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### Nanostructured Materials and Shaped Solids for Essential Improvement of Energetic Effectiveness and Safety of Nuclear Reactors and Radioactive Wastes

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#### 1. Introduction

It is generally considered that nuclear energetics started with the discovery of nuclear fission in 1939 [1,2]. But much earlier, in 1913, H.Moseley, a prominent pupil of E. Rutherford [3], analyzed the possibilities of direct conversion of ionizing radiation to electricity and was the first to experimentally demonstrate that ionizing radiation did produce electricity [4,5]. Henry Moseley is known among most physicists as the author of "Moseley's law", connecting the energy characteristics of X-Ray emission of chemical elements with their atomic numbers [1]. On the other hand, his experiments in 1913 on creating an electrostatic potential between two insulated electrodes subjected to ionizing irradiation can be considered as the starting point for nuclear power engineering. At least his electric circuits are widely applied in modern nuclear reactors for detection of ionizing radiation by means of direct charging devices and separation of electric charges by ionizing irradiation for direct conversion of radiation to electricity is the subject of wide speculation [4–7]. These considerations seem to be quite natural because ionization means separation of positive and negative electric charges which is the basis of any kind of generation of electrical energy. In the middle of the 20th century when Nuclear Power Plants (NPP) started their scheme of electric energy production using a fairly long chain of "nuclear fission – heating energy – water vapor – vapor electric generator" transformations, discussions about direct "radiation - electricity" conversion continued [6,7]. Nowadays, after two global disasters at NPP (1986 - Chernobyl, 2011 - Fukushima), the issue of direct generation of electricity from nuclear and radiation processes has become extremely urgent because



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this way of energy production would convert nuclear power engineering from a hazardous phenomenon to the most economic and ecologically safe technology.

The abovementioned disasters at nuclear power plants with significant radioactive contamination of the environment and a large amount of human victims induced a severely negative attitude of the officials and human society to nuclear power engineering in most civilized countries. Besides, the society is highly anxious about the ecological danger of radioactive wastes because their amount will grow inevitably with increased production of nuclear energy. These two negative factors connected with nuclear power plants induce serious obstacles for further development of nuclear power engineering. Moreover, several governments have decided to gradually close their NPP (e.g. Germany). But objective comparative analysis of the ecological dangers of nuclear-based and carbon-based energy production shows that the latter is much more harmful to human life due to continuous accumulation of carbon dioxide in the Earth atmosphere and the related green house effect. Its negative influence on our climate is growing inevitably resulting in weather instabilities with extreme hazardous phenomena. Therefore, an intensive search for ways to improve the safety of Nuclear Power Plants in order to make them more acceptable to our society is a burning problem. In addition to ecological problems, the competition between nuclear, carbon, solar, hydro and other techniques of electrical energy production involves their economical effectiveness. Our studies of the prospects of application of nanostructures and shaped solids demonstrate that these two kinds of materials are capable to improve essentially either the ecological safety or the effectiveness of NPP as well as radioactive waste storage. Such improvements are based on development of units and devices for: 1) operative differential monitoring of radiation flows inside active reactors zones for instantaneous detection of damages of uranium oxide rod cladding inside nuclear reactors and optimization of regulation of reactor functioning; 2) direct conversion of radiation flows to electrical energy inside the reactors, in water pools with depleted nuclear fuel rods pulled out from the reactors and in radioactive waste stores anywhere.

The next part of this chapter is devoted to analysis of the current situation with active zone control, energy production effectiveness, resource electricity supplies, utilization of radioactive wastes at nuclear power plants around the world. The drawbacks of these systems as well as possible ways of elimination of these drawbacks are discussed. Two subsequent parts describe the properties of nanostructures and shaped sapphire which can be useful for nuclear power engineering. Manifold enhancement of radiation hardness in nanostructures and possibilities of direct transformation of radiation to electricity connected with active migration of electron excitations between nano-grains are described. A set of experimental facts confirming good radiation hardness of shaped sapphire is presented. The subsequent part deals with application of nanostructures and shaped solids to continuous and informative monitoring of radiation flows, nuclear fuel elements and other constituents inside active reactor zones. The prospects of improving the safety and economic characteristics of NPP connected with these innovations are described. The sixth part is devoted to practical ways of direct conversion of radiation to electricity at NPP, radioactive waste storage and other objects of nuclear engineering based on nanostructures and shaped solids. The influence of application of direct radiation-to-electricity conversion on safety and effectiveness of NPP is discussed.

The conclusion of this chapter summarizes the improvements in safety and effectiveness of nuclear power engineering resulting from application of nanostructures and shaped crystals. The terms of their possible realization are estimated.

## 2. Current situation with control of active zones, effectiveness and safety of nuclear energy production

In spite of the wide variety of designs of nuclear reactors, their operation is described with a set of typical processes. Uranium or plutonium nuclear fission produces two secondary nuclear splinters with the total kinetic energy of about 80 % of the total energy of the process (Fig. 1) [1, 2]. The rest of the fission energy is distributed between the secondary neutrons, alpha and beta particles and gamma radiation. The fissile nuclear fuel in the form of enriched uranium oxide pellets is contained inside the cylindrical fuel elements with metal cladding made of either zirconium alloy or stainless steel. Usually, when the cladding is not damaged, the neutron and gamma particles escape from the fuel element to the outer space whereas other fission products remain inside the rod. The major part of the energy is converted to heating directly inside a rod. Water or some other cooler is circulating between the assemblies of fuel rods, absorbs their thermal energy and delivers it from the active zone of the reactor to the outer space where heating is converted to electricity.. The gamma and neutron radiation emitted from the fuel rods is absorbed either inside the active zone or in the reactor shielding. Monitoring of the reactor operation inside the active zone is usually performed with radiation detectors registering neutron and gamma flows and with thermoelectric devices measuring the distribution of the temperature inside the reactor [1–7]. The rate of the nuclear fission is regulated with absorbers of neutron flows (cadmium, boron, etc.) by means of variation of their content inside the active zone (in the most cases by means of introduction of rods with absorbing material into the active zone or dissolving of the absorbing elements in the water used for cooling).

Typical nuclear reactors contain thousands of fuel rods with fissile material (enriched with U<sup>235</sup> or Pu<sup>239</sup>), collected to assemblies containing several tens of rods. The temperature of the cooling liquid when it leaves the active zone and its flow intensity are the main factors determining the electric power that can be achieved by the electric converters installed outside. The liquid is heated as it passes by the fuel rods where the nuclear fission takes place. The specific fission power is determined by the total amount of the fissile nuclei and the flow of fast and thermal neutrons irradiating the rod. On the other hand, these flows are determined by the intensities of the fission in other rods, the processes of moderation and absorption of neutrons, etc. Thus, the set of the fuel rods is a multi-component system with a huge amount of links and feedbacks that are flexible, so it is natural to expect local fluctuating instabilities of the fission rate and corresponding oscillations of temperatures within separate rods and their assemblies. For example, one can assume that thermal growth of a certain fuel rod will



Figure 1. Scheme of <sup>235</sup>U nuclear fission induced by a thermal neutron.

produce a vapor bubble in its vicinity, resulting in local focusing or defocusing of the neutron flows related to their additional scattering produced by water nonhomogeneities and further increase or decrease of the fission rate and the corresponding variations of the local temperature and radiation instabilities. Severe fluctuations of the fission rate and local temperatures can develop in fractions of seconds. For instance, the fission rate and corresponding power of the Chernobyl reactor before its disaster increased many fold in a second due to the drawbacks of the regulation system, boiling of water and the human errors. The fission rate is determined mainly by absorption of thermal neutrons. But the presence of the Pu<sup>239</sup> isotope (about 1%) in the depleted fuel rods after their removal from the active zones reveals the participation of fast neutrons also in the nuclear fission processes. The exchange with the fast neutrons occurring between the neighboring rods provides a positive feedback as an additional factor for local fast oscillations of the reactor operation. These temperature oscillations create oscillating internal stresses in the claddings of the rods which are capable of inducing their cracking with subsequent penetration of the radioactive splinters from the rod interior to the cooling liquid and then to the outside of the reactor core.

Thus, observations of the amplitudes and characteristic times of fast instabilities of fissile rates, radiation flows and temperatures in separate rods seem to be important for better understanding of the internal processes in reactors and further improvement of their operation control, regulation and energy effectiveness. From this viewpoint, monitoring of the averaged values of these parameters commonly done by means of measurements with thermoelectric and direct charging devices seems to be too slow (the characteristic times of these devices are of the order of several tens of seconds [1,2]). The averaging of the temperature and radiation flows proceeds inevitably due to the very long characteristic times of these devices. So, we must confess that up to now there is no available information on faster instabilities. We suppose that this is due to the absence of fast radiation detectors and temperature sensors with satisfactory radiation hardness for stable functioning within intense gamma and neutron flows inside active zones of nuclear reactors.

Our studies on nanocrystalline scintillators show that their radiation hardness can be improved up to the level required for measurements inside active zones of nuclear reactors (see part 3 of this chapter). In Part 4 we describe radiation hard light guides made of (based on) shaped sapphire and glass fibers with a complex cross-section in the form of photonic crystals. Part 5 describes the experimental schemes of local measurements of radiation flows and temperatures by means of nano-scintillators and radiation hard light guides with a temporal resolution better than one millisecond. It should be emphasized that radiation flows can be measured differentially, i.e. separate sets of data on gamma-, alpha-, beta-particle flows, fast and thermal neutrons can be registered. The intensities of scintillation flashes can be used for estimation of fast temperature variations of the rod claddings. On the other hand, the data on the oscillations of the cladding temperatures enable determination of the amplitudes of the thermo-elastic stresses and strains of the rods. By these means much more adequate information on the working resources of the claddings can be obtained.

It is worth noting that differential measurements of intensities of beta- and alpha- flows provide reliable information on local cladding damages, because these particles are usually localized inside the rods and their presence in the external region points to perforation of the cladding. Hence, the damage of the cladding can be revealed much earlier than the moment when the radioactive splinters washed out by the cooling liquid from the rods appear outside the active zone. Prompt detection of such perforations ensures fast removal of the damaged fuel assembly before the total radioactive contamination of the cooling chain. Thus, shutdown of the reactor can be avoided resulting in improvement of its ecological safety and economic effectiveness.

On the other hand, operative information on the neutron and gamma flows from definite fuel rods would result in better optimization of the fuel usage, because the degree of fissile material depletion will be determined for every separate fuel rod (or at least for separate assemblies of rods). So, more exhausted assemblies will be substituted earlier and the total coefficient of fuel usage will be increased.

Generally speaking, the data on differentiated flows of various ionizing radiations obtained with good temporal and spatial resolutions could ensure deeper understanding of the physical processes in nuclear reactors providing their better safety and effectiveness.

The Fukushima NPP disaster showed that the resource electrical supplies of the reactor cooling system based on diesel engines are not reliable because their moving parts can be broken by a water flow or any other accident, whereas fuel delivery can become problematic due to destruction of roads. Our experiments on propagation of ionizing radiation through triple-layer nanostructures including a strongly absorbing electrical conductor, an insulator and a

weakly absorbing conductor showed that such adequately constructed structures can provide direct, sufficiently efficient conversion of neutron and gamma radiations to electricity. These devices could be attached to the assemblies of the depleted fuel rods taken out of the active zones and stored in cooling pools until exhaustion of their radiation. On the other hand, besides essential improvement of reliability of resource electrical supplies of NPP these converters could change (transform) the situation with nuclear wastes. Nowadays they are the cause of social tensions due to their radioactivity, because such radioactive materials will become low cost sources of electricity and a source of commercial profit (see part 6). Annual exploitation of a nuclear power plant results in 1.500 cubic meters of radioactive waste per one GWt of electrical power which is the power of an average nuclear reactor [1,2]). Considering the fact that the Nuclear Power Plants around the world have now more than 400 nuclear reactors, the annual amount of radioactive wastes can be estimated as 600,000 cubic meters As it will be seen from the arguments presented below, direct conversion of their radiation to electricity can produce the amount of energy equivalent to construction of several tens of new nuclear reactors.

## 3. Unique features of nanostructures applicable to nuclear power engineering

The current situation with monitoring operation of nuclear reactors, utilization of depleted fuel rods and radioactive wastes described in the previous part shows that the safety and effectiveness of nuclear power engineering could be essentially improved with application of much faster detectors for separate control of different kinds of ionizing radiation inside the active zones and direct radiation-electricity converters for application at NPP as reliable sources of reserve electricity supplies as well as in nuclear waste stores for production of electrical energy for NPP own needs and supply of the surrounding areas. These problems have been pressing since the very beginning of nuclear engineering (see, for instance, books [4,6,7]). Moreover, nuclear reactions inside fuel rods are connected with the electrical charge separation process that is the main constituent of any generator of electrical energy. So, direct conversion of nuclear fission energy to electricity would be the most natural decision. But very short distances of charged particle propagation inside condensed materials require nanoscopic dimensions of construction elements of such converters [7–14]. So, when the main construction elements of experimental converters had thicknesses exceeding tens of micrometers, the energy effectiveness of the radiation - electricity converters was extremely low (less than 0.01 % with respect to the energy of absorbed ionizing radiation, see, for instance, one of the patents on this issue [16]). Hence, prior to active studies of technologies based on nano-dimensional materials and their properties, development of fast and radiation hard detectors as well as sufficiently effective radiation- electricity converters remained problematic.

A great body of the experimental and theoretical results on interaction of ionizing radiations with nanostructures demonstrated the prospects for developing either fast and radiation hard detectors for selective registration of gamma, neutron, alpha, beta and proton flows or direct converters of such types of radiation to electricity [10 - 14, 17 - 20]. Among the main features of nano-dimensional materials which provide successful development of their based devices

are the high probability of structural defects or electron excitation to reach quickly the external boundary of the nano-dimensional grain as well as a significant ratio of the amount of surface atoms to the total amount of the atoms in the grain. The surface -to-volume ratio varies inversely with nanoparticle size. On the other hand, absorption of any kind of external irradiation by a nanoparticle is proportional to its volume whereas re-emission of the absorbed energy to the external space is proportional to the surface. Hence, the absorption rate decreases in accordance with the third power of the particle radius whereas the decrease of the reemission rate is much slower, i.e. proportional to the square of the radius. So, at a certain radius value the re-emission rate becomes equal to the rate of external pumping. This means that particles with smaller radii will not change their stationary state when subjected to this kind of irradiation because the absorbed energy returns to the external space by means of reemission via the surface (by surface re-emission). This consideration belongs to the arguments confirming the increase of radiation stability of nano-particles following the decrease of their dimensions.

It is (rather) easy to show that the stability of nano-particles with respect to accumulation of radiation defects should be immediately enhanced with the decrease of their dimensions. Surfaces of solids are well known channels of annihilation of structural defects [22]. Thus, intensive irradiation-induced migration of point defects towards the external grain boundaries is able to enhance manifold radiation hardness of materials with respect to their bulk radiation hardness. This effect is explained by fast annihilation of radiation-induced defects as compared to the rate of irradiation pumping of the material. The time interval between the moment of creation of a point defect by radiation and its annihilation at the surface is inversely proportional to the square of the radius of the particle divided by the coefficient of diffusion of defects of this kind [20]:

$$\frac{1}{\tau_a} \approx \frac{D}{R^2} \tag{1}$$

On the other hand, the frequency of absorption of the ionizing radiation particle quanta by this particle is proportional to its volume multiplied by the intensity of the radiation flux and the absorption coefficient of the material (2)

$$v_{\tau} = F \cdot k \cdot R \cdot \pi R^2 = \pi F \cdot k R^3 \tag{2}$$

Comparison of expressions (1) and (2) enables to conclude that the critical intensity of radiation flux  $F_c$  that the particle subjected to irradiation can withstand without accumulation of defects and the corresponding degradation of its structure and properties is inversely proportional to the fifth power of the particle radius, i.e. it increases fast with decreasing dimensions:

$$F_c = D / \pi k R^5 \tag{3}$$

Therefore, it is clear that in this version of determination of radiation hardness for any irradiation intensity it is possible to select the critical nanoparticle radius when particles with smaller radii will withstand such radiation intensity, because annihilation of the point defect created by radiation due to the particle diffusion to the surface will proceed faster than production of a new one.

Besides, surface atoms belong to the category of structural defects themselves, because the atomic structure around them does not resemble the bulk structure. When the particle dimensions become less than 100 nm, the averaged concentration of these defects (i.e. the number of the surface atoms with respect to the total amount of the atoms in the particle) exceeds 1 %. This value is much higher than the averaged concentration of point defects which can be created in the bulk by radiation. So the influence of radiation-induced defects on the electron properties of the particles will be negligible with respect of the influence of the surface. Hence, either the atomic or electron structure of the nanoparticles subjected to irradiation should preserve their initial parameters even with high intensities appropriate for active zones of nuclear reactors.

The good stability of nanoparticles observed during their studies in scanning and transmission electron microscopes when they are subjected to irradiation of electron beams with high energy flow density shows that most of the incident energy is scattered outside. The possible scattering channels are secondary electrons and X-rays, thermal and optical radiation. Our studies of Xray excited luminescence of composites composed of inorganic polymer (polystyrene, etc.) bound nanoparticles showed that in this case most absorbed X-ray energy is not stored in the absorbing particle [21, 23-26, 28]. Other authors confirm enhancement of the radiation hardness of the materials transforming ionization radiations when the dimensions of their grains become nanoscopic [29, 30]. The energy is transferred to the surrounding organic molecules by means of secondary electrons and soft X-rays, excitons, light photons. These phenomena can be used for direct conversion of radiation energy to electricity in composite structures when one of the components is characterized by much stronger absorption of ionizing radiation than the other. In this case the radiation will produce separation of electric charges between these two components resulting in generation of an electromotive force. The idea of direct conversion of radiation to electrical energy has been discussed in scientific publications since the beginning of the 20th century [3-5]. But as it mentioned above, the experiments provide too low effectiveness of this conversion (not more than 0.01%) [16]. The abovementioned direct charging detectors can be classified as a type of low-efficiency radiationelectricity converter [2] which is too small for practical energy generators. But application of layered structures of radiation-electricity converters with nanoscopic ranges opens up new prospects for increasing conversion effectiveness. Theoretical estimations show that about 80% of radiation energy can be transformed to electricity by means of nano-dimensional converters with optimized structure, see, for example, patent application and publications [13,14, 17-20]. But the scheme of nano-structural converters described there implies artificial constructions made of nano-tubes and nano-layers which can be rather expensive to produce. Below we will present one of the possible and more economic ways for construction of radiation-electricity converters with an effectiveness sufficient for practical applications and a relatively low production cost [45, 49-51].

Up to now a wide variety of different techniques of preparation of nanoscintillators and other nanoscopic solids applicable to nuclear engineering have been developed [41, 42, 44–48].

# 4. Shaped sapphire and glass fiber radiation hard light guides with hollow cores

The considerations presented above show that applications of scintillating nanoparticles are rather promising for fast monitoring of radiation fluxes inside cores of nuclear reactors, because they possess sufficient radiation hardness for stable work within high intensity radiation fluxes. But the scintillation light emitted by these particles should be delivered to the outside of the core where the radiation intensity is much lower and photodetectors transforming the scintillation pulses to electric signals can be installed.

So radiation hard light guides are quite necessary. Usual silica fibers applied widely as light guides do not possess sufficiently high radiation stability [34]. We propose two other kinds of light guides that are radiation hard for effective transfer of light signals from cores of reactors to photodetectors. The first kind uses shaped sapphire fibers grown by the EFG technique. Several papers of independent authors devoted to shaped sapphire single crystals show that this material is able to withstand intensive radiation flows inside active zones of nuclear reactors [31–34]. Moreover, the color centers produced by irradiation deteriorate the optical transparency of sapphire in the ultraviolet region whereas we can choose nanoscintillators emitting light in the visible range. On the other hand, the color centers can be bleached by a laser beam with a wavelength in their absorption band. It should be emphasized that single crystalline sapphire tubes manufactured at the Institute of Solid State Physics (ISSP) RAS (Chernogolovka, Russia) were successfully used as guides of neutron flows inside the core of nuclear reactor in Grenoble and demonstrated a high radiation stability of their properties and structure [38, 39]. Another original technique for preparation of sapphire fibers with a modulated core of lateral structures made by regulation of the chemical composition of the melt delivered to the growth region have been developed [35-37]. Modulation of the chemical compositions of the fibers by varying the content activators of light emission along or across the fiber ensures control of spectral compositions of light emission from different fiber regions. Below we will describe the possibilities of differential radiation flow monitoring achieved by modulation of local chemical compositions of sapphire fibers doped by titanium. The examples of sapphire fibers grown in Chernogolovka are presented in Figs. 2–5.

The core of another radiation hard fiber light guide does not contain any solid, i.e. it is empty [43]. So, nothing can be damaged by irradiation of the core. The localization of the light flow along the fiber axis is achieved by the specific geometry of the micro-channels in the fiber cross – section. The channels are parallel to the fiber axis whereas in the transverse direction they form a photon crystal limiting light propagation in radial directions (Fig. 6). The design was patented by ISSP [46].



Figure 2. Non-doped single crystalline sapphire fibers grown by EFG technique at ISSP [35-37].



### 5. Differential and fast monitoring of radiation flows inside active zones for better safety and optimized operation of nuclear reactors

Shaped sapphire nanocrystalline scintillators and fibers possess high radiation hardness and can be used effectively for fast differential control of radiation fluxes in active zones of nuclear reactors. Due to their small dimensions, the composition and structures of nanoscintillators

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**Figure 4.** Cathode luminescence micrograph of the cross-section of the sapphire fiber with doped core (upper photo) and radial distribution of the luminescence intensity across the radius (below). Titanium was used as the dopant.

are regulated over a wide range providing excellent opportunities for optimized selection of materials for registration of specific types of radiation. Nanocrystalline particles are placed at lateral surfaces of fiber light guides which also possess high radiation hardness.. The light signals delivered to the outside of the active zone are characterized by millisecond duration and transfer separate information on the intensities of the radiation flows of fast and thermal neutrons, alpha and beta particles, gamma - quanta in definite points of the reactor core to the entrance of the control system. Such a design provides well - timed preventing of accidents and optimized control of the reactor operation. For instance, fiber light guides placed along the claddings of the fissile rods enable timely control of the wall tightness. With intact walls the scintillators register neutron and gamma radiation only, because alpha and beta particles cannot penetrate through the cladding. But as soon as the wall has become too thin, beta emission occurs and with thinning of the wall there also appears alpha - emission. In this case the light guide transfers a warning signal to the regulation system much earlier than the radioactive substances reach the outside of the active zone. Thus, fiber- scintillator detectors ensure timely preventing of diverse accidents which increases significantly the reliability of the reactor operation.



**Figure 5.** Cathode luminescence of shaped sapphire fiber with modulation of activator doping along the fiber axis. Titanium was used as the dopant.



**Figure 6.** Radiation hard light guide with empty core, transverse propagation of light limited by photon crystal geometry [43, 46].

As it was mentioned above, development of differential monitoring of radiation fluxes inside active zones of nuclear reactors with temporal resolution much better than a second is desirable for deeper understanding of the physical processes inside nuclear reactors, preventing of leakages of radioactive wastes, more complete burning of fission fuel, etc., in other words, for general improvement of reactor safety and energy effectiveness. Application of nano-dimensional scintillators and radiation hard light guides which can work reliably inside active zones of reactors allow for resolving this problem. High radiation hardness of nano size scintillators as well as other nano-dimensional objects was discussed in the previous part. Scintillation light registration of the level of radiation flow in a certain point of the active zone will be transferred

by of the light guides to the outside of the active zone where photodetectors can be installed for transformation of light to electric signals for further computer processing (Fig. 7).



Figure 7. Scheme of transfer of scintillation signals to photodetectors outside the reactor core.

At least two kinds of radiation hard light guides can used for the delivery of scintillation signals to the outside of the active zone: shaped sapphire and glass fibers (Fig. 2) with empty cores with cross-sections in the form of photonic crystals (Fig. 6). According to the measurements made by V.V. Nesvizhevskiy at the Institut Laue-Langevin (Grenoble, France) [38, 39], the products made from shaped sapphire crystals at ISSP RAS invariably retain their parameters in the active zone of the reactor. The data are supported by the direct experiments on the transparency of sapphire fibers subjected to ionizing irradiation [31-34]. On the other hand, light guides with transversal geometry of photonic crystals contain empty cores whose transparency cannot be deteriorated by radiation defects [30, 46]. Based on the above, the researchers at the ISSP RAS are developing various combinations of nanocrystal scintillators and light sapphire fiber waveguides for rapid differential control of radiation flows inside nuclear reactors [40 – 42, 47]. Radiation hard nano-scintillators are placed at the lateral guide surfaces (Fig. 8).

As one of the nano-particles absorbs ionizing radiation it produces a light outburst of a definite color. Here differentiation means to obtain separate information about the intensities of the fluxes of the fast and slow neutrons, alpha-, beta-, and gamma radiation in specific points of the active zone. Such information appears quite useful for improving the safety of the reactor operation and optimizing its operational parameters (for example, for improve the burnup fraction of the fissile materials in the fuel elements).

The information on the intensities of different ionizing radiations is differentiated with the aid of the nanoscintillators on the basis of a wide variation of their structure. The ISSP RAS has developed the techniques of synthesis of nanocrystal scintillators with largely varied compo-



Figure 8. Deposition of nanoscintillators with various dopants at lateral surfaces of radiation hard light guides.

sitions and sizes to ensure selective information on any specific type of radiation (Fig. 9) [21, 23-25, 44].



Figure 9. Lutetium-sodium fluoride nanoscintillators doped with cerium activator.

Thus, the prevalence of heavy elements (tungsten, lutecium, lead, etc.) enhances sensitivity to hard gamma-radiation. When alloying with gadolinium, lithium, boron increases the role of scintillations induced by thermal neutrons. Hydrogen saturation of nanoparticle allows receiving scintillation signals from the fast neutrons. The selective sensitivity of the radiation detectors to fast neutrons can be ensured by making compositions of inorganic nanoscintillators and organic molecules [23, 24, 41, 47, 48]. When fast neutrons collide with the protons of

the organic matter, the protons penetrate fast into the inorganic particles contacting with these molecules and exciting their scintillation radiation. For selectivity in relation to alpha-ray and beta-ray radiation the depth of its penetration into the material may be strongly limited (alpha-rays: by units of microns, beta-rays: by tens of microns). Thus, for selective recording of the charged particles, it is sufficient to place the relevant nanoscintillators in a thin layer on the outer surface of the light waveguide. The scintillation signals from different radiations may be of different spectral compositions. For example, nanoscintillators activated with cerium will produce blue flashes, green flashes will be produced by terbium and red flashes by europium.

As stated above, nanoscintillators may be fixed on the lateral surface of the same sapphire light waveguide in order to register different radiations. Separating the scintillators by the spectral compositions of the light emission at the light waveguide output enables to determine the specific radiation in the relevant point of the active zone. When nanoscintillators are uniformly positioned along the whole light waveguide length, the coordinate of the scintillation radiation point may be determined from the ratio of the signal amplitudes at the opposite ends of the light waveguide. As the light passing along the light waveguide reduces exponentially, the difference of the distances between the point of radiation of scintillation flash and the ends of the light waveguide is determined as the logarithm of the ratio of intensities of the relevant signals (Fig. 10).



Figure 10. Scheme showing localization of point of damage of fuel rod cladding.

The control of impermeability of the fuel-element cladding may serve an example of the efficiency of separate recording of ionizing radiations for improving the reactor safety. When a fiber sapphire light waveguide with fixed nanoscintillators is positioned along its outer wall,

as described above for specific radiations (see Fig. 8), and the cladding is fully impermeable, only gamma and neutron radiation signals will be recorded from the light waveguide (as the alpha and beta particles generated by the nuclear reactions cannot pass through an impermeable wall). With thinning and further perforation of the cladding wall the alpha and beta radiations will start. Hence, the sapphire fiber located near the fuel element will send a signal "colored" for the alpha and beta radiations to the control equipment (first, the beta-ray signal will appear; then, as the cladding "perforation" develops, the alpha particle signal will occur). As nanoscintillators are quite fast radiation detectors, and the signals go along a light waveguide at the speed of light, the response time of the control system to a cladding defect will not exceed milli-seconds. This is much faster than the moment when a radioactive leak appears at the active zone outlet. Hence, efficient emergency actions can be taken much faster. Hence, fixing a sapphire light waveguide with specially adjusted nanoscintillators to every fuel element will significantly improve the reactor safety. Moreover, this method of control ensures, without extra risk, the service life of a fuel element in the active zone, improving the fuel burn up fraction and the reactor efficiency as a whole. It is important to note that beta radiation control ensures detection of corrosion-induced thinning of the fuel-element cladding down to several tens of microns prior to perforation. Thus, active zone emergencies can be prevented as such, increasing, in parallel, the service life of the fuel assembly.

Composite scintillators made of a combination of inorganic nanoparticles and organic luminofors efficiently record fast neutrons, and, due to the fast transfer of the electronic excitations from the inorganic nanoparticle to the organic phosphor increase considerably the response speed of the radiation detectors.

### 6. Practical development of direct radiation-electricity converters based on nanostructures

In the formation of nanostructures, due to the small sizes of their grains, the role of the interactions of the atomic and electronic excitations with the interface surfaces is extremely great. The radiation defects diffuse rapidly to the surface and annihilate on it. The nonequilibrium electrons formed upon interaction of the ionizing radiations with the nanoparticles collide repeatedly, with their edges during their lifetime. Bremsstrahlung and transition radiations are generated during these collisions and electrons are also emitted.. The above processes rapidly carry the consequences of the radiation effects outside; hence, the radiation strength of the nanostructures increases manyfold as their sizes decrease. On this basis, several new devices are proposed; which could to stabilize work in a field of intense radiation, ensuring efficient transformation of ionizing radiations into electric power, based on the external photoelectric effect and thermal electromotive force. For example, in a nanostructure of alternating "light weight conductor – insulator – heavy metal" layers gamma radiation is absorbed, mainly, in the heavy metal. This induces emission of photoelectrons; some of them coming, via the insulator layer, to the light weight conductor (these may be, for example, thin aluminum or graphite layers of). Thus, the heavy metal gets a positive charge while the light weight conductor acquires extra electrons and gets a negative charge, i.e., an electromotive force is generated. When the electrodes interlock via a current receiver, work is produced in the latter. Thus, the energy of the ionizing radiation is directly transformed into electric energy. The produced power is determined by the difference of the potentials between the electrodes and summed current which is directly related to the number of the electrons transferred from one electrode to the other. Meanwhile, this number depends upon the configuration of the heavy metal surface because only the electrons whose normal to the surface component of the momentum exceeds a certain threshold will take part in the charge transfer. From this viewpoint, the flat geometry of the electrons can move in the direction required. The coaxial configuration with an insulator layer and a tubular light weight conductor around the heavy metal axis (this configuration is close to that of the direct charge transducers used to record the ionizing radiations [1, 2] is much closer to the optimum. When the thickness of the heavy metal conductor is comparable to the track length of the hot photoelectrons, a significant share of them will take part in the electrode charge exchange, and, hence, in the transformation of the absorbed radiation into electric energy.

From the viewpoint of the above, the honeycomb structure of the cylindrical cells with conductive walls when the core of the cells is filled with heavy metal separated from the walls with a thin insulator layer seems appropriate. At the ISSP RAS we use biomorphic silicon carbide structure as the basis for such a device (see Fig.11). Such structure is obtained as follows: a cross-cut piece of wood (in this case spruce was used) was subjected to pyrolysis, i.e., thermal disintegration in an oxygen-free medium. The pyrolysis yielded a carbonic frame reproducing the structure of the channels parallel to the tree growth axis (upper Figure). Following this, the carbonic frame was saturated with silicon yielding a silicon carbide frame reproducing the original morphology of the wood [49-51]. The silicon carbide surface was oxidized to obtain a silicon dioxide insulating layer. Next, a metal layer was applied to the inner surfaces of the channels. In such a configuration, the total current-forming surface is fairly large: with the spruce cross-section surface of 1 cm<sup>2</sup> and thickness of 1 cm the inner surface of the cells is about 3000 cm<sup>2</sup>. The Figures 11 and 12 show the inner configuration of the cell with visible nanograins of the silicon carbide (that make the surface even larger). The carbon of the pyrolyzed wood frame forming the integrated structure for the whole frame works as a light weight conductor. In this case, the silicon carbide plays the role of a mechanical frame. It is made sufficiently thin to allow transfer of photoelectrons emitted from the heavy metal to the light weight conductor.

In the second variant of thermoelectric generator the silicon carbide formed as described above plays the key role in transforming the heat into electricity. In one of the variants of the generator the contact surface of the silicon carbide with the carbon frame acts as the double charge layer. On thermal generation inside this layer of nonequilibrium electrons and holes the inner difference of potentials carries them in opposite directions generating electric current. The intensity of the current is determined by the surface of the silicon carbide — carbon contact that is, as it has been shown above, quite large.

A set of tubular radiation-electric and thermal-electric converters that may be coaxially put onto a cylindrical assembly of fuel cells will, at a 100 kW total flow of energy from the assembly,

generate at least 20 kW of electric energy. As the fuel containment sump of the reactor can contain several hundred assemblies of the kind (depending upon the reactor type) the total power output from the sump will be several megawatts. Compared with the working reactor power this makes only fractions of a percent, and, hence, is insignificant. However, the Fukushima 1 accident has shown the critical need, in an event of emergency, for a redundant and absolutely reliable source of electric power. For this purpose NPPs usually have diesel generators. However, their start requires at least a minute, and they need fuel to be supplied. Moreover, a serious accident may damage the moving parts of the diesel generators. Meanwhile, radiation and thermoelectric generators need no fuel, have no moving parts, and work nonstop; hence, they can be much more reliable. The key issue of reliability of such generators is the radiation strength of their generating units. As the main elements of these generating units are nanocrystals, a high radiation strength of the generating units may be expected.

As for nuclear power engineering, it is important to point out that the silicon carbide is a wide zone semiconductor, with a high thermochemical stability, and capable to retain its high semiconductor capacities up to 600÷800°C. Thus, generally speaking, it can work not only in the sumps of the used fuel assemblies but in the active zones of reactors, as well.

There is another prospect for the nuclear power engineering use of the above radiation-electric and thermal-electric generators based on biomorphic silicon carbide: standalone electricity sources for radioactive waste storage sites. As such storage sites are arranged, as a rule, in back lands, the possibility to get the electric energy without fuel supplies or power transmission lines seems extremely attractive..



Figure 11. Electronic microscopy image of cross cut surface of spruce after pyrolysis.



**Figure 12.** High magnification image of morphology of inner wall of channel in pyrolyzed wood after synthesis of silicon carbide by siliconizing.

 $\gamma$ -Ray irradiation of layered structures consisting of interleaving layers of heavy metal, insulator, and lightweight metal produces electromotive power [49-51]. This occurs due to the prevailing absorption of  $\gamma$ -radiation with heavy metal, thus resulting in emission of fast electrons. Some fraction of them penetrates the lightweight metal making it negatively charged. The contact of the lightweight and heavy metals under an external load makes these electrons return through it into the heavy metal, thus performing useful work. The ratio of this work to the radiation energy absorbed by the layered structure determines the efficiency of radiation-electricity conversion in devices of this kind.

Ideally, all electrons dislodged from the heavy emitter reach the lightweight conductor and are absorbed there. If each of these electrons returns from the collector to the emitter through the external circuit, electric current *I* will occur in the circuit.

$$I = e \cdot N = e \cdot \gamma \cdot \hbar \cdot \omega / (K + W), \tag{4}$$

where e is elementary charge;  $\hbar$  is Plank's constant;  $\omega$  is circular wave frequency of electromagnetic field; *K* is electron kinetic energy;  $\gamma$  is photon flux absorbed in the emitter material; *W* is electronic work function.

In the external circuit with electrical resistance R, electric power will occur. It is possible to estimate the efficiency ( $\eta$  is the efficiency factor) of the system, which is determined as the ratio of power generated in the external circuit  $W_{ext}$  to the power of the absorbed  $\gamma$ -rays  $W_{abs}$ :

$$\eta = W_{ext} / W_{abs} = I^2 \cdot R / \gamma \cdot \hbar \cdot \omega = \left[ \left[ e \cdot \gamma \cdot \hbar \cdot \omega / (K + W) \right]^2 \cdot R \right] / \gamma \cdot \hbar \cdot \omega,$$
(5)

From formula (5) one can see that as the electron kinetic energy K in the denominator decreases, the efficiency factor is expected to increase monotonously until the energy reaches zero; however, in order to have current *I* in the external circuit, there should be potential difference  $\phi = I \cdot R$  between the emitter and the collector.

For the electrons escaping from the emitter to overcome this potential difference and to reach the collector, their kinetic energy should be not lower than this difference multiplied by elementary charge:

$$K \ge e \cdot \phi = e \cdot I \cdot R = e^2 \cdot \gamma \cdot \hbar \cdot \omega \cdot R / (K + W).$$
(6)

From (6) there follows a quadratic equation for the minimum acceptable value of kinetic energy of emitted electrons, by solving which we obtain:

$$K = -W/2 + \sqrt{(W^2/4 + e^2 \cdot \gamma \cdot \hbar \cdot \omega \cdot R)}.$$
(7)

As one can see from expression (7), in the ideal case, when the system is pumped with gammarays so much that the second term under the square root  $e^2 \cdot \gamma \cdot \hbar \cdot \omega \cdot R$  is much higher than the first one  $W^2/4$  (therefore, power consumption of the work function can be neglected), the efficiency factor value in expression (5) approaches unity within the accuracy of the energy spent on the work function.

The physical sense of this derivation is as follows: when the kinetic energy of electrons emitted from the emitter to the collector becomes equal to their electrostatic energy between the emitter and the collector, the electrons enter the collector with zero kinetic energy, thus eliminating heat release losses. Therefore, the efficiency factor approaches unity, i.e. the ideal value. But, if kinetic energy exceeds the value

$$K = e \cdot \sqrt{\gamma \cdot \hbar \cdot \omega \cdot R}, \tag{8}$$

then, according to expression (5), the efficiency factor starts to decrease in inverse proportion to kinetic energy.

With the opposite approach, i.e. when the first term under the square root in equation (7) is much higher than the second one, the kinetic energy of emitted electrons only slightly exceeds the work function; therefore, a sufficiently high charge cannot be accumulated on the collector, and the efficiency of the system will be always low.

Despite the fact that the efficiency of an idealized system could reach 100 %, the experimental data reported in the patent [16] have shown an efficiency factor of ~  $10^{-5}$  %. Below we analyze the reasons of such a considerable discrepancy, and discuss the ways to improve the efficiency of conversion of ionizing-radiation energy into electricity up to a practically significant level.

Non-optimal geometry of converters. In the experiments mentioned above, the layered 1. converters were of planar geometry. At photoelectric absorption and Compton scattering of gamma-rays, the directions of outgoing electrons receiving their energy do not coincide with the initial direction of radiation propagation; hence, the planar geometry means that traveling of the predominating fraction of these electrons is strongly deviated from the normal line to the outflow surface. Due to this, the distance to the collector increases and can considerably exceed the full range of the fast electron. Accordingly, such electrons fail to leave the plate and participate in formation of radiation EMF. From this point of view, spherical geometry for the heavy metal fragments absorbing gamma-rays seems to be optimal. But for these photoelectrons to travel into the lightweight metal, the size of the spheres should not exceed their ranges. Besides, all spheres should be electricallyconnected to each other. Since the ranges of fast electrons in heavy metals are of the micron or even submicron scales, manufacture of such a device is a complicated engineering problem. As an intermediate stage, the geometry of biomorphic silicon-carbide or carbon matrices described earlier [49-51] can be used, where heavy metal (lead or tin) fills parallel micro channels formed by means of pyrolysis and siliconizing of wood sections (see figure below). Although the geometry of parallel metal micro fibers is far from the geometry of spheres, it is much more beneficial for photoelectron emission in comparison with the planar geometry.



**Figure 13.** Matrix of silicon carbide microchannels obtained by wood pyrolysis filled with tungsten (left picture); and with lead (right picture).

2. *Excessive thickness of heavy metal layers.* In the patent [16], the planar structure "heavy metal – insulator – lightweight metal" is formed by layers, each of tens of micrometers thick. This thickness is acceptable for the light metal (collector) that is intended to capture emitted electrons; but for the heavy metal (emitter) such thicknesses significantly exceed the optimum values and contribute to a considerable decrease of the effect of radiation separation of charges. A considerable fraction of both primary and secondary electrons (occurring in collisions with the primary ones via collision ionization) fail to travel such long distances and stop in the heavy metal. Naturally, these electrons do not participate in the generation of useful current in the external circuit.

3. *Excessive thickness of the insulator.* In the case of excessive insulator thickness, many of the electrons emitted by heavy metal are stopped in the insulator layer. Apart from the fact that these electrons do not reach the light metal and accordingly drop out of the process of formation of useful current, they considerably decrease the electrical resistance of the insulator, contributing to the return of the emitted electrons from the lightweight metal to the heavy metal not via the external circuit, but immediately back through the insulator layer. Reduction of the insulator thickness will allow increasing the conversion efficiency considerably. For example, with the insulator thickness reduced from 200  $\mu$ m to 1  $\mu$ m in the papers [49 - 51], the conversion efficiency increased by 3 orders. At the same time, this value should be sufficient to prevent breakdown, as well as large leakage current.

The most preferable insulator is a vacuum layer, where emitted electrons make no impact such as collision ionization, and consequently, the probability of backflow is minimized, being determined only by the tunnel process. With the purpose to create an insulator with properties close to vacuum, it is possible to use a layer of aerogel (material formed by fibers of such dielectrics as aluminum oxide or silicon oxide, the porosity of which exceeds 95 % [15]), and mechanical strength allows holding the layers of heavy and lightweight metals at a certain distance preventing their direct contact.

4. *Non-optimal energy of electrons leaving the emitter*. The optimal energy of electrons leaving the emitter corresponds to the situation, when by the moment of entering the collector, all the kinetic energy has already been spent on overcoming the repulsive potential between the emitter and the collector. Besides, the electron needs a certain kinetic energy store for losses when travelling in the insulator. Note that in a real situation, electrons leaving the emitter have certain statistical spread of energy; therefore, it is almost impossible to achieve an exact situation, when all electrons would precisely spend their all kinetic energy before reaching the collector. The energy required for electrons to exit from the emitter is largely determined by the range of the electron in the absorbent material and by the energy of initial radiation. Therefore, adjustment of exit energy primarily implies selection of the optimal size and shape of the emitter surface, and this adjustment should be performed with reference to the technical parameters of particular operating conditions.

Conversion of energy of fast neutrons created in fission processes to electricity needs preliminary transfer of a neutron kinetic energy to kinetic energy of a proton. This necessity is explained by the too small cross - section of a neutron interaction with electrons and a much bigger corresponding cross - section of a proton. The micro-channels of biomorphic matrices can be filled with material containing a high percentage of hydrogen atoms. For example, pure water  $H_2O$  can be used as a preliminary converter of this kind. Parrafin and other organic materials are acceptable as well. Protons generated by fast neutrons will produce showers of fast electrons which will penetrate through the insulator nano-layer to the light conductor as in the case described above.

It should be noted that sources of ionizing radiation can be placed just inside the microcapillaries. For example, it can be metal uranium or uranium oxide. The experimental studies of nuclear energy conversion to electricity in plane layered structures with U<sup>235</sup> [13, 14] revealed a conversion efficiency of 25 %. Microcapillary structures based on biomorphic matrices provide much better geometry for penetration of electrical charges generated in nuclear reactions through the insulating layer to the collector. So we can assume that filling of biomorphic matrices described above with nuclear fuel will increase the conversion efficiency to higher values.

#### 7. Conclusion

Energy effectiveness and ecological safety of nuclear power engineering can be improved essentially with application of nanoparticles, layered nanostructures, shaped sapphire and microporous light guides as well as microcapillary biomorphic matrices. On the one hand, usage of these materials can improve essentially the situation at operating NPP and at stores of radioactive wastes. Nanoscintillators fixed at lateral surfaces of radiation hard light guides are capable to deliver instantaneous information about local situations at separate fuel rods and their assemblies with temporal resolution of milliseconds and spatial resolution of centimeters. This information will form the basis for much deeper understanding of operation of rather complicated multi-component objects such as NPP. Besides, this information will improve essentially the safety and effectiveness of performance of nuclear reactors.

Direct converters of radiation energy to electricity will improve significantly the safety of operation of nuclear reactors and transform radioactive wastes from the object of continuous anxiety of the society to ecologically safe and low cost fuel.

Moreover, qualitatively new designed of nuclear reactors can be developed based on nanostructured fissile fuel deposited inside micro-capillary matrices. Reactors of this kind will produce most energy by direct conversion of nuclear energy to electricity. Due to this radical improvement their construction will be more simple, economic and safe. The volume of radioactive wastes will also be reduced.

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