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INVESTIGATION OF NUCLEAR FUEL CHARACTERISTICS BASED ON URANIUM ISOTOPES

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Abstract
This article analyzes the application areas of natural uranium isotopes — uranium-238, uranium-235, uranium-234 — as well as thorium-232, and examines the main characteristics of nuclear fuel isotopes — plutonium-239, plutonium-240, and uranium-233 — which are produced through nuclear reactions in reactors. The reasons for the fission of uranium-238 and thorium-232 under fast neutrons, and of uranium-235, uranium-233, and plutonium-239 under thermal (slow) neutrons, are explained. Furthermore, reactions for the production of uranium dioxide (UO_2), which is used as nuclear fuel in power reactors, are proposed. The article also evaluates the advantages and disadvantages of this compound as a nuclear fuel and provides explanations for these properties.

Keywords: Actinide uranium compounds (UO_2, UO_3), Active zone, Alpha decay, Barn, Beta decay, CANDU reactor.

Introduction

In various nuclear reactors, including RBMK, VVER, and CANDU types, uranium-238 and thorium-232 nuclei are commonly employed as nuclear fuel and raw material. The primary nuclear fuel isotopes comprise uranium-238, thorium-232, uranium-235, and the rare uranium-234 isotope. Secondary nuclear isotopes include uranium-233, plutonium-239, plutonium-240, and plutonium-241. These isotopes are characterized by relatively high fission and neutron capture cross-sections. Depending on the specific isotope, fission can occur either under thermal neutron irradiation (E_1 = 0.025 eV) or under fast neutron irradiation (E_2 = 10–14 MeV) The probability of fission is governed by the neutron capture cross-section of the isotope.

In nuclear reactors, uranium dioxide and plutonium dioxide, along with their mixed oxides and uranyl nitrate compounds, are considered important nuclear fuels. The

formation reactions of these compounds were analyzed, and the mechanisms of uranium dioxide synthesis were presented. In addition, the formation reactions of uranium tetrafluoride and uranium hexafluoride, as well as their conversion into UO_2 through interactions with hydrogen and oxygen atoms, were examined. The advantages and drawbacks of utilizing the resulting UO_2 compound as nuclear fuel were also critically assessed.

Nuclear fuel refers to nuclides that undergo fission under the influence of neutrons. As a rule, uranium, plutonium, and thorium isotopes with even mass numbers (^{232}U, ^{240}Pu, ^{242}Pu, ^{232}Th) can only undergo fission when exposed to fast neutrons with energies above a threshold of approximately 1.5 MeV. In contrast, isotopes with odd mass numbers such as ^{239}Pu, ^{241}Pu, ^{235}U, and ^{233}U are capable of fission at any neutron energy, including thermal neutrons (~0.025 eV). The lower the neutron energy, the higher the microscopic cross-section for fission of odd-mass isotopes.

The neutrons produced during fission have an average energy of about 2.1 MeV, which is below the fission threshold energy of even-mass isotopes, meaning that they are predominantly fast, slowing-down neutrons. This explains why a chain reaction cannot be sustained with even-mass isotopes: only a very small fraction of neutrons reach energies above their fission thresholds. Consequently, to sustain a chain reaction with odd-mass isotopes, the fission neutrons must be moderated, reducing their energies down to the thermal range.

-Nuclear fuels consisting only of naturally fissile isotopes (^{235}U, ^{238}U, ^{232}Th) are referred to as primary nuclear fuels.

-Isotopes produced artificially (^{233}U, ^{239}Pu, ^{241}Pu) are referred to as secondary nuclear fuels.

The ^{238}U and ^{232}Th isotopes are natural nuclear materials that undergo fission only under the influence of fast neutrons. Therefore, they are not used directly as nuclear fuel but rather serve as fertile materials for the production of secondary nuclear fuels [1].

1.^{238}U -present in natural uranium at 99.25% $\left(T_{1/2} = 4{,}5 \cdot 10^9 \text{years}\right)$.

2.^{235}U - present in natural uranium at 0.71% $\left(T_{1/2} = 0{,}7 \cdot 10^9 \text{ years}\right)$.

3. ^{234}U - present in natural uranium at 0.0056% $\left(T_{1/2} = 2{,}5 \cdot 10^5 \text{ years}\right)$.

The ^{234}U isotope is produced from the ^{238}U isotope through one α-decay, followed by two successive β^- decays of the resulting ^{234}Th isotope. The decay chain can be written as follows:

$$^{238}U(\alpha) \quad ^{234}Th\left(\beta^-, T_{1/2} = 24\ day\right) \quad ^{234}Pa\left(\beta, T_{1/2} = 6{,}7 hours\right) \quad ^{234}U \qquad (1)$$

All uranium isotopes are radioactive, emitting alpha particles with energies of 4.5–4.8 MeV, and are capable of spontaneous fission accompanied by neutron emission (e.g., 1 kg of ^{238}U emits ~13 neutrons per second). Among them,^{235}U is the only naturally occurring fissile isotope, undergoing fission with neutrons of any energy, including thermal neutrons. In practice, most power reactors operate with uranium enriched to 2–5% in ^{235}U, while fast while fast reactors typically require enrichments of 15–25% [2].

It is well established that the primary isotopes used as nuclear fuel are ^{235}U, ^{239}Pu, and ^{233}U. Among these, uranium-235 is a naturally occurring isotope of uranium, although it can also be bred from ^{238}U nucleus. The natural abundance of ^{235}U is approximately 0.71%, and its half-life is $T_{1/2} = 0{,}71 \cdot 10^9$ years. As the head of the 4n+3 radioactive decay series, ^{235}U undergoes fission when irradiated with thermal neutrons, releasing about 200 MeV of energy per fission event.

Some isotopes of uranium undergo induced fission when exposed to neutrons. The isotopes ^{235}U and ^{233}U primarily undergo fission with thermal neutrons (0.025 eV); however, due to their high fission cross-sections, they can also partially undergo fission under fast neutrons. In contrast, the isotope ^{238}U, which is considered a nuclear raw material, undergoes fission only under the influence of fast neutrons (energy > 1 MeV).

To verify this, it is necessary to calculate the energy released during neutron capture by ^{235}U and ^{238}U, which corresponds to the neutron binding energy in the formation of intermediate nuclei ^{236}U and ^{239}U, respectively. For example:

For the reaction (^{235}U+n):

$E_{heavy}=(M_{235}+M_n-M_{236})c^2=40{,}913+8{,}071-42{,}440=6{,}5$ MeV

For the reaction(^{238}U+n):

$E_{heavy}=(M_{238}+M_n-M_{239})c^2=47{,}305+8{,}071-50{,}570=4{,}8$ MeV

The fission barrier for ^{235}U and ^{238}U nuclei is equal to 5.7 MeV.

Thus, in the case of ^{235}U+n, the neutron effectively raises the nucleus above the fission barrier even with “zero energy,” meaning that fission can occur without the need for additional kinetic energy. However, in the case of ^{238}U+n, the neutron must have an energy of at least 1 MeV to induce fission. At lower neutron energies, the ^{238}U nucleus cannot undergo fission—it can only capture the neutron.

From this, we can conclude that the ^{235}U nucleus can undergo fission even with neutrons that have negligible or negative kinetic energy. Secondly, the reason ^{235}U undergoes fission effectively with thermal neutrons is due to its large fission cross-

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section $\sigma_{^{235}U} = 560$, which is several hundred barns, whereas the fission cross-section of ^{238}U is only a few barns $\sigma_{^{238}U} = 1$

When the ^{238}U nucleus captures a high-energy neutron, it transforms into ^{239}U, which subsequently undergoes two β^- decays and is converted into the fissile isotope ^{239}Pu:

$$U^{238} + n\,(14\,MeV) \rightarrow U_{92}^{239} \xrightarrow{\beta^-} Np_{93}^{239} \xrightarrow{\beta^-} Pu_{94}^{239} \qquad (2)$$

The thermal neutron capture cross-sections of the uranium isotopes ^{234}U, ^{235}U, and ^{238}U are approximately $98 \cdot 10^{-28} m^2$, $683 \cdot 10^{-28} m^2$ and $2{,}7 \cdot 10^{-28} m^2$, respectively. It is evident that this value for ^{238}U is about 232 times smaller than that for ^{235}U.

The complete fission of one kilogram of ^{235}U yields approximately $2 \cdot 10^7$ kWh of thermal energy, often referred to as its "thermal energy equivalent."

Nearly all uranium isotopes are alpha emitters. In addition, the isotopes ^{233}U, ^{238}U, and ^{239}U exhibit spontaneous fission properties; however, the probability of spontaneous fission is significantly lower than that of alpha decay.

The nuclear-physical properties of the isotopes ^{235}U and ^{239}Pu, which are used as nuclear fuel, are quite similar. This similarity is reflected in the fact that both isotopes undergo fission under the influence of thermal neutrons [3]

Table 1

Isotopes	$\sigma_{thermal}$ (barns)	σ_{fast} (barns)	Number of Neutrons Produced per Fission Event
^{235}U	562,2	2,0	2,41
^{239}Pu	742,5	1,8	2,87

When the ^{235}U isotope captures a thermal neutron, it forms an excited ^{236}U compound nucleus, which undergoes fission within $10^{-14} \div 10^{-16}$ seconds, splitting into two fission fragments and releasing 2–3 free neutrons.

A 60 kg mass of ^{235}U can undergo approximately 96 fission events per second, which not only makes it a viable source of nuclear energy but also enables its use in the production of nuclear weapons.

One kilogram of ^{238}U produces approximately 35 times more neutrons than the same amount of ^{235}U, meaning that even a small quantity of this isotope can significantly increase radiation levels. In contrast, ^{234}U produces 22 times fewer neutrons than ^{235}U, making its effects similar to those of ^{238}U in terms of undesirable radiation.

The specific activity of ^{235}U is about 2.1 microcuries per gram, and its contribution to total radioactive contamination is around 0.8%. Meanwhile, ^{234}U has a much higher specific activity—51 microcuries per gram. Although the natural abundance of ^{234}U is very low (only 0.0056%, or about 140 times less than ^{235}U, it is more easily and rapidly fissioned. In highly enriched uranium (HEU), the content of ^{234}U can reach about 2%. The fission of the ^{235}U isotope is used not only for nuclear energy generation but also in the development of nuclear weapons and the synthesis of certain important actinides. The chain reaction is sustained by the excess neutrons released during the fission of ^{235}U, which are captured by ^{238}U nuclei. After two successive β^- decays, the ^{239}Pu isotope is formed, which is also capable of undergoing fission under the influence of thermal neutrons.

Uranium-233 Isotope. The ^{233}U isotope is an alpha (α) emitter with a half-life of $T_{1/2} = 1{,}5 \cdot 10^5$ years.

Its parent nuclides include ^{237}Pu (alpha emitter), ^{233}Np (beta-plus emitter, β+), and ^{233}Pa (beta-minus emitter, β–), while its daughter nuclide is ^{229}Th.

The ^{233}U isotope is produced in nuclear reactors from the thorium-232 (^{232}Th) isotope via the following reaction:

$$^{232}_{90}T_n + n(fast) \rightarrow {}^{233}_{90}T_n \xrightarrow{\beta(22{,}2\ minutes)} {}^{233}_{91}Pa \xrightarrow{\beta(27\ days)} {}^{233}_{92}U \qquad (3)$$

The ^{232}Th isotope captures a fast neutron to form ^{232}Th, which then undergoes beta decay to form ^{233}Pa. After another beta-minus (β–) decay, the isotope ^{233}U is produced. The ^{233}U isotope exhibits both spontaneous and induced fission properties and can undergo fission upon interaction with neutrons of any energy. Therefore, it is used as nuclear fuel in reactor applications.

The effective fission cross-section of ^{233}U under fast neutron irradiation is approximately 533 barns. This isotope does not occur naturally. The critical mass of ^{233}U is significantly smaller than that of ^{235}U, approximately 16 kg. The spontaneous fission rate of ^{233}U is about 720 fissions/(s·kg).

The ^{233}U nucleus primarily undergoes fission upon neutron absorption, and only rarely captures a neutron to form ^{234}U, although this process has a very low probability.

Thus, as primary nuclear fuels, enriched isotopes consisting of 5–10% ^{235}U, ^{233}U, and 20–25% enriched ^{239}Pu are utilized.

The ^{232}Th isotope captures a fast neutron and transforms into ^{232}Th, which then decays into the ^{233}Pa isotope. After another beta-minus (β–) decay, the isotope ^{233}U is formed. The ^{233}U isotope exhibits both spontaneous and induced fission properties and can

undergo fission under neutrons of any energy. Therefore, it is used as a nuclear fuel in reactor applications.

The effective fission cross-section of ^{233}U under fast neutron irradiation is approximately 533 barns. This isotope does not occur naturally. The critical mass of ^{233}U is significantly smaller than that of ^{235}U, approximately 16 kg. The spontaneous fission rate of ^{233}U is about 720 fissions per second per kilogram.

"When a ^{233}U nucleus absorbs a neutron, it undergoes fission, and in rare cases, it captures the neutron to become the ^{234}U isotope; however, the probability of such a process is very low. Therefore, the primary nuclear fuel typically consists of 5–10% ^{235}U, ^{233}U, and 20–25% enriched ^{239}Pu isotopes."

"Now let us discuss the enrichment of uranium isotopes: Enriched uranium refers to uranium that contains a higher concentration of ^{235}U than is found in natural uranium—that is, uranium in which the proportion of uranium-235 exceeds its natural abundance. The enrichment levels of uranium isotopes are classified based on the following concentration ranges [4]

- Low-enriched uranium (LEU): $C_{uranium} < 5\%$
- Medium-enriched uranium: $C_{uranium} = 5–20\%$
- High-enriched uranium (HEU): $C_{uranium} = 20–90\%$
- Very highly enriched uranium: $C_{uranium} > 90\%$

The level of enrichment used for nuclear fuel varies depending on the reactor type. For instance, in CANDU-type reactors, natural uranium is used as fuel."

A reactor with an electric capacity of 1 GW(e) has an initial core loading of 100 tons of UO_2. The annual plutonium yield from the reactor is approximately 350 kg per GW(e)·year. The isotopic composition of plutonium in CANDU reactors is similar to that in VVER reactors, consisting approximately of 66% ^{239}Pu, 27% ^{240}Pu, 5% ^{241}Pu, and 2% ^{242}Pu. This corresponds to about 71% fissile nuclides (66% ^{239}Pu + 5% ^{241}Pu), with similar ^{240}Pu content in both reactors (25% in VVER and 27% in CANDU).

In RBMK-type reactors, low-enriched uranium oxide fuel (~2% ^{235}U) is used. The initial fuel loading for a 1 GW(e) RBMK reactor is approximately 150–180 tons of UO_2. The amount of separated secondary nuclear fuel generated per year is about 250 kg of plutonium per GW(e)·year. However, the isotopic composition of plutonium extracted from spent nuclear fuel (SNF) in RBMK reactors contains lower amounts of fissile isotopes and ^{240}Pu compared to VVER reactors. Specifically, the isotopic composition in RBMK SNF is about: 45% ^{239}Pu, 36% ^{240}Pu, 11% ^{241}Pu, and 8% ^{242}Pu—resulting in approximately 56% fissile nuclides.

The most prominent type of power reactors are VVER reactors, which are light-water energy reactors that use low-enriched uranium dioxide (UO_2) fuel (3–5% ^{235}U). The

initial fuel loading for a 1 GW(e) VVER reactor is approximately 100 tons of UO_2. The amount of separated secondary nuclear fuel from such reactors is about 200 kg of plutonium per GW(e)·year.

However, the isotopic composition of reactor-grade plutonium is not suitable for nuclear weapons production. Weapons-grade plutonium consists primarily of the ^{239}Pu isotope, with no more than 7% ^{240}Pu [5]. In contrast, plutonium separated from the spent nuclear fuel of light-water power reactors contains roughly: 60% ^{239}Pu, 25% ^{240}Pu, 11% ^{241}Pu, and 4% ^{242}Pu—resulting in about 71% fissile isotopes (60% ^{239}Pu + 11% ^{241}Pu).

The critical mass of metallic reactor-grade plutonium is approximately 1.5 times greater than that of weapons-grade plutonium (23 kg vs. 15 kg, respectively). However, this is not the most significant difference. Reactor-grade plutonium contains about four times more ^{240}Pu compared to weapons-grade material, which drastically reduces the efficiency (by approximately 30 times) of a nuclear explosive device due to ^{240}Pu being a strong source of spontaneous fission neutrons."

The main isotopes and compounds used as nuclear fuel are as follows [6-8].

- **Natural isotopes** (used as raw materials): ^{238}U, ^{232}Th, and ^{235}U
- **Artificial isotopes**: ^{233}U, ^{239}Pu, ^{240}Pu, ^{241}Pu
- **UO_2** – uranium dioxide
- **PuO_2** – plutonium dioxide
- **$(PuO_2 + UO_2)$** – a mixed oxide (MOX) compound
- **UO_3** – uranium trioxide (also known as uranyl oxide)
- **PuO_3** – plutonium trioxide
- **UF_6** – uranium hexafluoride
- **UF_4** – uranium tetrafluoride
- **$(UO_2)(UO_2)\cdot 3H_2O$** – uranophane
- **U_3O_8** – triuranium octoxide

In addition to these, there are other uranium-containing compounds that are also used as nuclear fuel. Below, we will review the chemical reactions of some of these fuel compounds and carry out corresponding calculations.

1.UO_2–Uranium Dioxide:

Triuranium octoxide (U_3O_8) reacts with hydrogen gas at high temperatures to produce uranium dioxide and water vapor. This reaction is represented as:

$U_3O_8 + H_2 \rightarrow UO_2 + H_2O$

In this process, when U_3O_8 is mixed with water and heated up to a temperature of approximately 750°C, water droplets (H_2O) form on the inner walls of the reaction vessel, while a solid powder consisting of 99% pure UO_2 is formed inside the furnace. Additionally, in real-world applications (such as during hydrolysis), uranium hexafluoride (UF_6) can also be converted into uranium dioxide through a series of multi-step reduction reactions involving hydrogen. These involve multiple intermediate transformations that ultimately yield UO_2.

The sequence of transformations can be written as follows:

$UF_6 \rightarrow UO_2F_2 \rightarrow UO_2$

$UF_6 \rightarrow UF_2 \rightarrow UO_2$

$UF_6 + H_2 = UF_4 + 2HFUF_4 + 2H_2O = UO_2 + 4HF$

The overall reaction of gaseous uranium hexafluoride interacting with water vapor and hydrogen can be written as follows:

$UF_6 + 2H_2O + H_2 = UO_2 + 4HF$

Through the above reactions, solid uranium dioxide (UO_2) can be obtained. It is important to emphasize that this compound is used as nuclear fuel in power reactors. The transformation sequence (or formation process) of uranium dioxide via the interaction of uranium hexafluoride with hydrogen and oxygen can be written as follows:

Under the influence of hydrogen at high temperatures, uranium hexafluoride (UF_6) first converts into uranium tetrafluoride (UF_4). This compound then reacts with water at elevated temperatures (750–800°C) to form uranium dioxide (UO_2)."

$UF_6 + H_2 = UF_4 + 2HF$,

$UF_4 + 2H_2O = UO_2 + 4HF$,

$2H_2 + O_2 = 2H_2O$

or $UF_6 + 2H_2O = UO_2F_4 + 4HF$, $UO_2F_2 + H_2 = UO_2 + 2HF$

or $O_2F_2 + H_2O = UO_3 + 2HF$, $UO_3 + H_2 = UO_2 + H_2O$

These processes are carried out through chemical reactions.

Pure uranium tetrafluoride (UF_4) contains approximately 1% uranyl fluoride (UO_2F_2) and 1% uranium dioxide (UO_2), as well as some triuranium octoxide (U_3O_8). Uranium dioxide (UO_2) fuel is primarily manufactured in the form of solid pellets, which are used as fuel in VVER-1000 reactors. Thus, through the chemical reactions described above, the essential nuclear fuel UO_2 for reactors is produced.

2.UF_4 – Uranium tetrafluoride

UF_6 – Uranium hexafluoride reacts with hydrogen molecules to form uranium tetrafluoride (UF_4). The chemical reaction can be written as follows:

$$UF_6 + H_2 \rightarrow UF_4 + 2HF + 16{,}5\frac{Kj}{mol}. \qquad (4)$$

This reaction is exothermic, releasing 16.5 joules of energy.

Uranium hexafluoride reacts with hydrogen to form uranium tetrafluoride and hydrogen fluoride. Subsequently, uranium tetrafluoride reacts with calcium in the form of an alloy to produce metallic uranium.

Reaction (4) proceeds with a relatively low energy release; however, the activation energy for the reduction of UF_6 by hydrogen is quite high. The reduction of uranium hexafluoride with hydrogen is considered a first-order reaction.

Currently, fast breeder reactors predominantly use uranium fuel rather than uranium-thorium compounds. This fuel consists of uranium dioxide (UO_2) enriched to 20–25%. The load for a reactor with a capacity of 1 GW(e) is approximately 10–15 tons, with 2–3 tons of fissile isotopes maintained continuously in the reactor core.

The annual reprocessing of plutonium from the spent fuel is about 1500 kg/(GW(e)·year). During the fuel cycle, approximately 80% of the accumulated plutonium is reprocessed in a “pure” form, amounting to 250 kg/(GW(e)·year).

Fast reactors achieve nearly complete fuel burnup at the level of 80–100 GW·day/ton, which is approximately three times higher than that of light-water thermal reactors. Consequently, the plutonium accumulated in the active zone of fast reactors contains a large fraction of higher isotopes, making this plutonium unsuitable for nuclear weapons.

The main advantages of uranium dioxide (UO_2) as nuclear fuel are as follows:

1. Extremely high melting temperature (2780 °C);
2. Chemical stability of coolants used in nuclear reactors (such as light water, heavy water, sodium, etc.);
3. Construction materials made from corrosion-resistant steel and zirconium alloys;
4. Radiation resistance under high neutron flux (10^{14} neutrons/cm²·s);
5. Established technology for producing fuel in the form of high-density pellets;
6. Isotropy of the crystal lattice structure.

Disadvantages of Uranium Dioxide (UO_2):

1. With decreasing temperature, thermal conductivity sharply decreases: at 45 °C, the thermal conductivity (λ) drops from 0.084 W/cm·K to 0.024 W/cm·K.

Due to the small size of the uranium dioxide pellet (radius R = 3 mm), it is highly sensitive to temperature changes. The temperature gradient at the pellet's surface reaches approximately ΔT ~ 1000–1500 °C, while at the pellet center it can reach ΔT ~ 1500–2000 °C. This results in a temperature gradient between the center and edge of the pellet, which hinders the complete combustion (burnup) of uranium dioxide in the reactor core.

2. At room temperature, UO_2 undergoes light oxidation when exposed to high humidity. Therefore, it must always be operated under dry air conditions. Fuel based on (UO_2 + PuO_2) compounds is produced in the form of small pellets using a specialized scheme and is exclusively used in VVER reactors. Its efficiency in other reactor types is relatively low.

Nuclear Fuels Loaded and Produced in Nuclear Reactions [9] (Table 2)

№	Reactor Type	Loaded Fuel	Produced Fuel, kg/(GW·year)
1	Research reactors	510kg (90 % , ^{235}U)	-
2	VVER	100 t UO_2 (3-5 % , ^{235}U)	200, (25% , ^{240}Pu)
3	RBMK	150-180 t UO_2 (1,8-2 %, ^{235}U)	250, (36% , ^{240}Pu)
4	CanDU	100 t UO_2 (0.7 % , ^{235}U)	350, (27% , ^{240}Pu)
5	LMFBR	10-15 t UO_2 (15-20 % , ^{235}U)	

Thus, in this study, the nuclear-physical properties of the isotopes ^{235}U, ^{233}U, and ^{239}Pu, which are used as nuclear fuel in reactors, were analyzed. A series of chemical reactions involved in producing uranium dioxide (UO_2) nuclear fuel was presented. Additionally, the advantages and disadvantages of uranium dioxide compounds as nuclear fuel were examined, and conclusions were drawn.

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