This book addresses the various issues associated with the buildup of reactor-grade plutonium in nuclear power reactors and analyzes its properties critical to the nuclear proliferation risks. An independent “inventory check” of reactor-grade plutonium production is performed according to the types of plutonium, nuclear reactors and countries, and a new approach to the identification of proliferation risks associated with different types of reactor-grade plutonium is presented.

The book is intended for those who deal with non-proliferation and security issues, nuclear power engineering and production, and for students and professors.

PREFACE

This book addresses the issues associated with the production of reactor-grade plutonium in nuclear power reactors, which are used to generate electricity. Properties of reactor-grade plutonium, which are critical in assessing nuclear proliferation risks, are analyzed.

Plutonium is the first “man-made” chemical element (its atomic number in the periodic table of elements is 94), which does not occur in nature and which, however, has been produced by the mankind. For this purpose, a unique facility – a nuclear reactor – has been developed, the first prototype of which was launched in December 1942. Soon after that the United States started large-scale production of weapon-grade plutonium, and in July 1945 the fact of nuclear weapons development was broadcasted with the first nuclear explosion in the Alamogordo Desert. Following this, the USSR, Great Britain, France and China entered the nuclear arms race. The nuclear weapon stockpiles of these five countries use plutonium as a major material. Of the four “unofficial” nuclear armed states, India, Israel and North Korea acquired the nuclear-weapons status through plutonium production. Although Pakistan initially relied on highly enriched uranium, it has substantially increased weapons-related plutonium production since then. Thus, plutonium is a byword for the material essence of nuclear weapons.

As nuclear reactors generate large quantities of energy during their operation, it was recognized at the dawn of nuclear age in the 1940s that this energy can also be utilized for peaceful purposes – primarily, for the production of electricity. This instigated the process of conversion of nuclear reactors from producers of weapon-grade material into producers of electricity. A lot of designs and modifications of nuclear power reactors have been developed, and peaceful nuclear programs have grown with time to exceed those aimed at nuclear weapons development.

All nuclear power reactors produce plutonium, which is called reactor-grade plutonium as distinct from weapon-grade plutonium produced by dedicated plutonium production reactors. Whereas the primary purpose of plutonium production reactors is to produce plutonium, nuclear power reactors are intended for power generation, while plutonium is a byproduct of this process.

The International Atomic Energy Agency (IAEA) was established for the purpose of promoting international cooperation in the area of peaceful uses of nuclear energy while preventing the diversion of nuclear material from peaceful nuclear activities to the manufacture of nuclear explosive devices. This organization gives special attention to the production and handling of materials, which can be used for manufacturing nuclear explosive devices, in particular, plutonium. Corresponding facilities in the non-nuclear-weapons states parties of the Non-proliferation Treaty (NPT) are placed under the IAEA safeguards, and are subject to monitoring and inspections.

Over the last 30 years, nuclear weapons have been developed by five states outside the official nuclear club. In addition to India, Pakistan, Israel and North Korea, they were developed in South Africa, which however gave up nuclear weapons later and eliminated its small nuclear

stockpile under the supervision of IAEA. Several states have sought to start their nuclear weapons programs, but did not cross the “red line”. Some states have advanced their nuclear technology to the level allowing them to manufacture nuclear weapons in a year or two, if this becomes necessary and if a respective political decision is taken. All this is indicative of certain instability of the existing nuclear non-proliferation regime.

Proliferation risks are determined by political, military and technological incentives to develop nuclear weapons and by the proliferation potential.

The proliferation potential is determined by the availability of nuclear materials, nuclear technologies, special technologies and personnel needed for the development of nuclear weapons. Since the mid-20th century, all the constituents of this potential have been growing continuously. This growth is largely associated with the development of nuclear power engineering, and is basically a by-process related to the dual-use nature of nuclear technologies. Among the major constituents of this potential, let us emphasize the growth in production and stock of reactor-grade plutonium as a by-product of nuclear power engineering.

What determines the relevance of this book? First of all, it is the fact that plutonium produced in nuclear reactors is a mixture of five isotopes Pu-239, Pu-240, Pu-241, Pu-242 and Pu-238 (in the descending order of their content in most plutonium materials), the isotopic composition of which has a considerable influence on the properties of plutonium. There exists a stereotype, according to which weapon-grade plutonium should contain at least 93% plutonium-239 and no more than 7% plutonium-240, while all the other combinations are different types of plutonium. As any stereotype dating back to the early 1960s, it does not completely correspond to the situation in the early 21st century and may lead to the underestimation of the nuclear weapons proliferation risks.

Within this study, we made an attempt to analyze the properties of all major isotopic compositions of reactor-grade plutonium and assess comparative proliferation risks associated with these materials. At the same time, we conducted an independent “inventory check" of the production of such plutonium materials by their type, the type of nuclear reactor and by countries. We hope that this study will contribute to the strengthening of nuclear non-proliferation, probably within the framework of the currently discussed Fissile Material Cut-Off Treaty.

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It is important to note that all the assessments and conclusions presented in the book are the sole responsibility of the authors. They in no way reflect the views of the John D. and Catherine T. MacArthur Foundation or any other institutions.

CONTENTS

INTRODUCTION

One of the major risks of nuclear proliferation is associated with large quantities of plutonium of different isotopic compositions produced in commercial nuclear power reactors. Such plutonium is commonly referred to as “civilian” or “reactor-grade” plutonium. In the context of the nuclear proliferation threat, relative advantages and drawbacks of possible nuclear power engineering roadmaps are assessed subject to a number of factors, such as the volume of plutonium produced, its quality (isotopic composition), choice of technology for spent nuclear fuel (SNF) handling and disposal, procedures for accounting, control and protection of plutonium etc. There are a lot of studies devoted to different aspects of the “plutonium proliferation” problem. Among the best-known of them are the studies of the Institute of Science and International Security (ISIS) in Washington. This institute issues annual updates of estimated global plutonium stocks, including plutonium buildup, separation of plutonium from spent nuclear fuel, its use in MOX fuel, and distribution by countries. A detailed analysis of both global weapon-grade and reactor-grade plutonium stocks up to 1996 is provided in [1]. A wide range of scientific and technological issues related to plutonium properties have been discussed in detail in [2].

Any plutonium produced in nuclear reactors is a mixture of five isotopes: Pu-239, Pu-240, Pu-241, Pu-242 and Pu-238 (in the descending order of their content in typical plutonium materials). These isotopes have different physical properties, which directly determine the quality of plutonium material. This book presents an approach, which makes it possible to compare plutonium materials of different isotopic composition from the standpoint of nuclear proliferation threat, and provides examples of its application. In this book, we also discuss the influence of the isotopic composition on the following properties of plutonium material:

- critical mass properties;
- processing characteristics; and
- radiation safety.

Note that we do not draw absolute conclusions on the quality of some or other plutonium material. Instead, we compare relative properties of different types of plutonium. Such comparative properties of plutonium materials with respect to some or other parameters can generally be represented in the quantitative form.

An important factor is that one of the major plutonium isotopes, Pu-241, has a comparatively short lifetime (its half-life is $T_{1/2}$ = 14.4 years). Then it becomes americium-241 through beta decay. This process entails considerable changes in the isotopic composition of plutonium during long-term storage of spent nuclear fuel. To a certain degree, this also applies to the isotope Pu-238 ($T_{1/2}$ = 87.7 years), which becomes uranium-234 through alpha decay.

The processes of radioactive decay in the plutonium material separated from spent nuclear fuel are of special importance, because they result in radiation effects on the crystalline structure of plutonium, buildup of other actinides and helium isotope He-4, release of heat etc. These issues

can in particular be important for plutonium metal, which is commonly associated with the risk of manufacturing nuclear explosive devices. In this study we consider the proliferation risks associated with reactor-grade plutonium metal in the δ-form.[1]

The following categories are used rather often for plutonium classification [1]:

- weapon-grade plutonium (WGPu) contains no more than 7% of the isotope Pu-240;
- fuel-grade plutonium (FGPu) contains more than 7%, but less than 18% of the isotope Pu-240;
- reactor-grade plutonium (RGPu) contains more than 18% of the isotope Pu-240.

Any plutonium material contains the isotopes Pu-239, Pu-240, Pu-241 and Pu-242 produced in a chain of successive neutron captures and radioactive decays on the nuclei of uranium isotope U-238, and the isotope Pu-238 is produced mostly in a chain of successive neutron captures and radioactive decays on the nuclei of uranium isotope U-235 and partly in a chain of captures and decays on the nuclei of U-238. The isotopic composition of produced plutonium depends heavily on the burnup of nuclear fuel, isotopic composition of uranium fuel (i.e. initial enrichment in the isotope U-235), and on the type of nuclear reactor, which determines its neutron spectrum.

The above classification uses the content of the isotope Pu-240 as the key parameter to determine the quality of plutonium material. While being historically attributed to nuclear weapons programs in some states, this parameter can hardly qualify for playing such a key role (in terms of nuclear non-proliferation). According to our analysis, the key parameters critical to the risk of nuclear proliferation are the content of the isotopes Pu-241 and Pu-238.

Here, we consider the issues related to the isotopic composition of plutonium produced in thermal reactors, which constitute nearly the entire fleet of nuclear reactors in the world. We do not address the issues related to possible plutonium production in fast reactors.

Major types of nuclear power reactors include:

- PWR – Pressurized Water Reactor, uses ordinary light water as both coolant and neutron moderator; uses low enriched uranium oxide fuel with a typical content of U-235 from 3.2% to 4.4%;
- BWR – Boiling Water Reactor, uses ordinary light water as both coolant and neutron moderator; uses low enriched uranium oxide fuel with a typical content of U-235 from 2.5% to 3%;
- PHWR – Pressurized Heavy Water Reactor, uses heavy water as both coolant and neutron moderator; uses natural uranium oxide fuel;
- GCR – Gas-cooled Reactor, uses carbon dioxide (helium can also be used) as coolant and graphite as the neutron moderator; uses natural uranium metal fuel;
- LWGR – Light-Water Graphite Reactor, uses ordinary light water as coolant and graphite as the neutron moderator; uses low enriched uranium oxide fuel with a typical content of U-235 from 1.8% to 2%;

[1] Plutonium has six different structural modifications or forms that have significantly varying densities and crystal structures. Nuclear weapons usually use metallic plutonium in the δ-form.

- AGR – Advanced Gas Reactor, uses carbon dioxide as coolant and graphite as the neutron moderator; uses low enriched uranium oxide fuel with a typical content of U-235 about 2%.

As of the end of 2004, 438 thermal nuclear power reactors were in operation worldwide with a total net installed electricity generating capacity of P_{el}^{Σ} = 366 GW and 103 reactors with P_{el}^{Σ} = 33.4 GW, which have been in the state of long-term or permanent shutdown.

The total estimated quantity of reactor-grade plutonium produced in these reactors is approximately 2000 tons. Annually the global stockpile of reactor-grade plutonium grows by about 106 tons.

Table 1 shows the number (*N*) and net installed electricity generating capacity (*P*) of operational and shutdown nuclear reactors by types of reactors as of the end of 2004.

Table 1. Distribution of global nuclear electricity generating capacity by types of reactors

Reactors		PWR	BWR	PHWR	GCR	LWGR	AGR	Total
Operational	*N*	267	93	40	8	16	14	438
	P, GW	240.9	82.7	20.5	2.3	11.4	8.4	366.2
Shutdown	*N*	27	19	16	33	7	1	103
	P, GW	13.8	4.55	4.15	5.9	4.95	0.03	33.38

Table 1 shows that PWR and BWR light-water reactors are the most prevalent commercial power reac6tors in the world (88% of operational electricity generating capacity), but the contribution of heavy-water reactors is also significant.

An important fact in the context of nuclear proliferation is that SNF of GCRs cannot be stored for a long period of time and has to be reprocessed with separation of plutonium. Spent fuel of light-water reactors is also partially reprocessed.

As part of this study, an approximate numerical model was developed to determine the quantity and the isotopic composition of plutonium produced by different types of reactors. Its results have been used to analyze the risks of proliferation associated with reactor-grade plutonium.

Of importance for assessing the “proliferation potential” of any type of reactor-grade plutonium is the comparison of its properties with those of WGPu. Different authors provide data on the isotopic composition of WGPu in their papers. For example, according to Ref. [1], WGPu contains 93% Pu-239, 6.5% Pu-240 and 0.5% Pu-241; in Ref. [3] WGPu is said to contain 93.5% Pu-239, 6% Pu-240 and 0.5% of other plutonium isotopes; and according to Ref. [4], WGPu contains 93.8% Pu-239, 5.8% Pu-240, 0.35% Pu-241, 0.022% Pu-242 and 0.012% Pu-238. One can see that the isotopic compositions of WGPu given in these papers are close to each other. In the present study, for simplicity of comparison, we will use the isotopic composition of WGPU from Ref. [1].

As the nuclear power industry has been developing since the 1960s, there is now a large stock of reactor-grade plutonium produced dozens of years ago. Although most of such plutonium

stays in SNF, some of it has been separated. Over this period, most of the originally contained isotope Pu-241 decayed (this also applies partly to the isotope Pu-238), and the quality of the plutonium material improved. This aspect, which is essential for nuclear non-proliferation, is also addressed in this study.

1. CRITICAL MASS

Critical mass parameters constitute the most important characteristic of the material, which determines its weapons-related "quality".

At present, there is a lot of data on the critical mass of fissionable isotopes, and plutonium isotopes belong to those studied particularly thoroughly. Data of different up-to-date studies on the critical mass of plutonium isotopes are in a fairly good agreement with each other. We will use data from Ref. [5] by U.S. scientists from the Oak Ridge National Laboratory (ORNL). In 2000, they performed simulations using the ENDF/B-6 library of nuclear data. Results of critical mass simulations for bare spheres of plutonium isotopes are presented in Table 2.

Table 2 also presents densities of the materials under study. It is well-known [3] that the most US nuclear weapons use gallium-stabilized δ-form of metallic plutonium having a density of $\rho = 15.8$ g/cm^3 [3, 6]. The last row in Table 2 provides critical masses of plutonium isotopes for the density of plutonium in the δ-form recalculated as follows:

$$M_{cr}(\rho) = \left(\frac{\rho_0}{\rho}\right)^2 M_{cr}^0 .$$

Table 2. Critical masses of plutonium isotopes

Isotope	Pu-239	Pu-240	Pu-241	Pu-242	Pu-238
M_{cr}^0, kg	10.1	36.95	13.02	85.35	9.75
ρ_0, g/cm^3	19.851	19.934	19.84	20.101	19.84
$M_{cr}^{\delta\text{-form}}$, kg	15.94	58.81	20.53	138.14	15.37

In order to estimate the critical mass of a mixture of different isotopes, we will use an approximate relationship, which can be derived from elementary considerations:

$$M_{cr}^{-1/3} = \sum_i \alpha_i \left(M_{cr}^i\right)^{-1/3} ,$$

where α_i is the mass fraction of the *i*-th isotope of plutonium in the mixture; M_{cr}^i is the critical mass of the *i*-th isotope of plutonium.

As a rule, when fuel of nuclear power reactors reach its rated burnup, the isotopic composition of reactor-grade plutonium lies in the range indicated in Table 3. This table also presents a typical isotopic composition of plutonium in spent PWR fuel calculated using the developed approximate numerical model.

According to these ratios, the critical mass of PWR plutonium from Table 3 is 23.62 kg, which is 1.38 times larger than the critical mass of WGPu M_{cr}^{WGPu}.

Table 3. Isotopic compositions of reactor-grade plutonium at the rated burnup of reactor fuel

Isotope	Pu-239	Pu-240	Pu-241	Pu-242	Pu-238
Content, %	50-70	20-33	4-18	0.5-6	0.05-2
Pu(PWR), %	55.85	21.9	13.75	6.75	1.75

Table 4 presents the critical mass of a mixture of two plutonium isotopes, Pu-239 and Pu-240, as a function of the Pu-240 content, which underlies the conventional classification of plutonium.

Table 4. Dependence of the critical mass of plutonium on the content of Pu-240

Pu-240, %	0	7	10	18	25	30	40	50	100
M_{cr}, kg	15.94	17.18	17.76	19.42	21.03	22.30	25.17	29.66	58.81
$M_{cr} / M_{cr}^{\mathrm{WGPu}}$	0.93	1	1.04	1.14	1.23	1.28	1.47	1.735	3.44

It follows from Table 4 that the threshold values of the Pu-240 content, α = 7% (which distinguishes WGPu from FGPu) and α = 18% (which distinguishes FGPu from RGPu), are in no way distinctive in terms of critical mass. The critical mass of plutonium containing α = 25-30% of the isotope Pu-240 is just slightly different from that of WGPu. Thus, the conventional thresholds to distinguish FGPu and RGPu are unreasonable in terms of critical mass.

Another physical property to determine the quality of a fissionable material is the neutron multiplication rate in infinite medium χ_0 defined as

$$\chi_0 = \frac{d \ln n}{dt},$$

where n is the neutron density. The values of χ_0 for different plutonium isotopes are given in Table 5 (ρ_0 = 15.8 g/cm^3).

Table 5. The values of neutron multiplication rate in infinite medium

Isotope	Pu-239	Pu-240	Pu-241	Pu-242	Pu-238	Pu(PWR)	WGPu
χ_0, 1/ns	0.25	0.149	0.201	0.098	0.271	0.211	0.243

The following relationship is valid for a mixture of isotopes:

$$\chi_0 = \sum_i \alpha_i \chi_0^i; \quad \chi_0 \sim \rho .$$

Table 5 provides the values of χ_0 for PWR plutonium (see Table 3) and WGPu calculated using this relationship. It follows from Table 5 that $\chi_0(\mathrm{Pu(PWR)})/\chi_0(\mathrm{WGPu}) \cong 0.87$.

Table 6 shows the dependence of χ_0 on the content of Pu-240 for a mixture of Pu-239 and Pu-240. These data indicate that the threshold values of the Pu-240 content, α = 7% and α = 18%, are in no way distinguished in terms of the neutron multiplication rate.

Table 6. Dependence of the neutron multiplication rate χ_0 in infinite medium on the content of Pu-240

Pu-240, %	0	7	10	18	25	30	40	50	100
χ_0, 1/ns	0.25	0.243	0.24	0.232	0.225	0.22	0.21	0.2	0.149
χ_0 / χ_0^{WGPu}	1.029	1	0.988	0.955	0.926	0.905	0.864	0.823	0.613

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