## 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  $\delta$ -form.<sup>1</sup>

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%;

<sup>&</sup>lt;sup>1</sup> 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  $P_{el}^{\Sigma} = 366$  GW and 103 reactors with  $P_{el}^{\Sigma} = 33.4$  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.

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

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.