Physics – Uspekhi **65** (11) 1188 – 1192 (2022)

CONFERENCES AND SYMPOSIA

PACS numbers: 28.41.Kw, 28.41.Vx, 92.70.Mn

FORUM 'USPEKHI-2021': CLIMATE CHANGE AND GLOBAL ENERGY ISSUES
(Joint Scientific session of the Physical Sciences Division, Chemistry and Materials Sciences Division, Division of Earth Sciences and Division of Energetics, Machinery, Mechanical Engineering and Control Processes of the Russian Academy of Sciences, January 19, 2021)

Solving scientific problems of nuclear power engineering as a source of 'green' energy

S N Kalmykov

DOI: https://doi.org/10.3367/UFNe.2022.10.039256

Contents

Abstract. A way to prevent adverse global climatic change is to essentially alter the strategy of power generation, which is one of the major sources of greenhouse gases. A task of paramount economic importance is to ensure a balance among various energy sources, depending on the social economical features of a particular region. Major industrial regions cannot develop without highly concentrated energy sources, among which nuclear energy is actually the only 'green' one. However, progress in the nuclear power generation industry and an increase in its share in the total power generation both in Russia and worldwide depend on solving challenging scientific and technological problems related to safe processing of spent nuclear fuel and radioactive wastes. An urgent task is to develop a next-generation nuclear power industry based on a combination of thermal- and fast-neutron reactors, recycling of fissile nuclides, deep fractioning of radioactive wastes with subsequent 'afterburning' of long-lived radionuclides, and minimizing deep disposal in geological formations.

Keywords: nuclear power generation industry, recycling of fissile nuclides, balanced nuclear fuel cycle, fractioning of radioactive wastes

S N Kalmykov
Presidium of the Russian Academy of Sciences,
Leninskii prosp. 14, 119991 Moscow, Russian Federation;
Lomonosov Moscow State University,
Leninskie gory 1, 119991 Moscow, Russian Federation
E-mail: stepan_5@hotmail.com

Received 4 October 2022
Uspekhi Fizicheskikh Nauk **192** (11) 1275 – 1279 (2022)
Translated by M Zh Shmatikov

1. Introduction

A task of utmost importance in the development of science and technology is to create new, economical, and environmentally friendly energy sources, which, in combination with conventional sources, will ensure the sustainable development of individual regions and countries. The global problems of humankind, the ever increasing demand for material goods, and the limited resources, including energy resources, have been discussed previously in *Physics–Uspekhi* [1–3]. Conventional energy sources based on the combustion of fossil fuels, primarily coal, which accounts for about 38% [4] of the world's total energy production, fail to meet increasingly stringent requirements for protection against environmental pollution and greenhouse gas emissions (primarily CO_2 when burned). However, economic development is not possible without using highly concentrated energy sources. Nuclear energy is such a highly concentrated source of energy production that neither results in significant greenhouse gas emissions nor affects the climate. The Organization for Economic Cooperation and Development (OECD) [5] has declared a desired indicator of 50 g CO_2 eq. emissions per generated kWh of electricity. Figure 1 shows the values of CO_2 emission in g (eq.) per kWh from the main sources of electric power (taking into account the full life cycle of objects). It is of importance that the combustion of fossil fuels leads, in addition to CO_2 emission, to the emission of other hazardous substances, including sulfur dioxide, nitrogen oxides, and carbon black particles [6].

The most important misconception is that nuclear power plants negatively impact the dose loads of the population residing near them. A number of studies [7, 8] have shown that, under regular operation, the dose exposure of the population residing near coal-fired power plants is significantly higher than that of the population residing near

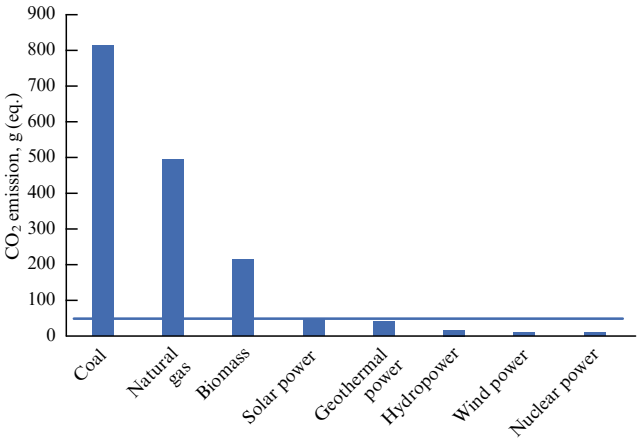

Figure 1. Emission of CO_2 in g (eq.) in production of 1 kW of electric power from various energy sources.

nuclear power plants. For example, the equivalent dose from external exposure, the effective dose to the skin and internal organs from gaseous emissions from a nuclear power plant were 2.93×10^{-4} mSv yr^{-1}, 2.90×10^{-3} mSv yr^{-1}, and 1.78×10^{-2} mSv yr^{-1}, respectively, compared with the values of 1.13×10^{-2} mSv yr^{-1}, 5.33×10^{-2} mSv yr^{-1}, and 1.17×10^{-1} mSv yr^{-1}, respectively, for emissions from a coal-fired power plant. These significant differences are due to the emission of natural radionuclides into the air, including short-lived decay products of radium-226, in the combusting fossil fuels, which does not occur in the operation of nuclear power plants. It was shown that natural radionuclides, such as uranium, can be encapsulated in carbon fullerene-like nanostructures in the form of oxide nanoparticles with the composition of UO_{2+x} upon incomplete combustion of coal [9] (Fig. 2).

2. Modern nuclear power and the problem of radioactive wastes

Nuclear energy apparently can in the future make up a significant share of the global energy balance, but this can occur only provided the scientific and technological problems of the full closing of the nuclear fuel cycle (NFC), minimization of the resulting radioactive wastes (RWs), and multiple recycling of fissile isotopes are solved. This will make it possible to achieve both ecological and economic goals: abandoning or minimizing the deep disposal of long-lived radionuclides and switching to their 'afterburning' (transmutation) with subsequent near-surface disposal of only short-lived radionuclides.

The industrial use of nuclear energy began with the start-up in 1954 in the USSR of the first nuclear power plant with a

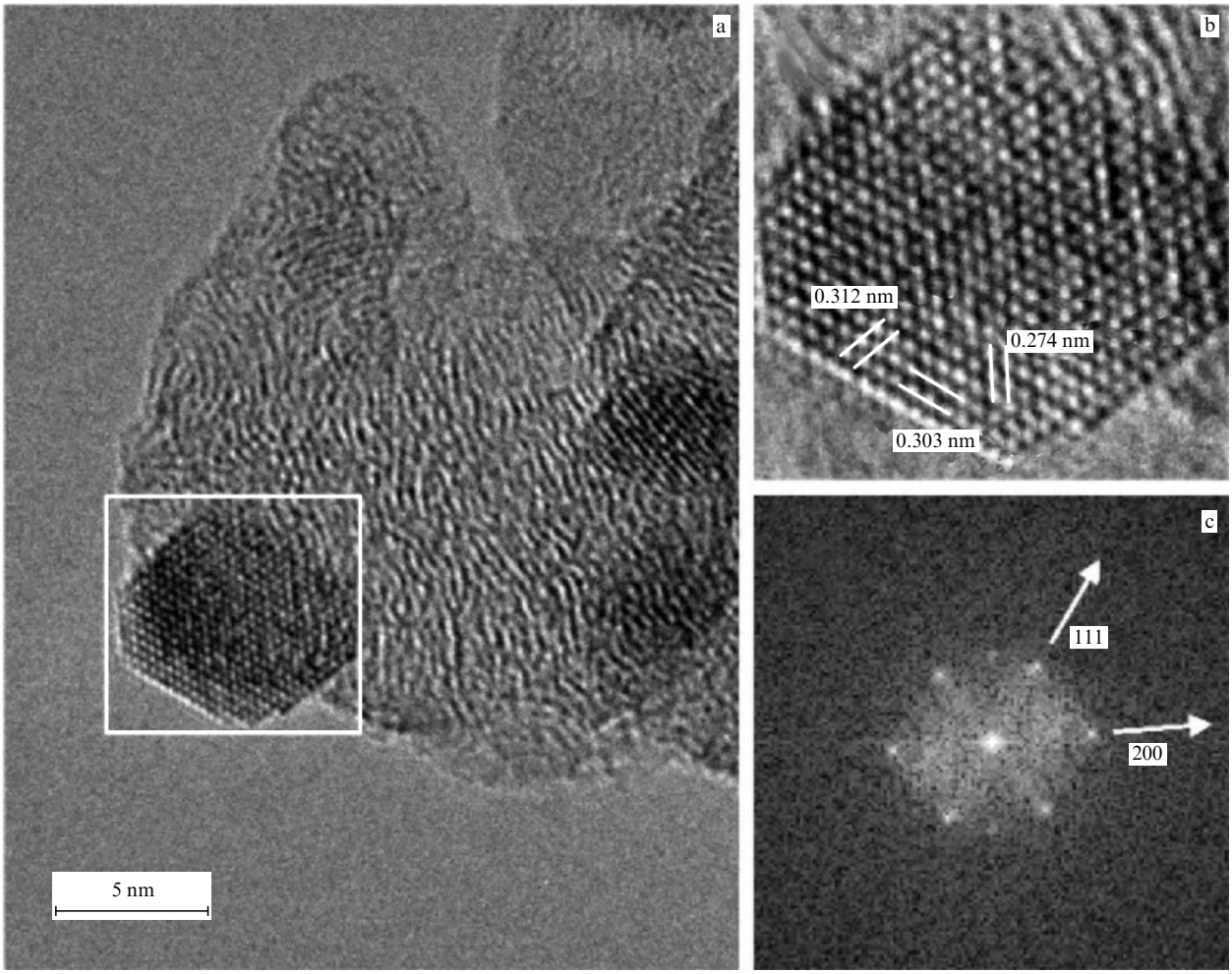

Figure 2. UO_{2+x} nanoparticles encapsulated into carbon onion-like structures discovered in smokestack exhaust gases of a coal-fueled power plant.

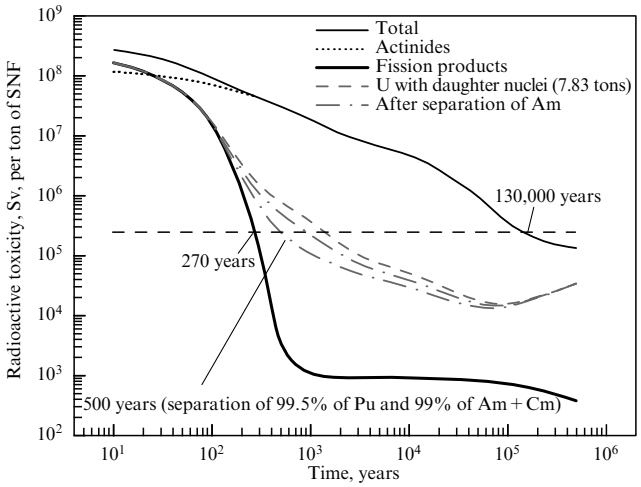

Figure 3. Changes in radioactive toxicity of SNF, actinide fraction of SNF, fission product fraction, and SNF after separation of 99.5% of plutonium, americium, and curium.

capacity of 5 MW(e). International Atomic Energy Agency (IAEA) reports that, by the end of 2021, 437 nuclear power units were in operation worldwide, which generated about 10% of the world's electricity, corresponding to 28% of the contribution to all low-carbon energy sources. According to forecasts of the same agency, by 2030, the nuclear energy sector will grow from 17% (under the pessimistic scenario) to 94% (under the optimistic scenario) [10].

There are two fundamentally different strategies for handling spent nuclear fuel (SNF): open and closed nuclear fuel cycles (NFCs). According to the former (accepted in the USA, Sweden, Finland, Switzerland, and other countries), SNF is not processed, but stored with the option of processing or final geological disposal in the future (postponed decision). More than 300,000 tons of spent nuclear fuel have already been accumulated in the world, and although at present there is not a single licensed SNF deep geological disposal facility on the planet, it is stored until a final decision is made on its fate, while the volume of SNF increases annually by about 8,000–10,000 tons [11].

In a number of countries with developed nuclear power (France, Great Britain, Russia, and others), SNF (in whole or in part) is processed with the extraction of uranium and plutonium in the Purex process and subsequent solidification of liquid RW. Deep geological disposal of solidified RW is carried out at the final stage of the closed nuclear fuel cycle. Highly radioactive wastes (HRWs) in SNF processing, as a rule, is solutions and slurries containing fission products — isotopes of Cs, Sr, Zr, Tc, Mo, Ru, I, rare earth elements, and corrosion products (isotopes of Cr, Mn, Fe, Co, Ni, Zr), minor actinides (Np, Am, Cm), and residual amounts of U and Pu.

Similar to SNF, there is not a single licensed HRW deep storage facility in the world. The reason is that the safety of such repositories should be justified for a period of up to one million years. Calculated data on the reduction of the potential long-term environmental impact during disposal of, on the one hand, SNF and, on the other hand, RW formed during a closed nuclear fuel cycle with the release of fissile actinides (^{239}Pu and ^{235}U) for reuse in nuclear power generation and minor actinides (^{237}Np, ^{241}Am, ^{244}Cm) for the purpose of their transmutation in fast neutron reactors are shown in Fig. 3 [12]. For example, the radiotoxicity of RW after isolation of long-lived actinides from SNF decreases with time by many orders of magnitude compared to that of the initial SNF. Thus, the closure of the NFC with multi-recycling of fissile components and fractionation of radioactive waste to isolate the most environmentally hazardous long-lived isotopes (such as americium) for their subsequent transmutation and a significant reduction in the RW volume is a strategic scientific and technological task for Russia's nuclear industry.

3. Strategy for closing the nuclear fuel cycle

Closing the NFC in a two-component nuclear power system with thermal and fast-neutron reactors involves recycling fissile materials; minimization or complete abandonment of the deep disposal of HRW by separating the actinide-lanthanide fraction followed by separation of americium and curium for americium transmutation and storage of curium until its decay; RW fractionation to isolate the short-lived fraction (Cs, Sr), platinum group metals, and technetium and other elements; and minimization of RW volumes. Thus, the implementation of this concept [13–16] will make it possible to:

- solve pending problems of handling SNF and RW, consistently reduce the amount of accumulated SNF, and prevent its further accumulation;
- involve plutonium in NFC as a product of processing SNF in thermal neutron reactors (VVER) for the production of fuel for fast neutron reactors and the full use of the energy potential of natural uranium (^{238}U) through multiple recycling of fuel materials;
- fractionate radioactive waste and extract long-lived minor actinides for afterburning or transmutation in nuclear reactors, which, along with multiple plutonium recycling, will significantly reduce the environmental hazard of RW;
- technologically support non-proliferation of fissile materials through stage-by-stage phasing out of the circulation of enriched uranium and the separation of pure plutonium.

One of the essential incentives for the transition to a closed NFC using 'fast' reactors is the successful operation of BN-600, an industrial 'fast' reactor at the Beloyarsk NPP, which has no foreign equivalent. In 2016, the 4th power unit with a BN-800 was commissioned, initially focused on testing NFC closure technologies [17, 18].

While extraction systems for the isolation and separation of such elements as uranium, plutonium, and many others that change their oxidation state have been developed and, in general, successfully implemented, the task of separating trivalent rare earth elements (REEs) and minor actinides (Am and Cm) with close chemical properties remains very urgent.

4. Methods for separating Am(III) and Cm(III)

The mass content of minor actinides does not exceed 0.1% of HRW, but it is these elements (primarily americium) which are the most hazardous during further geological disposal, since they feature significant half-lives and high heat release ($T_{1/2}{}^{241}\mathrm{Am} = 432$ years, $T_{1/2}{}^{243}\mathrm{Am} = 7370$ years). Curium (primarily ^{244}Cm with a half-life of 18 years) spontaneously decays with a probability of 1.4×10^{-6}% and can emit 1.08×10^{7} neutrons $(\mathrm{s\ g})^{-1}$, a value which is

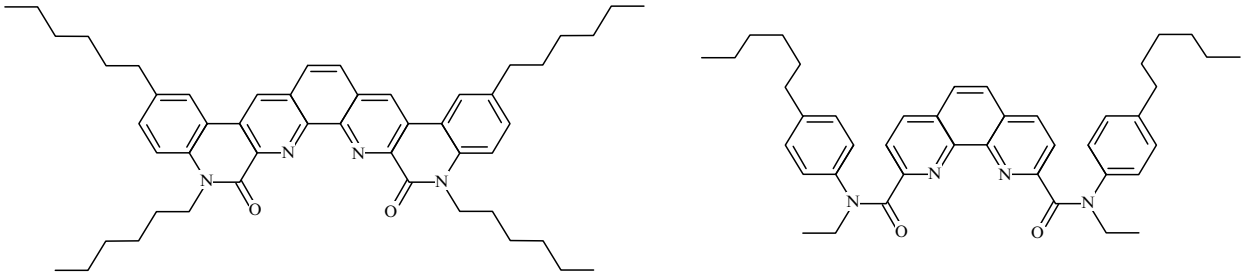

Figure 4. Base structures of phenanthrolinedicarbon acids studied for separation of REEs, minor actinides, and americium and curium.

almost 9 orders of magnitude greater than that for ^{239}Pu (2.18×10^{-2} neutrons $(\text{s g})^{-1}$) [19]. Among the main neutron poisons, in addition to ^{135}Xe, REE isotopes (including stable ones) are of the greatest importance, primarily Sm, Gd, Pm, and Eu. Radioactive ^{152}Eu ($T_{1/2} = 13.5$ years) is formed in fission with a high yield of about 5%. Thus, in the next-generation NFC, long-lived isotopes of americium should be 'afterburned' in fast neutron reactors, ^{152}Eu disposed in geological formations, and ^{244}Cm either stored for the accumulation of ^{240}Pu or also disposed. For such 'afterburning,' options are considered that involve both a heterogeneous solid-fuel reactor and a homogeneous molten-salt reactor. Ya B Zeldovich and Yu B Khariton were the first to propose, in 1939, a homogeneous reactor [20]. Consequently, the efficient separation of Eu, Am, and Cm is the most challenging scientific and engineering problem.

Various highly selective extraction systems have been studied recently for the separation of both rare earth elements and minor actinides and americium and curium. Supercomputer modeling and subsequent studies enabled identification of extractants (phenanthralenedicarboxylic acid diamides (Fig. 4)), which, according to a theoretical prediction, should exhibit high selectivity in the extractive separation of trivalent f-elements.

A vast series of compounds of these types has been developed and synthesized, and the selectivity factors for the separation of the standard americium/europium pair SFAm/Eu $\geqslant 150$ and separation of the americium/curium pair SFAm/Cm $\geqslant 7$ have been obtained, which are record high values achieved in the world to date. A new approach to the creation of extraction systems for the separation of actinides and lanthanides has been developed and successfully tested; based on the use of hydrophilic and hydrophobic ligands in aqueous and organic phases, respectively, (strategy of a hydrophilic-hydrophobic pair), it enables a significant enhancement of the selectivity of separation of target metals.

It is of importance that the proposed ligands have high radiation and hydrolytic stability, maintain their extraction properties under α-irradiation up to doses of 500 kGy and under α-irradiation up to doses of the order of 200–300 kGy, while the extraction process be fast and completely reversible. In addition, the developed extraction system must be fire and explosion safe.

In testing in centrifugal extractors (V G Khlopin Radium Institute), it was possible to isolate from nitric acid solutions containing a mixture of americium and curium (total concentration of 0.11 g l^{-1}) at least 99% of the americium into a separate portion with a purity of at least 99.9% (by curium). Moreover, the initial solution contained interfering components such as ammonium nitrate (570 g l^{-1}), diethylenetriaminepentaacetic acid (10 g l^{-1}), aminoacetic acid (5 g l^{-1}), and a plutonium impurity. These admixtures did not hinder the separation of the target components.

5. Conclusion

Nuclear power is the only highly concentrated 'green' source of electric power, the share of which in world production will most probably increase. According to the decarbonization scenario (production of no more than 50 g of CO_2 per kW of generated electricity), by 2040 the share of nuclear power will increase by 55%, to 4320 TWh of generated electric power. However, the implementation of a next-generation nuclear fuel cycle should include solving the problems of accumulating radioactive waste and minimizing the amount while maximizing the potential of fissile materials through multirecycling. This strategy is based on two-component nuclear power technology: thermal reactors with a low breeding factor generate cheap electric power, while fast breeder reactors with a breeding factor greater than 1 provide the fuel base for the entire system and utilize the SNF of thermal reactors.

References

1. Kapitza P L "Global problems and energy" *Sov. Phys. Usp.* **20** 547 (1977); "Global'nye problemy i energiya" *Usp. Fiz. Nauk* **122** 327 (1977)
2. Zavoiskii E K, Kadomtsev B B, Okun' L B, Smirnov B M "Man and environment — problems of the future" *Sov. Phys. Usp.* **20** 553 (1977); "Chelovek i okruzhayushchaya sreda — problemy budushchego" *Usp. Fiz. Nauk* **122** 337 (1977)
3. Fortov V E, Makarov A A "Avenues for the innovative development of energetics in the world and in Russia" *Phys. Usp.* **52** 1249 (2009); "Napravleniya innovatsionnogo razvitiya energetiki mira i Rossii" *Usp. Fiz. Nauk* **179** 1337 (2009)
4. Coal and Electricity, World Coal Association, London, UK, 2015, https://www.worldcoal.org/coal/uses-coal/coal-electricity
5. "The costs of decarbonisation: system costs with high shares of nuclear and renewables", NEA No. 7299 (Paris: Nuclear Energy Agency, Organisation for Economic Co-operation and Development, 2019)
6. Du X, Jin X, Zucker N, Kennedy R, Urpelainen J "Transboundary air pollution from coal-fired power generation" *J. Environmental Management* **270** 110862 (2020)
7. McBride J P, Moore R E, Witherspoon J P, Blanco R E "Radiological Impact of Airborne Effluents of Coal-Fired and Nuclear Power Plants", ORNL-5315 (Oak Ridge, TN: Oak Ridge National Laboratory, 1977)
8. Purevsuren B, Kim J *Sci. Technol. Nucl. Install.* **2021** 6685385 (2021)
9. Utsunomiya S, Jensen K A, Keeler G J, Ewing R C *Environ. Sci. Technol.* **36** 4943 (2002)
10. Myasoedov B F, Kalmykov S N "Nuclear power industry and the environment" *Mendeleev Commun.* **25** (5) 319 (2015)

11. Ceyhan M, Lee J S, in *Proc. of the Intern. Conf. on Management of Spent Fuel from Nuclear Power Reactors, IAEA, Vienna, Austria, 19–22 June, 2006* (IAEA-CN-144) (Vienna: International Atomic Energy Agency, OECD Nuclear Energy Agency, 2006)
12. "Implications of Partitioning and Transmutation in Radioactive Waste Management", Technical Reports Series No. 435 (Vienna: International Atomic Energy Agency, 2004)
13. Khapersaya A V, Kryukov O V, Ivanov K V "Spent nuclear fuel management in Russia: Status and future development", in *Management of Spent Fuel from Nuclear Power Reactors*: *Learning from the Past, Enabling the Future, Proc. of the Intern. Conf., Vienna, Austria, 24–28 June 2019* (IAEA-CN-272/53) (Vienna: International Atomic Energy Agency, 2020) p. 93–98, Paper ID #25
14. Fedorov Yu S, Baryshnikov M V, Bibichev B A et al. "Multiple recycle of REMIX-fuel based on reprocessed uranium and plutonium mixture in thermal reactors", in *Proc. of GLOBAL 2013*: *Intern. Nuclear Fuel Cycle Conf., Nuclear Energy at a Crossroads, Salt Lake City, UT, September 29 – October 3, 2013*
15. Sheremetyev A, Koulupaev D "Experience and prospects of spent nuclear fuel reprocessing at Mayak. Management of spent fuel. From nuclear power reactors. Learning from the past, enabling the future", in *Management of Spent Fuel from Nuclear Power Reactors*: *Learning from the Past, Enabling the Future, Proc. of the Intern. Conf., Vienna, Austria, 24–28 June 2019* (IAEA-CN-272/53) (Vienna: International Atomic Energy Agency, 2020) p. 161–166, Paper ID #53
16. Ponomarev-Stepnoi N N "Two-Component Nuclear Power System with a Closed Nuclear Fuel Cycle Based on BN and VVER Reactors" *Atom. Energy* **120** 233 (2016); "Dvukhkomponentnaya yadernaya energeticheskaya sistema s zamknutym yadernym toplivnym tsiklom na osnove BN i VVER" *Atom. Energ.* **120** (4) 183 (2016)
17. Adamov E O, Dzhalavyan A V, Lopatkin A V et al. "Conceptual framework of a strategy for the development of nuclear power in Russia to 2100" *Atom. Energy* **112** 391 (2012); "Kontseptual'nye polozheniya strategii razvitiya yadernoi energetiki Rossii v perspektive do 2100 g." *Atom. Energ.* **112** (6) 319 (2012)
18. Adamov E O et al. *Belaya Kniga Yadernoi Energetiki. Zamknutyi YaTTs s Bystrymi Reaktorami* ('White Book' of Nuclear Power. Closed NFC with Fast-Neutron Reactors) (Gen. Ed. E O Adamov) (Moscow: NIKIET, 2020)
19. Stewart J E, Eccleston G W, Ensslin N, Cremers T L, Foster L A, Menlove H O, Rinard P M "Neutron-Based Measurements for Nondestructive Assay of Minor Actinides Produced in Nuclear Power Reactors", LA-UR-96-3690 (Los Alamos, NM: Los Alamos National Lab., 1996)
20. Ponomarev L I "Ya B Zeldovich and nuclear power" *Phys. Usp.* **57** 215 (2014); "Ya B Zeldovich i yadernaya energetika" *Usp. Fiz. Nauk* **184** 227 (2014)