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Research Article

Radiation characteristics of reactor grade platinum group metals*

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Abstract

The paper examines the radiation characteristics of noble platinum group metals (PGMs) extracted from spent nuclear fuel (SNF) of the VVER-1000 reactor. These are the so-called reactor-grade ruthenium, rhodium and palladium. PGMs are radioactive when extracted from SNF, but after the decay cooling of ruthenium for about 27 years, and of rhodium for about 13 years, they can be used in unlimited quantities. There is no sense in decay cooling of reactor grade palladium due to its radioactive isotope ^{107}Pd having a half-life of 6.5 million years. As specified by regulatory documents, such palladium can be freely used only in quantities of up to 34 g. Pd is a soft beta emitter with a maximum beta particle energy of 34 keV. The calculation results show that the mean free path of beta particles from ^{107}Pd in palladium metal is 0.8 μm, so reactor-grade palladium emits only from the surface layer, and other electrons are absorbed in the material itself. The mean free path of electrons with an energy of 34 keV in biological tissue is about 20 μm, which does not exceed the thickness of the skin epidermiscorneous layer. Calculations have shown that the equivalent dose rate (EDR) on the surface of reactor-grade palladium is 0.04 μSv/h, which is below the public EDR value. As a result, a conclusion is made that reactor-grade palladium does not pose a danger in the event of external contact.

Keywords

spent nuclear fuel, noble metals, platinoids, platinum group metals, PGMs, reactor grade PGMs, man-made PGMs, ruthenium, rhodium, palladium, spent nuclear fuel, SNF

Introduction

Spent nuclear fuel (SNF) contains noble platinum group metals (PGM), including ruthenium (Ru), rhodium (Rh), and palladium (Pd) (Pokhitonov 2020). The amount of these depends on the type of nuclear fuel and its burnup depth. For example, platinum group metals account for more than 10% of fission fragments in the VVER-1000 reactor fuel with a burnup depth of 50.7 MW·day/kgHM, which makes about 5.6 kg/t SNF (Kovalev et al. 2024). When in the BN-1200 reactor core with a burnup of 99.2 MW·day/kgHM, their quantity amounts to 16.3 kg/t SNF. This value was obtained by the authors by calculations using a BN-1200 reactor model (Kovalev et al. 2023) for the Serpent code (Leppanen et al. 2015).

The Russian Federation has opted for a closed nuclear fuel cycle (Strategy for the development of nuclear energy in Russia until 2050 and prospects for the period until 2100, 2018). SNF is reprocessed, and uranium regenerate, plutonium, minor actinides, and fission products are extracted. Fission products are high-level waste (HLW) to be vitrified. The concentration of PGMs in HLW solutions

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reaches 850 to 1500 mg/dm^3 (Batorshin et al. 2015). In the process of vitrification, the presence of PGMs leads potentially to a number of problems (Labe et al. 2014; Onishi et al. 2017; Lopukh et al. 2023). For example, a melting temperature of 1100 to 1200 °C causes phases of solid solutions to form based on ruthenium oxide and elementary rhodium and palladium. Notably, the morphology of the particles formed is highly diverse. In the absence of sufficient convective mixing in the melt, a layer is formed at the furnace bottom, in which the viscosity and conductivity can increase tenfold compared to the total glass quantity. Such gradual accumulation of the solid metal phase on the electric furnace bottom will inevitably lead to its failure. In addition to the formation of a highly conductive layer, a major problem occurs associated with the drain hole plugging in the process of the melt discharge through the bottom. To address such problems, methods have recently been developed to extract PGMs prior to the vitrification process (Mishima et al. 2020; Wang et al. 2020; Davydova et al. 2022).

Platinum group metals extracted from SNF have multiple commercial applications, but it should be noted that they contain radioactive isotopes. This paper discusses the extent of hazard posed by PGMsextracted from SNF.

Composition of reactorgrade PGMs

The composition of reactorgrade PGMs in VVER-1000 SNF was calculated using a computational neutronic model of the reactor core (Kovalev et al. 2023) used in the Serpent code (Leppanen et al. 2015). Table 1 presents the estimated content of ruthenium, rhodium and palladium in one ton of SNF.Table 2 presents the calculated isotope composition of PGMsisolated from SNF taking into account its 10-year decay cooling prior to reprocessing.

Radioactivity of reactorgrade PGMs

Ruthenium, rhodium and palladium isolated from SNF contain radioactive isotopes. Table 3 presents the characteristics of radioactive isotopes in PGMs.The specific activity of each isotope is given for the isolated element. It needs to be noted that regulations require reactorgrade PGMs to be accounted and controlled in the national RS and RW accounting and control system, and a permit is required from executive power bodies responsible for national sanitary and epidemiological supervision to use reactorgrade PGMs while the activity and specific activity are greater than or equal to both the minimum significant activity (MSA) and the minimum significant specific activity (MSSA) (par. 3 of NP-067-16[1]/NRB-99/20092[2], Appendix 7, pars. 3 and 4). Also given are the MSA and MSSA for radioactive isotopes and the additional cooling time, after which the isolated elements can be used in an unlimited quantity.

Table 1. Calculated content of ruthenium, rhodium and palladium in one ton of SNF

Element	Weight, kg/t SNF
Ruthenium	3.04
Rhodium	0.60
Palladium	1.94

Table 2. Calculated isotope composition of PGMsisolated from VVER-1000 SNF taking into account its 10-year decay cooling prior to reprocessing

Ruthenium isotopes	Weight, %	Rhodium isotopes	Weight, %	Palladium isotopes	Weight, %
^{100}Ru	5.34	^{102}Rh	<0.01%	^{104}Pd	17.58
^{101}Ru	33.99	^{102m}Rh	<0.01%	^{105}Pd	26.92
^{102}Ru	35.96	^{103}Rh	100.00%	^{106}Pd	26.49
^{104}Ru	24.71			^{107}Pd	15.28
^{106}Ru	0.01			^{108}Pd	10.24
				^{110}Pd	3.49

In the process of beta decay, ^{106}Ru transforms into ^{106}Rh with a half-life of 29.8 s, which disintegrates into stable ^{106}Pd almost immediately. In this regard, the^{106}Rh isotope in isolated rhodium, separately from ruthenium, can be ignored.

It can be concluded from Table 3 that PGMs isolated from SNF can be freely used in the following quantities: ~12 μg (ruthenium), ~ 450 g (rhodium), and ~34 g (palladium). Having been cooled longer (ruthenium for ~27 years, rhodium for ~13 years), they can be used in unlimited quantities. It makes no sense to cool reactorgrade palladium.

Methods to isolate the radioactive ^{107}Pd isotope from palladium, e.g. using a laser technology (Shiho Asai et al. 2016) or electromagnetic separation (Gall et al. 2019), are rather expensive now. Therefore, one mayhandle a large quantity of reactorgrade palladium without extracting ^{107}Pd, as current law requires, either by its dilution with natural palladium or using it in nuclear industry (Pokhitonov and Tananaev 2022).

Calculation methodology

Using modern software tools for neutronic simulation, it is possible to study in detail the radiation characteristics of reactorgrade palladium.

The versatile and most accurate technique to calculate radiation transfer is the Monte Carlo method (Gurevich and Shkarovsky 2012). The method is used by common

[1] Federal Nuclear Regulations and Rules “Basic Rules for Accounting and Control of Radioactive Substances and Radioactive Waste within an Organization” (NP-067-16) (approved by Order No. 503, dated November 28, 2016, from the Federal Service for Environmental, Technological and Nuclear Supervision)

[2] Sanitary Rules and Standards. SanPiN 2.6.1.2523-09 “Radiation Safety Standards. NRB-99/2009” (approved by Resolution No. 47, dated July 7, 2009, from the Chief State Sanitary Inspector of the Russian Federation).

Table 3. Characteristics of radioactive isotopes in reactorgrade PGMs and their limits

Element	Isotope	Half-life	Specific activity, Bq/g	MSSA, Bq/g	MSA, Bq	Cooling time, years
Ruthenium	^{106}Ru	373.59 day	8.05E+09	1.00E+02	1.00E+05	~27
Rhodium	^{102}Rh	207.0 day	2.22E+03	1.00E+01	1.00E+06	~5
	^{102m}Rh	3.74 year	1.03E+03	1.00E+02	1.00E+06	~13
Palladium	^{107}Pd	6.5 million years	2.91E+06	1.00E+05	1.00E+08	~32 million

and generally accepted codes, such as MCNP, PHITS, PENELOPE, and others. This paper uses the licensed PHITS code (Sato et al. 2023). The PHITS (Particle and Heavy Ion Transport code System) is a general-purpose Monte Carlo particle transport simulation code developed collaboratively by the Japanese Atomic Energy Agency (JAEA), the RIST and KEK Institutes, and others. The code is capable to take into account the transport of all particles over a wide range of energies using all currently available nuclear reaction models and nuclear data libraries.

The PHITS code was used to determine the flux and equivalent dose rate (EDR) from beta particles and deceleration emission from reactorgrade palladium metal, as well as the path length of beta particles in palladium metal. The computational model has the form of a 1 mm thick palladium metal plate the composition of which is presented in Table 2.

Radiation hazard from reactorgrade palladium

The only radioactive palladium isotope, ^{107}Pd, is a soft beta emitter. The maximum energy of beta particles, E_β, is 0.034 MeV, and the average energy is 0.009 MeV. Fig. 1 presents the energy spectrum of beta particles and gamma quanta (deceleration emission) escaping the plate surface and entering a detector with an area of 1 cm^2. As a result, the flux of beta particles is 272 $\beta s^{-1} \cdot cm^{-2}$, which is less than the regulated flux of 820 $\beta s^{-1} \cdot cm^{-2}$ (for E_β = 0.05 MeV) (see table 8.4 in NRB-99/2009[2]).

The maximum calculated path length of beta particles from ^{107}Pd in palladium metal was 0.8 μm. Due to the small path length of beta particles, reactorgrade palladium emits only from the surface layer, and the remaining electrons are absorbed in the material itself.

The maximum biological tissue penetration depth of beta particles with an energy of 0.034 MeV is known to be ~20 μm (Georgia 2019), which does not exceed the thickness of the skin epidermis corneous layer (Lademann et al. 2008). Therefore, there is no probability for human biological tissues to be damaged when exposed to radiation from the outside.

Fig. 2 presents a diagram of the equivalent dose rate (EDR) from beta particles and gamma quanta of the reactorgradepalladium escaping the surface. The EDR at the surface is 0.04 μSv/h, which is below the EDR for the population (0.06 μSv/h) (see table 3.3.1 in OSPORB-99/2010[3]).

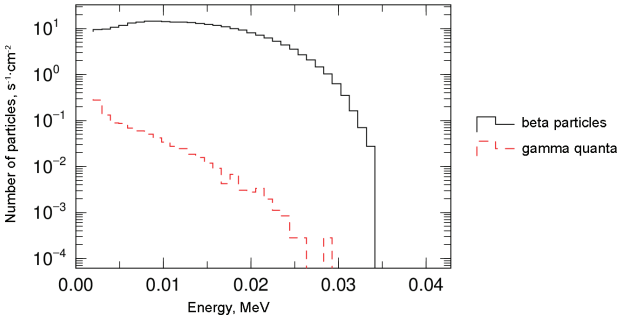

Figure 1. Energy-based distribution of beta particles and gamma quanta.

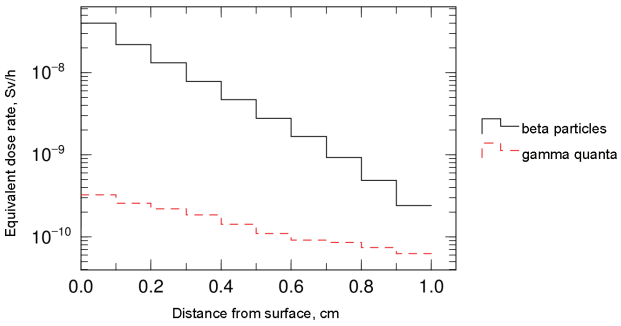

Figure 2. EDR from beta particles and gamma quanta of reactorgrade palladium.

Conclusions

Reactorgrade PGMs isolated from SNF contain radioactive isotopes, such as ^{106}Ru, ^{102}Rh, ^{102}m Rh, and ^{107}Pd. The findings show that reactorgrade PGMs cooled for ~ 27 years (ruthenium) and ~ 13 years (rhodium) can be used in unlimited quantities. Due to the ^{107}Pd isotope with a half-life of 6.5 million years, it makes no sense to cool palladium. However, the calculation results show that reactorgrade palladium is not radioactively hazardous when contacted externally. An experimental investigation into the radiation characteristics of reactorgrade palladium is described in Zaitsev et al. 1988, which is confirmed by the above conclusions.

It should be noted that the paper considers pure reactorgrade PGMs with no impurities in the form of other radioactive elements. There is still a question unanswered about the cost of isolating reactorgrade PGMs depending on their purification extent.

[3] Sanitary Rules and Standards. SP 2.6.1.2612-10 “Basic Sanitary Rules for Ensuring Radiation Safety (OSPORB-99/2010) (appr. by Resolution No. 40, dated April 26, 2010, from the Chief State Sanitary Inspector of the Russian Federation).

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