

TECHNICAL PHYSICS LETTERS

Founded by Ioffe Institute

Published since January 1975, 12 issues annually

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ISSN: 1063-7850 (print), 1090-6533 (online)

TECHNICAL PHYSICS LETTERS is the English translation of ПИСЬМА В ЖУРНАЛ ТЕХНИЧЕСКОЙ ФИЗИКИ
(PIS'MA V ZHURNAL TEKHNICHESKOI FIZIKI)

Published by Ioffe Institute

Radioisotope power source based on artificial diamond activated by carbon isotope and InGaP-photoconverter

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Received December 26, 2025

Revised March 25, 2026

Accepted March 28, 2026.

A prototype of a radioisotope power source has been created based on a self-glowing (450–550 nm) single-crystal diamond $^{12}\text{C}:(^{14}\text{C})$, enriched with the carbon isotope ^{14}C , and $p-n$ photoconverter based on InGaP. With a diamond self-luminescence power density of $\sim 0.8 \text{ nW/cm}^2$, the photoconverter provided an output electrical power of $16.5 \cdot 10^{-12} \text{ W/cm}^2$. The prospects for improving the optical radiation intensity of diamond, which will provide an output electrical power density of up to $\sim 1 \text{ nW/cm}^2$, are investigated.

Keywords: radioisotope power source, self-glowing single-crystal diamond, carbon isotope, InGaP-based $p-n$ photoconverter, diamond optical radiation intensity.

DOI: 10.61011/TPL.2026.07.63826.20614

The development of autonomous power sources for nanoelectronics [1] and microelectromechanical systems [2] with low energy consumption and a long service life is a pressing issue in the energy sector of low-current systems. One possible solution is creation of long-term radioisotope power sources (RPS) that convert the energy of radioactive decay into electrical energy. Compared to traditional chemical batteries [3], RPS [4,5] can operate continuously for long periods of time depending on the half-life of the radioisotope, even at extreme temperatures. With a relatively low output power density, by recharging a battery (secondary chemical cell or capacitor), the RPS can provide pulsed output power in the range of milliwatts to watts [6]. This allows for long-term power supply of remote electronic systems such as diagnostic microchips and wireless communication systems capable of operating in space, seawater, and other aggressive environments. RPS can be used in implantable medical devices: pacemakers, defibrillators, intraocular implants [7]. A challenging task for such RPS is to increase their active lifetime and electrical power density at the output.

Two types of RPS are distinguished: providing direct and indirect conversion of radioactive energy into electrical energy. The operating principle of the first type of RPS is based on the effect of generation of electron-hole pairs by radiation (most often beta radiation) in semiconductor $p-n$ and $p-i-n$ structures, Schottky diodes [5,8]. The most commonly used sources of beta isotopes include nickel-63, gadolinium-148, and strontium-90, with their half-lives of up to 100 years. A disadvantage of power sources based on these isotopes is the presence of accompanying penetrating radiation, which requires additional radiation protection. Producing nickel-63 is a complex technical

task, which includes, in addition to long-term irradiation in a nuclear reactor, a time-consuming procedure of isotopic enrichment by gas centrifugation, followed by reduction of nickel isotope as a metal. The service life of such power supplies is limited primarily by the radiation resistance of semiconductor converters. Si, GaAs, InGaP, AlGaP, 4H-SiC, 2H-GaN, AlGaN, 2H-AlN, ^{12}C , $c\text{-BN}$ are used as a material for semiconductor beta-radiation converters [8]. The paper [9] reports for the first time the fabrication of a prototype beta-voltaic battery, comprising 200 vertically arranged subcells based on diamond Schottky diodes, with the addition of a low-enriched isotope (24 %) ^{63}Ni . An open circuit voltage of 1.02 V and a maximum electrical output power of $\sim 4 \mu\text{W/cm}^2$ are reported with a total source design volume of $5 \times 5 \times 3.5 \text{ mm}^3$.

The second type of RPS are sources where the energy of radioactive alpha or beta decay is converted into radioluminescent radiation, and then, with the help of semiconductor structures, into electrical energy [10,11]. In this case, accounting for the particle path length and properties of the radioluminescent material, the photoconverters are not exposed to direct radiation. Thus, they can have minimal dimensions, higher reliability, environmental safety and maximum service life. The RPS lifetime depends on the half-life of the used isotope and the spinodal decomposition time of the solid solution of A^3B^5 -photoconverter semiconductor [12,13]. Radioluminescent radiation sources can be made, for example, on the basis of zirconium orthosilicate (zircon) $\text{ZrSiO}_4:\text{Eu, Tb}, \dots /(^{238}\text{Pu})$, yttrium phosphate (xenotime) $\text{YPO}_4:\text{Eu}, \dots /(^{238}\text{Pu})$, activated by alpha-emitting radionuclides [10,11], or in the form of a phosphor in combination with beta-radioactive tritium [14]. Depending on the spectrum of optical radiation of the radiolumines-

cent source, semiconductor compounds A^3B^5 :Ga(Al)As, InGaP, and etc. can be used in photoconverters.

This paper outlines the study aimed at fabrication of a radioisotope power source for indirect conversion of radioactive energy into electrical energy. The source of optical radiation in this case is an artificial crystalline diamond enriched with ^{14}C isotope. The carbon isotope ^{14}C is a source of beta radiation with an energy of 156 keV and a half-life of more than 5700 years. Beta radiation in diamond leads to excitation of doping centers with subsequent luminescence in the optical range.

For the first time, a domestic technology for growing a diamond doped with nitrogen and boron and enriched with ^{14}C isotope was developed using $\text{Ba}^{14}\text{CO}_4$ precursor [15,16]. The growth was carried out using the temperature gradient method at high pressure and temperature (HPHT) in BARS setups (non-pressing apparatus „Split sphere“). The content of ^{14}C isotope was 0.008 wt.%. The luminescence of such a diamond is associated with the donor-acceptor transition of nitrogen–boron [17] dimer with radiation predominantly in the blue-green region of the spectrum.

Radiation spectrum of the studied diamond enriched with ^{14}C and doped with boron [18] is shown in Fig. 1 (curve 1). Measurements of the self-luminescence spectrum were carried out on a specially designed optical stand. To effectively convert the radioluminescent radiation of diamond into electricity, it is necessary to ensure optimal matching of uminescence spectra with the external quantum efficiency of the photoconverter (curve 2 in Fig. 1) with a minimum reflection coefficient (curve 3 in Fig. 1). In addition, it is important to ensure minimization of volumetric and lateral leakage currents, which are characterized by the saturation currents of the tunnel-trap (Esaki) and recombination (Saa, Noyce, Shockley) [19,20] transport mechanisms. For this reason, and in connection with the fact that the most intense radiation of self-glowing diamond is observed in the range of 450–550 nm, a photoconverter (PC) based on InGaP semiconductor was used. The photoconverter was grown using metal-organic chemical vapor deposition (MOCVD) method at Ioffe Physical-Technical Institute. The spectral characteristics of external quantum efficiency and the reflectance spectrum of the photoconverter were measured using MSD-1 monochromator and S100 spectrometer.

Fig. 2, *a* shows experimental direct dark current-voltage characteristic (VAC) of the photoconverter, which is described by a three-component exponential expression

$$J = J_{0t} \left(e^{\frac{qV_j}{A_t kT}} - 1 \right) + J_{0r} \left(e^{\frac{qV_j}{A_r kT}} - 1 \right) + J_{0d} \left(e^{\frac{qV_j}{A_d kT}} - 1 \right), \quad (1)$$

where $qV_j = (F_n - F_p)$ is the difference between the electron and hole Fermi quasi-levels at the boundaries of the spatial charge region of p – n junction, $V_j = V - JR_S$ is the resistive voltage, independent of the series resistance of R_S structure (V is the voltage across the entire p – n structure, J is the dark current density). The values J_{0t} ,

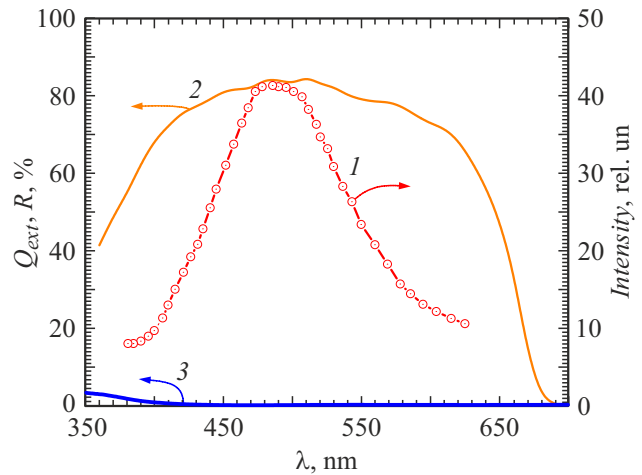


Figure 1. Experimental spectral dependences of the intensity of diamond luminescent radiation (*I*), external quantum efficiency (2) and reflectivity of InGaP photoconverter (3).

($A_t > 2$), J_{0r} ($A_r = 2$) and J_{0d} ($A_d = 1$) are the „saturation“ current densities and diode ideality factors corresponding to the tunnel-trap, recombination and diffusion mechanisms of the charge carrier transport in p – n junction.

VAC characteristics of the photoconverter were measured using MDC probe station and Keithley 2635A source-measure instrument. Depending on intensity level of optical radiation, the parameters of each component in expression (1) impact the efficiency of the conversion of optical radiation into electricity. The intensity of diamond self-luminescence (Fig. 1) according to Newport 843-R with a sensor 918D, was around $P_{opt} \sim 0.8 \text{ nW/cm}^2$. In this case, the main mechanisms of current flow that determine the efficiency of photoconversion into electricity in PC are the tunnel-trap with „saturation“ current J_{0t} ($A_t > 2$) and recombination with J_{0r} ($A_r = 2$). The values J_{0i} (A_i) were determined based on the experimental dark VAC (curves 1, 2 in Fig. 2) and made $J_{0t} = 10 \text{ pA/cm}^2$ ($A_t > 2$) and $J_{0r} = 10^{-2} \text{ pA/cm}^2$ ($A_r = 2$). The series resistance of PC was $R_S = 2.4 \text{ k}\Omega \cdot \text{cm}^2$.

In mathematical evaluations of the phase converter efficiency, the continuity and Poisson equations, the Shockley–Reed–Hall recombination model, and Fermi–Dirac statistics were used. The mobility of charge carriers was taken into account depending on concentration of nonequilibrium charge carriers, optical recombination, as well as a model of the band gap narrowing depending on the doping level.

Experimental and rated loading VAC are shown in Fig. 2, *a* (curves 3, 4), and the experimental value and rated dependence of efficiency on the photocurrent density — are given in Fig. 2, *b*. The short-circuit current density determined from the experimental and calculated curve is $J_{sc} = 273 \text{ pA/cm}^2$, and the no-load voltage is $U_{oc} = 0.172 \text{ V}$. The density of generated electrical power at the point of optimal load was $16.5 \cdot 10^{-12} \text{ W/cm}^2$.

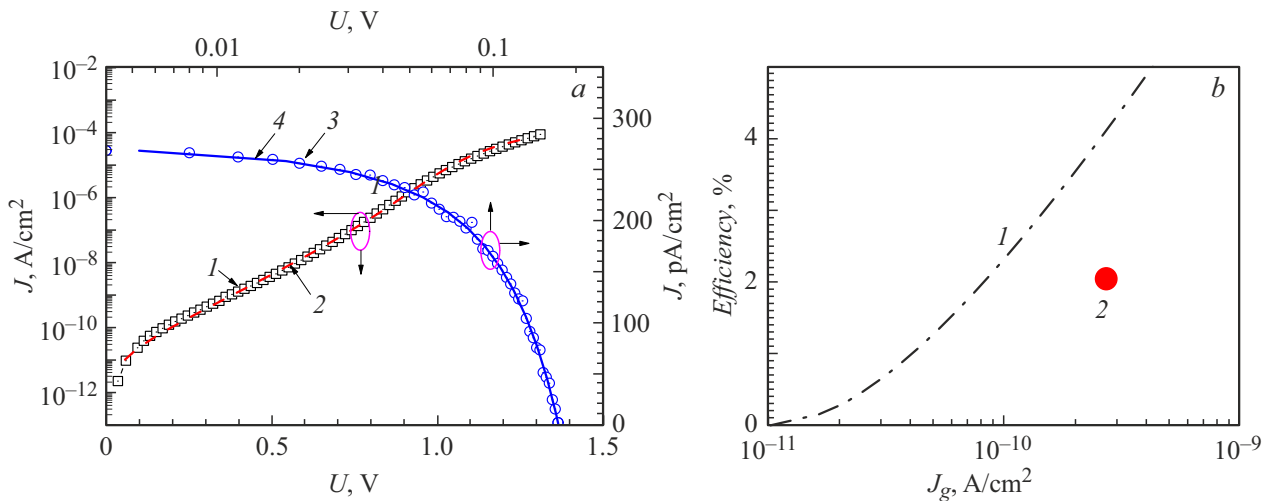


Figure 2. *a* — straight dark (1, 2) and loading (3, 4) VAC of InGaP-photo-converter. 1, 3 — experiment, 2, 4 — calculation of ($T = 300$ K). *b* — rated dependence of potential efficiency of InGaP-photoconverter on the generation current density (I) during excitation with monochromatic radiation ($\lambda = 490$ nm) and experimental efficiency of InGaP-photoconverter (2) when excited by a radioisotope source $^{12}\text{C}:(^{14}\text{C})$.

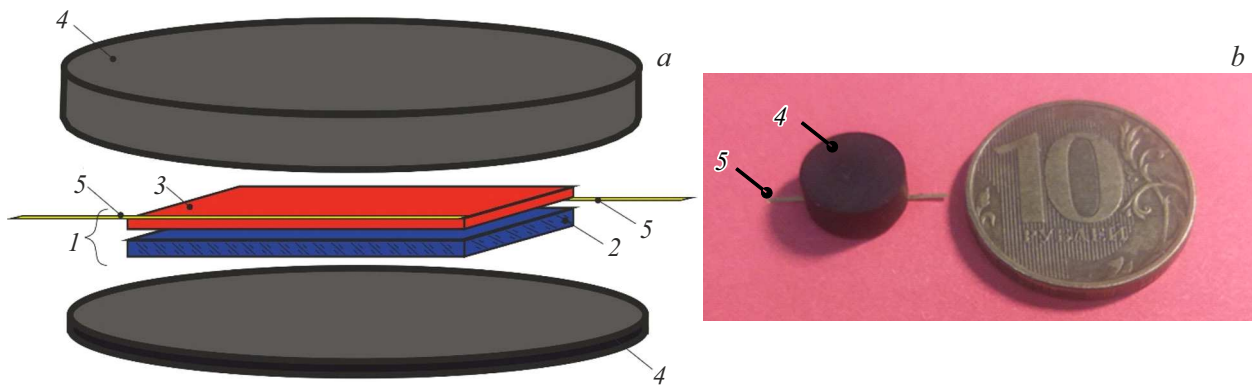


Figure 3. *a* — scheme of a radioisotope power source based on a self-glowing diamond $^{12}\text{C}:(^{14}\text{C})$ and InGaP photoconverter; *b* — photo of a radioisotope power source (RIP-C14) based on a self-glowing diamond crystal $^{12}\text{C}:(^{14}\text{C})$ and InGaP-photoconverter (in the center) and a 10-ruble coin (for comparison). The numbers indicate a power-generating part of source (1), which includes a self-glowing diamond (2) and a PC (3); the source body (4); electrical terminals on the PC (5).

Thus, for the first time, a model of a nuclear battery (Fig. 3) was created based on a self-glowing radioisotope source $^{12}\text{C}:(^{14}\text{C})$ with an optical radiation power density of $P_{opt} \sim 0.8 \text{ nW/cm}^2$ and PC. The „saturation“ current (10 pA/cm^2) of the photoconverter current flow tunnel-trap mechanism provided the no-load voltage of $U_{oc} = 0.172 \text{ V}$, output power of $16.5 \cdot 10^{-12} \text{ W/cm}^2$ and efficiency of 2%. For the obtained diamond self-luminescence intensity the rated efficiency of 4% and experimental efficiency of 2% (Fig. 2, *b*) were achieved in the study. To ensure that the calculated and experimental data are consistent, it is necessary to increase the efficiency of collecting radiation from a self-glowing diamond by optimizing its shape and dimensions, and to optimize the shape of the contact grid and design of the PC. It is necessary to reduce the influence of tunnel-trap and recombination transport mechanisms,

which have a significant impact on the efficiency of pico- and nanowatt optical signals. Since the half-life of ^{14}C isotope is more than 5700 years, and the content of the carbon isotope in it is negligible ($\sim 0.008 \text{ wt.}\%$), the developed nuclear battery is a safe source of electrical energy over a long period of time. In this case, it is necessary to account for the spinodal decomposition of the ternary solid solution of InGaP.

Calculations show that with a rise of optical radiation intensity by an order of magnitude $^{12}\text{C}:(^{14}\text{C})$, the efficiency of the photodiode will be $\sim 10\%$ with the possibility of providing an output electrical power density of $P_{max} = 7.2 \cdot 10^{-10} \text{ W/cm}^2$. A nuclear battery assembled, for example, from 200 such sub-elements connected in series [21] will provide a proportional increase in voltage with better radiation safety compared to other beta-voltaic

counterparts. Further research will be related to the development of technology for producing diamond with more efficient luminescence centers and increasing the efficiency of photovoltaic cells operating in photovoltaic mode.

Conflict of interest

The authors declare that they have no conflict of interest.

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Translated by T.Zorina