuel is deployed.

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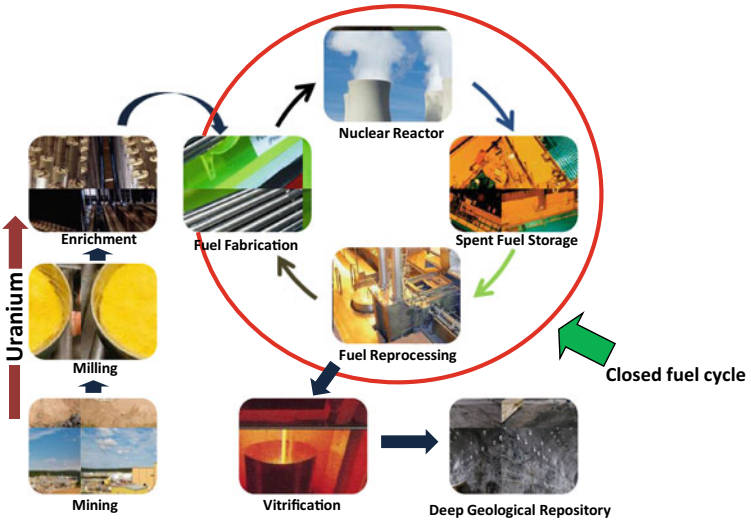

Fig. 1.1 Schematic representation of nuclear fuel cycle

The variations possible in the nuclear fuel cycle based on the factors mentioned above are described below.

(a) The fissile nuclide used, in a way, decides the options for the forms in which it can be used. Natural uranium is used in some reactor systems, while in some others, it is used in an enriched form (uranium with a higher content of U-235 isotope than that present in the natural uranium). Due to neutronic considerations, natural uranium, which consists mainly of U-238 isotope (99.274%) and about 0.72% of U-235 isotope, is used as fuel only in heavy water moderated reactors. The CANDU-type pressurized heavy water reactors (PHWRs) have delivered impressive performance records. However, light water moderated reactors, which use enriched uranium as the fuel, constitute the major fraction of thermal reactor systems, globally. Fast neutron reactors use fuels based on either enriched uranium or plutonium. Recycling of the fuel is especially an important option for reactor systems using fuels based on enriched uranium or plutonium.

(b) Uranium has been used as the fuel in various chemical forms in nuclear reactors. These forms of uranium used in nuclear reactors include the metal, alloy with other elements, and its oxide compound (UO_2), the most preferred form being the oxide (indeed, other forms including aqueous solutions or molten salts are known, but are not discussed in this book). The fuel cycle of the reactor system is influenced by the chemical form of uranium used as the fuel, since uranium does not occur in nature in any of these forms. The uranium obtained from natural resources has to be enriched as per requirement and converted to the desired chemical form, for its use in a reactor. The burn-up that can be reached with the fuel is also influenced by the form of the fuel, in addition to other factors.

The chemical form as well as the burn-up has a great impact on the flow sheet for the recovery of the fuel material from the irradiated fuel, for the purpose of recycling.

(c) Nuclear reactors are of various types, and their most important classification is based on the energy of the neutrons causing fission in the fuel—thermal and fast. World over, with just a few exceptions, nuclear power production is currently based only on reactors where fission is caused by thermal neutrons. The thermal reactors use either natural uranium or slightly enriched uranium as the fissioning material. (Uranium–plutonium mixed oxide has also been used in some cases). On the other hand, fast reactors use fuels of high fissile content (provided by the use of plutonium or enriched uranium) and also reach high levels of burn-up. The fast reactor fuel cycle needs to be closed, in order to be economically sustainable. The fabrication and reprocessing steps for fast reactor fuels need to take into account the presence of high concentration of plutonium. In addition to oxides, other chemical forms such as carbide or metal alloy have been proposed as the fuel for the fast reactor systems, and the fuel cycle has to be designed accordingly to suit the fuel form. For example, metallic fuels are reprocessed through non-aqueous routes.

1.2 Frontend and Backend of Fuel Cycle

1.2.1 Frontend of Fuel Cycle

The nuclear fuel cycle, in a generic sense, starts with the mining of uranium and ends with the disposal of nuclear waste. Some steps may not apply to some fuel cycle types, e.g., for fast reactor fuel cycle, no mining is involved. The steps of mining and milling, conversion, enrichment, fuel fabrication, etc. constitute the "frontend" of the nuclear fuel cycle, while the steps after the discharge of the irradiated fuel from the reactor, such as temporary storage of spent fuel, reprocessing, waste management, etc., are termed as the "backend" of the nuclear fuel cycle.

Uranium-rich minerals are radioactive in nature which is mainly due to the daughter products of uranium derived from radioactive decay processes. The world-wide production of uranium in 2019 amounted to about 54,750 tons [1], and the mined uranium was almost entirely used as the fuel for nuclear power plants. Uranium recovery is done by extraction from ores followed by concentration and purfication. Extraction is done by both excavation and in situleaching (ISL). In general, open-pit mining is used where deposits are close to the surface, and underground mining is used for deep deposits. The mined uranium ores are normally processed by grinding followed by uranium leaching mostly by chemical methods using either an alkali (Na_2CO_3) or an acid (H_2SO_4). The subsequent milling process yields the "yellow cake," which contains uranium as U_3O_8 with >80% uranium content. The milling process concentrates the uranium content by over 800 times from about 0.1% present in the ores. About 200 tons of U_3O_8 are required to keep a 1000 MWe nuclear power

reactor generating electricity for one year [2] (This is lower than the requirement for fossil fuel-based power generation by several orders of magnitude).

The U_3O_8 produced from the uranium mill is required to be enriched in the U-235 content from 0.72% to between 3 and 5% (for light water reactors). The CANDU-type pressurized heavy water reactors (PHWRs) do not require uranium to be enriched; fast reactors may need enrichment as high as 90% depending upon the size of the reactor. The isotope enrichment is usually a physical process (though there are reports of some chemical processes as well) and requires the uranium bearing compound to be in a gaseous form. In view of this, uranium is converted to uranium hexafluoride, which is a solid at ambient temperature and sublimes at 56.5 °C. The conversion step is one of the most important steps in the nuclear fuel cycle. The three major processes used in the isotope separation involved in the enrichment of U are: (i) gaseous diffusion, (ii) centrifugation, and (iii) laser separation. While the gaseous diffusion process is not used any more due to large energy consumption, laser isotope separation has not matured enough to be deployed at a large scale. Therefore, the main enrichment process used in commercial plants is centrifugation, where thousands of rapidly spinning vertical tubes exploit about 1% mass difference between the hexafluorides of two uranium isotopes leading to their effective separation.

The enriched UF_6 cannot be used as such as the fuel in the nuclear reactor and, hence, needs to be converted to ceramic pellets of UO_2 (containing LEU) sintered at >1400 °C. Alternatively, mixed U, Pu oxide (MOX) fuel pellets are fabricated. The chemical quality control (CQC) of the nuclear fuel is one of the major tasks entrusted to laboratory chemists who certify the U, Pu, and impurity content that can be tolerated in the reactor fuel. While the major components are measured by electrochemical methods like potentiometry, bi-amperometry, etc., the trace metallic impurities are determined by a host of techniques such as atomic emission and/or atomic absorption spectroscopy-based techniques, after removal of the matrix. Several critical non-metallic trace impurities (hydrogen, chlorine and fluorine, carbon, nitrogen, oxygen, sulfur, total gas analysis, etc.) are also determined by a variety of analytical techniques as a part of CQC to ascertain the desired level of purity of the fuel. The isotopic composition of actinides (U and Pu) used as major constituent of fuels is another important specification of the fuel.

During fuel fabrication, especially for the plutonium bearing fuels and those having highly enriched uranium, the possibility of a criticality incident needs to be taken into consideration carefully, in the design of equipment as well as processes. This issue is non-existent for the PHWR fuels and of lesser importance for LEU-bearing fuel. The dimensions of the fuel pellets and other components of the fuel assembly have to be precisely controlled to ensure satisfactory performance of the fuel in the reactor core.

Chapter 2 of this book deals with the frontend steps in fuel cycle, including uranium mining, milling, refining, enrichment, etc., in a detailed manner, while Chap. 3 deals with various aspects of the nuclear fuel fabrication.

1.2.2 Backend of Fuel Cycle

1.2.2.1 Irradiated Fuel

The composition of the irradiated fuel discharged from a reactor depends on the starting composition, and the extent to which the fuel has been "burnt." The "burn-up" seen by the fuel is a direct measure of the amount of fission energy that has been extracted from the fuel. A burn-up of 1 atom % of the fissile material corresponds to an energy generation of 10,000 MWd from one ton of fuel. It is important to emphasize that unlike in the fossil fuel-based power generation, in nuclear power generation, the fuel is not burnt out completely in one cycle. It is clear that since natural uranium contains only 0.72% of the fissile isotope (U-235), the burn-up of fuel based on natural uranium should be less than 0.72 atom %. However, because of fissions in Pu-239, generated in situ by neutron absorption in U-238, the burn-up can reach up to 1 atom % (10,000 MWd/t). In typical PHWRs, the burn-up level is of the order of 7000 MWd/t. In light water reactors which use enriched uranium, the burn-up levels reach 6–7 atom %, while in fast reactors, it can exceed 10 atom %. In some cases, up to 20 atom % burn-up has been reached.

After a certain level of burn-up, (corresponding to a typical irradiation period of 3–4 years for LWRs), the fuel has to be discharged from the reactor, since the accumulated fission products can affect the neutron economy. For fast reactor fuels, which reach high burn-up levels, other issues such as fuel clad chemical interactions and fuel swelling limit the fuel burn-up. However, a typical LEU-based fuel containing 5% U-235 discharged from the reactor after a burn-up of 55,000 MWd/t still has about 96% of its original U content (of which <1% constitutes U-235), while the fission products and Pu (activation product) constitute ca. 3 and 1%, respectively. Therefore, the fissile content (including Pu) needs to be recovered for its subsequent use as fuel in reactors. The U-235 content in the spent fuel is still higher than that in natural uranium and, hence, can be reused if subjected to the conversion and enrichment steps as mentioned above. In addition, Pu can be used in MOX fuel to increase the fissile content as required. Thus, it is obvious that irrespective of the type of fuel or reactor, the irradiated fuel is still a valuable source of fissile materials. Even if the uranium is depleted with respect to its fissile content, it can be used to prepare the fuel for a fast reactor by the addition of Pu. Thus, the irradiated fuel discharged from a nuclear reactor is not, strictly speaking, "spent fuel," even though the term "spent fuel" is used in literature, and also in this book, to be synonymous with the discharged fuel.

The process of recovering valuable fissile and fertile materials from the irradiated fuel is called reprocessing. This is briefly discussed in the next section but is dealt with in detail in Chap. 7.

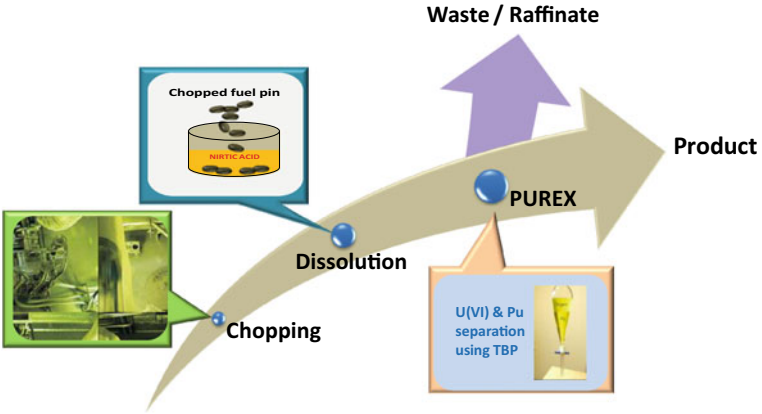

Fig. 1.2 Schematic presentation of spent fuel reprocessing using PUREX process

1.2.2.2 Reprocessing

The process adapted for the separation of U and Pu from the irradiated fuel by different countries is the Plutonium Uranium Redox EXtraction (PUREX) process which is schematically presented in Fig. 1.2. This process uses 30% tri-*n*-butyl phosphate (TBP) in an aliphatic hydrocarbon diluent (such as kerosene or a mixture of normal paraffinic hydrocarbons) for extracting U and Pu preferentially [3]. The first step involves the chopping of the fuel rods followed by their dissolution in 7–8 M nitric acid. The fuel material dissolves in nitric acid leaving behind the "hull" constituting the insoluble part which traps some amount of the radionuclides. The dissolution step is followed by feed clarification (filtration) and feed acidity adjustment followed by the extraction and partitioning steps. The decontamination factor (DF) from the fission products is very high (typically in the range of 10^4–10^6) and the process can be operated continuously for several cycles as required, without any significant change in the efficiency. Chapter 7 gives a detailed account of spent fuel reprocessing which also includes a discussion on the path of the minor actinides and fission products in the PUREX process. The chapter also describes reprocessing of Th-based spent fuel including the THOREX processes. Fast reactor spent fuel reprocessing is quite challenging due to the high Pu content of the fuel and also higher concentrations of the fission and activation products due to the high burn-up. This is also addressed in Chap. 7 along with a brief description of non-aqueous routes for reprocessing.

1.2.2.3 Waste Management

Minor actinides (MA) such as Np, Am, and Cm are streamed out of the PUREX cycle into the "raffinate" (a waste stream) which when concentrated results in the high-level liquid waste (HLLW). The HLLW is responsible for >95% of radioactivity of

all radioactive wastes and, hence, needs safe and acceptable management strategies for the success of the nuclear energy program. The common strategy involves the vitrification (using borosilicate glass) of solid waste oxides in glass blocks and their subsequent burial in deep geological repositories (DGRs). However, such disposal will require a long surveillance period, in view of the very long half-lives of minor actinides and some fission products making the waste management program highly expensive. Also, as the decay of the buried radionuclides takes millions of years, the radiotoxicity of the long-lived radionuclides present in the waste blocks is reduced to a level comparable to that of U from natural sources only after a very long time (Fig. 1.3). This prompted the development of processes for partitioning of minor actinides such as Np, Am, and Cm and fission products such as Cs-137 and Sr-90 which can drastically reduce the average exposure to the operating personnel. The strategy of "Actinide Partitioning" can be followed by the transmutation of the long-lived radionuclides in fast reactors or accelerator driven sub-critical systems (ADS). The "Partitioning & Transmutation" (P&T) strategy is of considerable interest in the long term, as it effectively addresses the concerns about radioactive waste management. However, if "Actinide Partitioning" is linked with the other emerging strategies such as "lanthanide–actinide separation" and "Am–Cm separation," it can have the additional benefit of the recovery of valuables like Am and Cm, which are immensely useful materials.

Apart from specific chemical methods used for various types of radioactive wastes, several physical methods are also used for waste immobilization/compaction prior to their subsurface disposal. The general categorization of radioactive wastes and their classification along with various strategies involved in their safe management are discussed at length in Chap. 8.

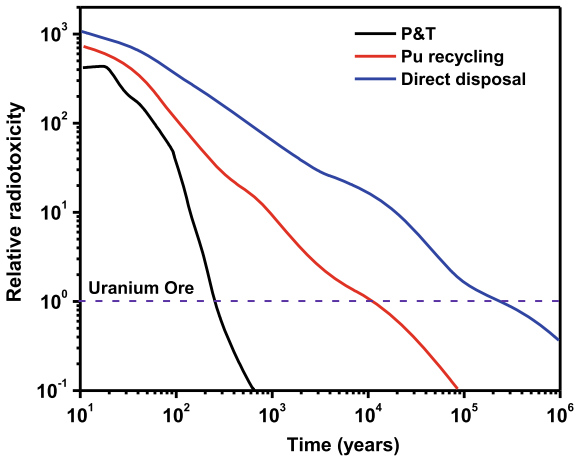

Fig. 1.3 Effect of Actinide Partitioning on the relative radiotoxicity of radioactive waste

1.3 Categories of Nuclear Fuel Cycle Based on Reuse Strategies

Nuclear fuel cycles are categorized into different types based on the different strategies employed on fronts like cost, safety, non-proliferation, and waste management. In general, there are two types of nuclear fuel cycles, viz., the "open" or "once-through" and the "closed" or "recycled." The "twice-through" fuel cycle is a variant that represents limited closure of the fuel cycle. There is also a strategy called "partially closed" fuel cycle which has limited spent fuel recycling option (similar to the "twice-through cycle") and is a concept that is in between the open and the closed fuel cycles.

1.3.1 Open or "Once-Through" Fuel Cycle

In the "open" or "once-through" fuel cycle (Fig. 1.4), the fuel material is used in the nuclear reactor only once; the fuel discharged from the reactor is kept in an "interim storage" facility until most of the short-lived radioactivity has decayed [2, 4]. The fuel can then be disposed into a repository as waste or reprocessed at a later stage for recovery of the fissile material. The open fuel cycle is practiced in the USA, UK, and South Africa where the discharged fuel is subjected to storage for possible vitrification and burial in deep geological repositories (DGRs) at a future date. At present, the irradiated, discharged fuel is stored either at the "at-reactor" (AR) or "away-from-reactor" (AFR) facilities. A block diagram, showing the "once-through" fuel cycle, is given in Fig. 1.4.

1.3.2 Twice-Through Fuel Cycle

In this case, the spent nuclear fuel is reprocessed in order to extract the uranium and plutonium and subsequently recycled once in the reactor. The recovered uranium and plutonium are suitably fabricated as mixed oxide fuel, to replace the enriched uranium oxide fuel. The MA and fission products are sent to the interim storage facility for further processing. The LWR is either fully loaded with MOX fuel or with a combination of MOX fuel bundles (about 30%) and UO_2 fuel bundles. After the second cycle of irradiation, the MOX fuel is not reprocessed and is cooled and stored similar to the "once-through" fuel cycle mentioned above. A schematic representation of the "twice-through" fuel cycle [2, 4] is presented in Fig. 1.5. Such recycling has been implemented in many countries especially with the oxide fuels. The major reason for recycling of the "spent nuclear fuel" (SNF) in the case of the "twice-through" NFC is due to the fact that the SNF still contains ca. 96% of the reusable material (U and Pu) and, hence, is still considered as an energy source due to an

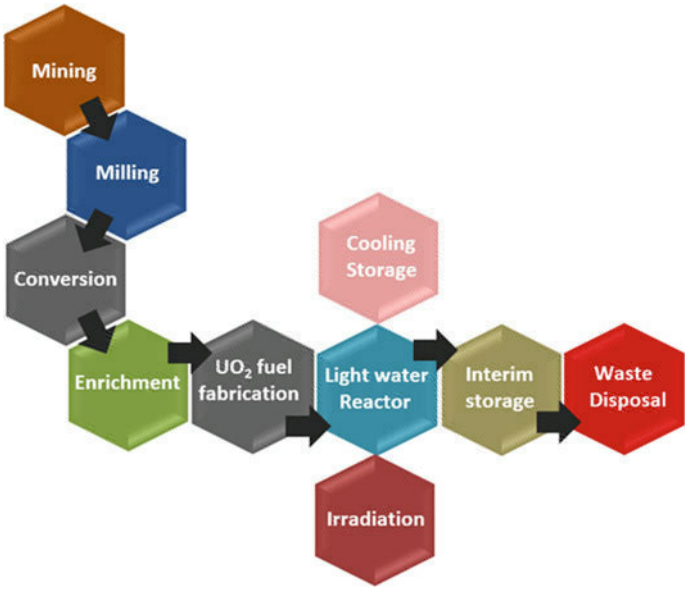

Fig. 1.4 Block diagram of the "once-through" fuel cycle

appreciable fissile material content. Additional cycles in a thermal reactor are not, however, practical due to degradation of the isotopic composition of Pu. As compared to the "once-through" NFC, in which the SNF is stored after use and buried in DGRs after vitrification, the "twice-through" NFC can utilize 17% more natural uranium by the MOX fuel option which can largely discount the heavy enrichment costs. The "twice-through" NFC can further yield a significant reduction in the waste volume (>80%) and radiotoxicity (>90%) of the HLLW.

1.3.2.1 Degradation of Isotopic Composition of Plutonium in Thermal Reactors

It is known that the isotopic composition of plutonium is not significantly changed due to irradiation in fast reactors. Thus, fast reactors can utilize the full energy potential of plutonium through multiple cycles. However, in thermal reactors, the isotopic composition of plutonium is degraded, and the use of plutonium in fuel becomes unviable after a few cycles. As a result, plutonium recovered from irradiated fuel can be used only in one additional cycle of irradiation, in a thermal reactor (twice-through cycle).

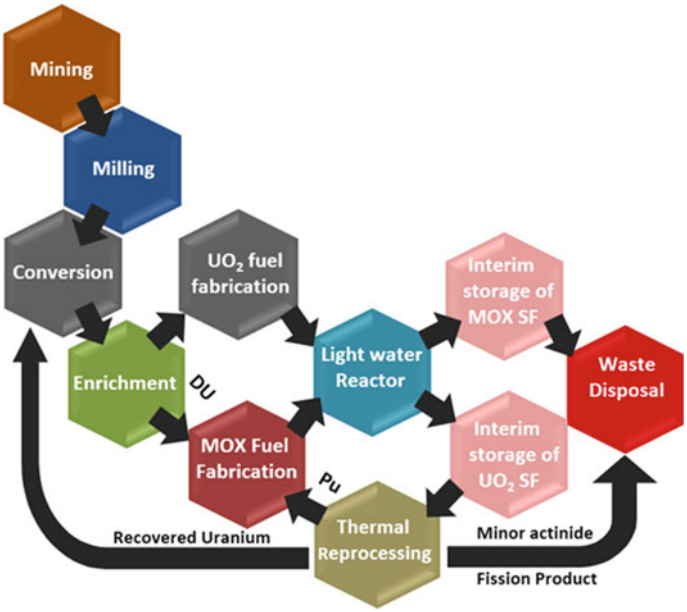

Fig. 1.5 Block diagram of the "twice-through" fuel cycle

1.3.3 Closed or "Recycled" Fuel Cycle

The closed fuel cycle (depicted in Fig. 1.1) is advocated by many countries including France, Japan, India, China, Russia, etc. and has the objective of effective utilization of the fissile material through multiple recycling and reducing the hazardous nature of the radioactive waste to be disposed by recovering the radiotoxic elements. The "closed" or "recycled" fuel cycle requires the development of advanced technologies for a sustainable NFC.

1.4 Open Versus Closed Fuel Cycle Options

The policy choice of the fuel cycle by a country depends on several factors. The common factor is a short-term approach caused by perceptions on economics, proliferation concerns, and assurance of uranium availability. In this approach, recycling of uranium is considered as an economic choice only when the cost of uranium is high. Several countries, therefore, have opted to postpone the implementation of recycling and resorted to store irradiated fuel in interim storages. Another reason for the adoption of open fuel cycle policy by many countries is the concern of nuclear proliferation, as a result of which the recovery of plutonium from the irradiated fuel is discouraged.

On the other hand, it is argued by several countries which advocate the use of a closed fuel cycle that it is a requirement for long-term sustainability of nuclear power. As pointed out in Sect. 1.2.2.1, the fissile content of the irradiated fuel discharged from reactor is quite valuable and does not qualify to be treated as waste material. The effective utilization of the uranium resources in nature is thus possible only through multiple cycles of irradiation. Since uranium resources are non-renewable, one can say that sustainable energy generation through nuclear fission can be realized only through the closed fuel cycle.

However, as discussed above, the number of cycles for which a given fuel material can be irradiated in thermal reactors is limited by the degradation of the isotopic composition of Pu. A high level (near quantitative) utilization of the fuel material through multiple cycles of irradiation is only possible in fast reactors. Fast reactors also need the closed fuel cycle from economic point of view. Thus, it can be argued that fast reactors with closed fuel cycle constitute the long-term sustainable option for nuclear reactors.

The impact of the closed fuel cycle on the long-term management of highly radioactive wastes is another important factor that is in favor of a closed fuel cycle. This aspect is discussed in some detail in the next section.

1.4.1 Impact of Closed Fuel Cycle on Waste Management

To understand the impact of the closed fuel cycle on waste management, one needs to study the composition of the radioactive wastes. The composition of a typical irradiated fuel discharged from the reactor is shown in Table 1.1. From the point of view of the radioactive waste management, one needs to particularly consider the radionuclides with relatively long half-life and significant radiotoxicity. Such nuclides include the isotopes of plutonium and the "minor" actinides (the term minor is used to convey that these elements occur at relatively low concentrations in the irradiated fuel, as compared to the "major" actinides, viz., uranium and plutonium). The radiotoxicity of these radionuclides is such that the wastes containing those are required to be disposed in repositories deep down the earth and not in near-surface repositories. In addition, due to their significant radiotoxicity and long half-lives, the waste form needs to be under surveillance for periods of the order of 10,00,000 years (please recall that the half-life of plutonium-239 isotope, one of the major constituents of the irradiated fuel, is of the order of 24,000 years). These factors not only pose several technological challenges but also considerably increase the cost of the fuel cycle.

The open fuel cycle, where the irradiated fuel as a whole is treated as waste, has a major impact on the radioactive waste management. In the closed fuel cycle, however, uranium and plutonium (in some versions, also the minor actinides) are recovered and recycled, reducing the quantity of the material to be disposed as waste to a very large extent (a factor of four or more). In addition, if the concentration of the radiotoxic long-lived alpha emitters is reduced below certain limits (presently

Table 1.1 Distribution of principal radioactive elements present in irradiated fuel discharged from a nuclear reactor [5]

Burn-up	6700 MWd/T (5 y cooling) (%)	33,000 MWd/T (10 y cooling) (%)
U	98.96	95.5
Pu	0.35	0.85
MAs (Np, Am, Cm)	0.011	0.112
Long-lived FPs	0.044	0.2
Short-lived FPs	0.038	0.17
Stable nuclides	0.60	3.2

accepted limit is 100 nCi/g), the waste can also be disposed near surface. Also, the surveillance period can be reduced to less than thousand years. These factors undoubtedly impact the economics of the fuel cycle.

Further value can be realized from the waste, through separation of radio-cesium (^{134,135,137}Cs), radio-strontium (^{90}Sr), PGMs, etc., which can be used in societal applications.

These variants in the implementation of the closed fuel cycle are discussed in brief in the following section.

1.5 Variants of Closed Fuel Cycle

1.5.1 Closed Fuel Cycle Without MA Recovery

In this type of closed fuel cycle, uranium and plutonium are recovered from the irradiated fuel, and minor actinides (Am, Cm, Np, etc.) and fission products are rejected into the raffinate stream for their interim storage and ultimate vitrification and burial in the DGRs. The plutonium recovered can be recycled in thermal or fast reactors. Depleted uranium can be used in MOX fuels (uranium–plutonium mixed oxide) constituted for thermal (or fast) reactors. This was the principle followed initially in countries with poor uranium reserves where the objective was to recover the depleted uranium and plutonium from the irradiated fuel for their subsequent utilization in the MOX fuel-based program.

1.5.2 Closed Fuel Cycle with MA Recovery

In Closed fuel cycle this type of fuel cycle, the HLLW arising out of the primary separation process (wherein uranium and plutonium are separated from the irradiated fuel through a solvent extraction scheme, for recycling) is subjected to another separation scheme wherein the minor actinides are recovered. The most well-developed schemes

for the recovery of minor actinides also recover the lanthanides along with the former because of the similarity in chemical properties of the trivalent actinides and lanthanides. The minor actinides recovered can be subsequently burnt in accelerator-driven sub-critical systems (ADSS) or fast reactors [6]. In this context, lanthanide–actinide separation [7] and subsequently, Am–Cm separation [8] are emerging as important steps for a meaningful transmutation of the long-lived actinides. It is important, however, to point out that in order that waste immobilization and its ultimate disposal in the DGRs are simplified, the recovery of the radiotoxic minor actinides has to be near quantitative and the remnant waste should meet the requirements for categorization as "non-alpha" waste.

1.5.3 Closed Fuel Cycle with MA and Fission Product Recovery

As shown in Table 1.1, the fission products constitute ca. 0.6% of the irradiated fuel out of which the long-lived fission products constitute only approx. 0.04% of all elements. However, the major amount of radioactivity of the HLLW is due to the presence of the highly radioactive fission product nuclides such as ^{137}Cs and ^{90}Sr, which are mainly responsible for the radiation dose burden. Apart from these, there are other radionuclides such as ^{106}Ru with moderate radioactivity and long-lived fission product nuclides such as ^{99}Tc, ^{93}Zr, ^{135}Cs, ^{129}I, and ^{107}Pd. Out of these, ^{137}Cs and ^{90}Sr can find use in societal applications such as in irradiators (^{137}Cs), nuclear medicine (^{90}Y), and as power sources (^{90}Sr), and hence, there have been efforts to separate these radionuclides from the HLLW prior to the radioactive waste vitrification. Also, long-lived fission product nuclides can be separated and transmuted to shorter-lived radionuclides for an effective reduction in the period of surveillance of the vitrified waste blocks. In India, the present strategy is to separate the fission product radionuclides prior to the Actinide Partitioning step (of the P&T strategy) so that the radioactive waste has very low radiotoxicity which can be conveniently managed through vitrification and burial in DGRs.

1.6 Challenges in the Development of Closed Fuel Cycle

The closed fuel cycle, when suitably designed, can enable efficient utilization of the uranium (fissile content) resources (in principle one can visualize utilization of >90% of uranium in fast reactors over several cycles) and thus provide a sustainable option for the development of nuclear energy. However, except for some, most countries have not developed the option of closed fuel cycle. The most important reason for support to the open fuel cycle option arises from the concern of proliferation of nuclear weapons that can be caused by the separation of plutonium in its pure form.

Also, the backend of the fuel cycle involves complex technologies which require chemical engineering operations in hot cells, and very few countries have been able to establish infrastructure to develop the technology. For plutonium utilization through closed fuel cycle, the fuel fabrication facilities also need to be housed in glove box containment, a requirement that does not exist for uranium based fuels. This needs additional infrastructure including human resources trained for remote operations and maintenance. Thus, only those countries with a strong human resource base and mastery of the complex technologies involved in nuclear recycle can opt for the closed fuel cycle.

1.7 International Scenario on Fuel Cycle

Very few countries have programs related to all stages of nuclear fuel cycle. Some countries possess uranium resources but do not have nuclear energy programs; some countries have nuclear reactors but do not have uranium resources and therefore, have to depend on the supply of fuel under international agreements. Only a few countries have nuclear reactors as well as fuel cycle programs. These include France, India, Russia, Japan, UK, and China. The profiles of nuclear fuel cycle programs of various countries have been summarized in Ref. [9].

As mentioned earlier, the most common fuel cycle option internationally is the "open" or "once-through" fuel cycle. The backend of the fuel cycle, however, is a subject of intense research in several countries, indicating the importance of closed fuel cycle as a long-term option. Partitioning of minor actinides from the HLLW is one of the important areas pursued for the development by several countries [10]. Minor actinide separation is proposed through several advanced aqueous partitioning methods using a variety of extraction systems. Several variants of separation schemes are being studied, and these are described in Chap. 8. These include the separation of all the actinides as a group (GANEX), separation of uranium in one cycle followed by the separation of other actinides as a group, etc. Many of these processes also separate the lanthanides along with the actinides, due to the similarity in the properties of the lanthanides and the transplutonium actinides. Variants of the separation processes under development include processes which separate the lanthanides from the minor actinides [7]. Such separations can further reduce the volume of the waste, reducing the demand of space of the repository.

Under the Global Nuclear Energy Partnership (GNEP) program led by the USA, efforts are being made to ensure that countries with developed nuclear technological base provide safe nuclear power to other countries without compromising on proliferation concerns. There is a renewed interest in the development of new separation schemes for the reprocessing of spent nuclear fuels. In this approach, the recycling of spent nuclear fuel is proposed to be carried out without separating out pure plutonium thereby reducing the proliferation concerns.

From the above, one could conclude that the fuel cycle is not only a complex issue that is addressed by country-specific approaches, but will continue to be a