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Research Article

Radiation-induced separation and accumulation of electric charge in supercapacitors*

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Abstract

In current sources with a radioactive isotope (CSRI), nuclear energy is directly converted into electricity due to the separation of electric charges during the decay of radioactive isotopes. It was previously shown that asymmetric supercapacitors can be used as CSRI prototypes if, after being exposed to pulsed reactor irradiation, the electric charge on their plates increases to several coulombs as a result of internal induced activity. In this paper, the electric charge separation and accumulation in supercapacitors were studied directly in the process of neutron irradiation.

The study was focused on the electrophysical characteristics of cylindrical supercapacitors with an organic electrolyte produced by JSC "ELEKOND". A comparison of symmetric and asymmetric supercapacitors showed that an effective charge accumulation occurs in the asymmetric capacitors: it is independent of the neutron flux density and determined by the absorbed radiation dose. The electrical voltage between the plates of a symmetrical supercapacitor with a capacity of 100 F during irradiation up to an absorbed dose of 50 Gy reaches 1.24 mV. When asymmetric supercapacitors are irradiated with the same dose, a significant increase in the potential difference up to 1.15 V is observed during irradiation and for a long time afterwards ($1.5 \cdot 10^5$ s) due to the electric charge redistribution (~ $5 \cdot 10^{-3}$ C) in the electrolyte and carbon particles with the formation of a double electrical layer. The post-radiation increase in the capacity of asymmetric supercapacitors is ~ 5 mF.

Keywords

Supercapacitor, neutron irradiation, radiation-induced electric charge

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Introduction

Currently, chemical (lithium) power sources are widely used for small-size equipment. However, these sources have limitations on miniaturization and a narrow range of positive and negative operating temperatures; they require periodic recharging, and their specific capacity does not exceed 1 kW/kg (Maltsev (ed.) 2005, Verner et al. 2008). The most promising sources can be current sources of a constant readiness, based on the direct conversion of nuclear power into electricity, in which the decay of radioactive isotopes leads to a radiation-induced separation of electrical charges (Anufrienko et al. 2006, 2008, Chernov et al. 2010, 2011, 2015, 2016). The expected characteristics of a current source of this type (Chernov et al. 2015, 2016) significantly exceed those of lithium ones and are characterized by an energy reserve of more than 10^3 kWh/kg, maximum power of 25 kW/kg and a minimum volume of $10^{-2} – 10^{-3}$ cm^3. In this case, the service life depends on the half-life of the isotope used, for example, the lifetime of promising isotopes of americium-241 is 460 years and of carbon-14 is 5700 years, respectively. The main application areas of CSRI are microsystem equipment and microelectromechanical systems, facilities in remote and hard-to-reach places operating under extreme conditions, monitoring, communication, and navigation systems.

In preliminary studies (Anufrienko et al. 2006, 2008, Chernov et al. 2010, 2011, 2015), various MDM (metal-dielectric-metal) structures were studied as converters of nuclear energy into electricity. They use the energy of fast particles due to the accumulated energy of secondary electrons emerging from the surface of MDM-structure layers. Calculated and experimental data were obtained on the specific energy, current and voltage under various radiation effects. It is shown that significant efficiency coefficients of radiation energy conversion can be obtained by nanostructuring MDM systems to achieve a larger working surface area of charge separation. Calculations based on the experimental data showed that, for CSRI at an absorbed dose rate of 1 Gy/s from the isotope decaying inside, currents up to 100 μA arise at interphase areas of $10^3 – 10^4$ m^2 (Chernov et al. 2015). Such areas are realized in supercapacitors and ionistors, where a nanostructured carbon electrode is the main component of the MDM structure.

Supercapacitors with capacities up to 22 F based on the carbon-graphite mixture and α-alumina, using a solid inorganic electrolyte Ag_4RbI_5, were examined as CSRI prototypes (Chernov et al. 2015). After pulsed reactor irradiation up to doses of 2 Gy (for fast neutrons), active isotopes were formed in the electrolyte of ionistors, mainly with I-131 and Rb-87, as sources of primary charged particles with an output up to 10^7 part./s. The post-radiation change in the electric charge on the plates as a result of internal induced activity reached 3 C. It was shown that gamma irradiation does not affect the charge separation and accumulation processes.

For further CSRI considerations, it is necessary to physically simulate electric charge separations in supercapacitors when radioactive isotopes are introduced into them. In this paper, the simulation was performed using measurements of the electrophysical characteristics of supercapacitors in which penetrating continuous neutron radiation plays the role of a radioactive isotope.

Methods and measurement result

The study was focused on the electrophysical characteristics of cylindrical supercapacitors with an organic electrolyte produced by JSC "ELEKOND" with a volume of cm^3 and a carbon layer weight of 3.5 g (specific surface = 1800 m^2/g) and a paper separator. For irradiation, a neutron source was used, providing an absorbed dose rate up to 0.1 Gy/s. The absorbed dose rate of accompanying gamma radiation did not exceed 0.03 Gy/s. The measurements were carried out using a combined multimeters SCH300 and DT 9208 in a sealed container with pre-annealed silica gel.

The voltage/current values were measured in a symmetrical supercapacitor with an electrical capacity of 100 F during and after irradiation at a load resistance of 10 kΩ. The measured voltage (U) values are shown in Fig. 1. In the asymmetric supercapacitors, one of the plates was a smooth aluminum foil. When their electrophysical characteristics were being measured, the voltmeter input resistance was 2 MΩ. The measurement results are shown in Fig. 2.

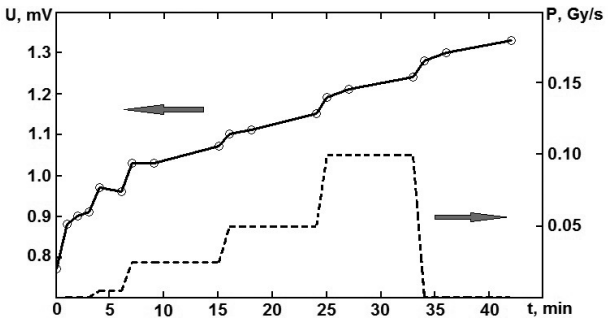

Figure1. Time-dependences of the absorbed dose rate on the neutron source P and voltage U between the plates of a symmetrical supercapacitor.

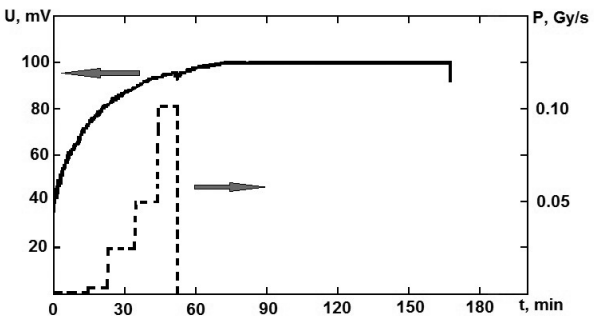

Figure 2. Time-dependences of the absorbed dose rate on the neutron source P and voltage U between the plates of an asymmetrical supercapacitor.

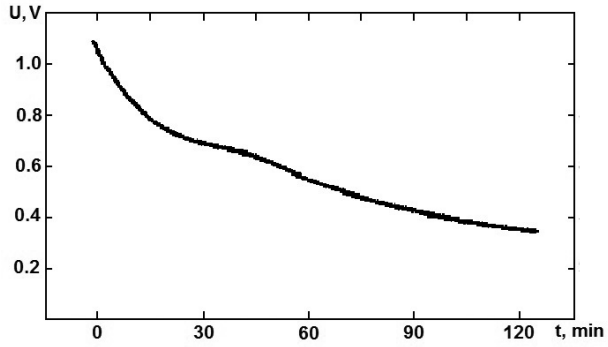

Figure 3. Changes in the electrical voltage between the asymmetrical supercapacitor plates the day after irradiation. The beginning of measurements corresponds to $t = 0$.

The electrical voltage between the asymmetric supercapacitor plates were also measured after $8 \cdot 10^4$ s after irradiation. The measurement results are shown in Fig. 3. Prior to the measurements, the supercapacitor was in the open state. The subsequent voltage measurements after another $8.6 \cdot 10^4$ gave a value of 1.15 V.

Discussion

Based on the results of measurements of the symmetric supercapacitors, it can be concluded that the electrical voltage between their plates does not depend on the neutron flux density, but is determined by the radiation dose. After irradiation ceased, the electrical voltage between the plates continued to grow. This means that the charge separation is associated with internal long (at times of 10^2–10^3 s) processes in the electrolyte. In the supercapacitors, a double electric layer with a specific area up to 10^3 m^2/g is formed on the extremely developed inner surface of the carbon material as a result of ion accumulated in the solvate shell of solution molecules near the plate surface. The charge of this layer is compensated by the electric charge directly on the plate surface.

In (Chernov et al. 2016), it was shown that the asymmetry (in area) of the plates provides an effective separation of the charge between them. In (Chernov et al. 2015, 2016), the radiation-induced specific surface density of the electric charge on the conductor of the MDM structure was obtained under neutron irradiation or heavy-ion irradiation: $\rho \sim 10^{-8}$ C/Gy·m^2. With a plate area up to 10^4 m^2 and an absorbed dose of 50 Gy, the value of the radiation-induced electric charge is about $5 \cdot 10^{-3}$ C. The curve in Fig. 2 reflects the charge accumulation kinetics during irradiation as well as the process of formation of a double electric layer due to the diffusion redistribution along the plate surface of the charge accumulated in the electrolyte. Since a significant increase in the potential difference is observed for a long time after irradiation, when the radiation charge separation has already occurred, it can be assumed that, at long times, the capacity of the asymmetric capacitor increases due to the electric charge redistribution in the electrolyte and carbon nanoparticles with the formation of a double electric layer. Thus, the overnight voltage measurements ($8.6 \cdot 10^4$ c) showed a tenfold increase. This corresponds to a post-radiation capacity increase by a factor of 10 to ~ 5 mF. The appearance of voltage and current almost two orders of magnitude less between the symmetric supercapacitor plates should be attributed to the presence of some asymmetry between them.

This interpretation of the experimental data fully corresponds to the modern concepts of a supercapacitor as a nonlinear system in which the capacity depends on measurement conditions, applied voltages, frequencies, etc. For example, in (Uchaikin et al. 2016), it was shown that the supercapacitor response to charging mode variations does not agree with the linear response theory. The relation of this nonlinearity to processes in the double electric layer, the morphology of porous plates, and the charge-exchange effect is discussed. Broadly speaking, the physical picture of processes occurring in the supercapacitors, even in "equilibrium" conditions, without irradiation, is not yet completely clear. In general, empirical approaches are used (Bertrand et al. 2010, Stldakova et al. 2015, Buller et al. 2002, Ricketts et al. 2000, Yu A et al. 2013, Shi L et al. 2008). The authors of (Bertrand et al. 2010), when simulating the supercapacitors, point to the existence of two classes of empirical models that take into account the charging dependence on the applied voltage (a characteristic of nonlinearity) or fractional-differential (linear) models.

The measured discharge kinetics of asymmetric supercapacitors (see Figure 3) suggests the existence of relaxation stages: the first is at times of 10^2–10^3 s, the second is more sloping at long times. The non-Debye relaxation in supercapacitors is a well-known experimental fact. In (Uchaikin et al. 2016), the discharge dynamics was measured in a remote time domain (up to 10^4 s) of sample Panasonic supercapacitors with a maximum capacity of 1 F, using an organic electrolyte and activated charcoal in the form of a finely dispersed fraction as plates – at short times, the time-current dependence is close to the stretched exponential law; at long times, the asymptotic behavior of the kinetic curves is power-dependent.

Concluson

Due to physically simulated processes in CSRI, neutron irradiation of the supercapacitors with an organic electrolyte produced by JSC "ELEKOND" shows that the electric charge separation and accumulation do not depend on the neutron flux density, but are determined by the radiation dose and the degree of asymmetry of the plates. As a result of long processes of formation of a double electric layer in an organic electrolyte after irradiation, the electrical voltage between the plates increases.

The electrical voltage between the plates of symmetrical supercapacitors with a capacity of 100 F during irradiation up to an absorbed dose of 50 Gy reaches 1.24 mV.

When asymmetric supercapacitors are irradiated with the same dose, a significant increase in the potential difference up to 1.15 V is observed during irradiation and for a long time afterwards ($1.5 \cdot 10^5$ s) due to the electric charge redistribution (~ $5 \cdot 10^{-3}$ C) in the electrolyte and carbon particles with the formation of a double electrical layer. The post-radiation increase in the capacity of asymmetric supercapacitors is ~ 5 mF.

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