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GROWTH IN WORKS ON ISOTOPIC POWER
IN THE USSR

by

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13. ABSTRACT

This article gives data concerned with developing isotopic thermoelectric generators in the USSR to be used as the power supply of oceanographic and navigational devices, hydrographic stations, high-mountain stations dealing with cosmic rays, and other scientific research stations and devices of ground-based purpose. Scientific and technical bases for the creation of such power sources are examined, and characteristics of some models of the generators are given. Original article has one table and eight figures.

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Н н	*Н н*	N, n	Э э	*Э э*	E, e
О о	*О о*	O, o	Ю ю	*Ю ю*	Yu, yu
П п	*П п*	P, p	Я я	*Я я*	Ya, ya

* ye initially, after vowels, and after ъ, ь; e elsewhere. When written as ё in Russian, transliterate as yё or ё. The use of diacritical marks is preferred, but such marks may be omitted when expediency dictates.

All figures, graphs, tables, equations, etc. merged into this translation were extracted from the best quality copy available.

GROWTH IN WORKS ON ISOTOPIC POWER IN THE USSR

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Given in this report are data concerned with the creation in the USSR of isotopic thermoelectric generators intended for the power supply of oceanographic and navigational devices, hydrographic stations, automatic radiometeorological stations, magnetic variation stations, high-mountain stations of cosmic rays and other scientific research stations and devices of ground-based purpose.

Scientific-technical bases for the creation of such power sources are examined, and characteristics of some models of the generators are given.

Radioisotope Fuel

Today more than 1000 radioactive isotopes are known. In order to select the fuel for the isotopic thermoelectric generators correctly, a detailed study of their nuclear physical properties is necessary, in the first place, the rate of the change in the number of initial nuclei (burning rate) and content of energy released at one event of the decay. The most suitable as a radioactive fuel are isotopes with a half-life ($T_{1/2}$) within limits of 100 days-100 years (~50 isotopes), from which 12-15 isotopes can be obtained in large quantities.

The major advantage of α-radioactive isotopes is the high value of the energy release for the event of the decay, whereupon its basic part turns into the kinetic energy of the α-particle and the recoil nucleus whose path completely fits into the volume of the radioactive preparation and in walls of the shell of the isotopic element. For some radioactive isotopes (which are the parent materials of the chain of radioactive conversions - ^{227}Ac, ^{228}Th and ^{232}U) the total energy release for one event of the decay of the parent isotope is 30-40 MeV, which is commensurable with the fission energy of heavy nuclei. Also the fact that with α-decay the majority of the transitions occurs at the main level of the daughter isotope, and when there is considerable probability of transition to the excitation level, the excitation energy is small. With β-radioactive decay (as a rule, the mean energy for decay $\bar{E}_\beta$ < I MeV) there is an accompanying braking and γ- emission, but the β-radioactive isotopes are more available.

<u>Possible sources and methods for the obtaining of isotopes</u>. On the basis of radiation and physical properties of isotopes, and also the possible sources and methods for obtaining them, it is

possible to produce the selection of the isotopes most suitable as a fuel for the isotopic sources of electrical energy (see Table 1).

The bulk of the fragmentation radioisotopes, including the most widely utilized strontium-90, are obtained as a result of the treatment of the discharged radioactive solutions. To ease the problem of the isolation of radiochemical pure elements, including strontium-90, the method for group concentration based on the deposition of the calcium of oxalate is used. The use of the indicated method makes it possible to divide all the radioactive fission-product elements into two groups: one - containing strontium, yttrium, rare-earth elements and americium, and the other - containing cesium, ruthenium, zirconium, niobium and ballast impurities.

Most promising today is the extraction separation of alkali-earth elements with the isolation of pure strontium. In this case extracting agents are used - di-2-ethylhexachlortophosphoric acids in kerosene from a nitric acid medium and a solution of salicylaldoxym (SAO) [Translators's note: this compound not found] in tributylphosphate from an alkaline (sodium hydroxode or ammonia) medium.

When evaluating the selection of a radioactive isotope as a source of thermal energy its cost is of great significance, which is especially for isotopes which rapidly decompose. For the cost of the latter, for example, use of the express method for the extraction, at which the rate of treatment reduces the losses of production for decay is of decisive importance. Thus, for polonium-210 the express method of the extraction of it by distillation from enriched bismuth with repeated recycling of the bismuth can be applied.

Table 1. Comparative characteristics of some isotopes which can be used in thermoelectric generators.

Isotope	Half-life, years	Basic form of radiation	Total energy arriving at the event of decay, MeV	Density g/cm^3	Volumetric heat power, W/cm^3	Activity per unit of heat power, C/W	Power: γ-radiation R/W·m^2	Power: neutron radiation s/W	Basic method of obtaining
1	2	3	4	5	6	7	8	9	10
^{210}Po	0,378	α	5,401	9,32	1340,0	31,4	$1,75 \cdot 10^{-4}$	–	Irradiation of ^{209}Bi
^{227}Ac	21,7	α	34,332	–	–	4,94	1,0	–	Irradiation of ^{226}Ra
(equilib.) ^{228}Th	1,9	α	34,746	11,7	1995,0	4,875	3,0	–	Irradiation of ^{226}Ra
(equilib.) ^{232}U	73,6	α	40,174	19,05	95,0	4,2	3,56	–	Irradiation of ^{229}Pa
(equilib.) ^{238}Pu	86,4	α	5,59	16,0	9,25	30,3	$6,35 \cdot 10^{-4}$	$4,85 \cdot 10^{3}$	Irradiation of ^{237}Np
^{242}Cm	0,445	α	6,213	13,5	1652,0	27,6	$6,7 \cdot 10^{-4}$	$1,52 \cdot 10^{5}$	Irradiation of ^{241}Am
^{244}Cm	17,9	α	5,895	13,5	39,0	28,6	$3,24 \cdot 10^{-4}$	$3,76 \cdot 10^{6}$	Irradiation of ^{243}Am or heavy isotopes of plutonium
^{90}Sr	27,7	β	1,1	2,6	2,44	154,0	Braking	–	Fission products
^{137}Cs	29,68	β,γ	0,786	1,873	0,77	215,0	66,2	–	The same
^{144}Ce	0,78	β,γ	1,409	6,9	184,4	120,0	2,0	–	The same
^{147}Pm	2,62	β	0,062	–	–	2725,0	Braking	–	The same
^{60}Co	5,27	β,γ	2,607	8,71	152,0	65,0	84,5	–	Irradiation ^{59}Co
^{170}Tu	0,354	β	0,321	9,3	105,5	525,0	Braking	–	Irradiation ^{169}Tu
^{192}Ir	0,204	β,γ	1,1	22,5	1345,0	154,0	76,6	–	Irradiation ^{191}Ir

The obtaining of thermally stable compounds of polonium from the vapor phase in this case ensures the high degree of the safety of the production and operation of the generator.

Radioactive isotopes are used as a fuel in the form of the special preparations placed into a hermetically sealing shell. From the viewpoint of radiation safety they should be a closed source of thermal energy with an activity of up to several hundreds of kilocuries with a high degree of airtightness, and should possess mechanical, thermal and corrosion resistance.

Since the radioactive isotopes are potentially biologically dangerous, the fuel has special requirements. The radioactive preparation must be in the form of a solid, which is non-crumbling, practically indissolvable in sea and fresh water, not sublimating and not entering into reaction with air, water and the material of an ampoule of the substance, which also should have a high radiation and thermal resistance.

From the viewpoint of radiation characteristics, the preparation should contain the smallest possible quantity of admixed radioactive isotopes with a hard neutron and γ- radiation, and the stable isotopes, which enters into the composition of the chemical compound or being the carrier, should consist predominantly of elements with small (in the creation of fuel on the basis of α- radioactive isotopes) and with large Z (in the creation of fuel on the basis of β-radioactive isotopes). The latter requirement makes it possible to decrease the yield of the braking and neutron radiations of the isotopic unit. The fuel material should also possess sufficiently high thermal conductivity and not contain large quantities of admixed radioactive isotopes with a half-life differing significantly from the basic isotope.

The low thermal conductivity of the preparation gives rise to a considerable temperature difference inside the thermal element and to the possible disturbance of its heat resistance, the content of a considerable quantity of short-lived isotopes - to a considerable decrease in the initial power and of long-lived isotopes - to a reduction in the specific activity.

The requirements for the design and material of the ampoule proceed from the fact that the construction of the radioisotope element would ensure its completeness and safety not only under operating conditions but also in emergency situations, which can take place both during the operation and transportation and with storage (Fig. 1).

Fig. 1. Radioisotope elements on the basis of ^{90}Sr.

Today there are created elements with an activity of dozens and hundreds of kilocuries on the basis of ^{144}Ce (20,000 curies), ^{90}Sr (9000-100,000 curies), ^{137}Cs (50,000-150,000 curies), and also elements on the basis of ^{238}Pu, ^{210}Po, ^{242}Cm and ^{60}Co, which satisfy the enumerated requirements. The heat power of these elements lies in the range of 1-1000 W.

Energy Release in Isotopic (Thermal) Element

It is evident that the total energy of decay Q is added from the energy of α - particles E_α, the recoil nucleus $\frac{m_\alpha}{m_{яд}} \sum E_{\alpha_i}$ and (when there is a thin structure) the energy of the emitted γ - quanta. When β - decay and spontaneous fission occur, it is necessary to consider the energy of the fission products (Еп.д.), β - particles and γ - quanta:

$$Q = \left(1 + \frac{m_\alpha}{m_{яд}}\right) \sum_i E_{\alpha_i} n_{\alpha_i} + \sum_i \bar{E}_{\beta_i} n_{\beta_i} + \sum h\nu_i n_{\gamma_i} + E_{п.д.}$$

The calculation of the spatial distribution of absorbed energy in the isotopic element and the subsequent integration of this distribution in volume in general cannot be completed analytically and require the use of bulky methods of numerical intetration. However, in connection with the fact that the paths α, β -particles and the recoil nucleus in solid radioactive fuel are very small (fractions of a millimeter), in practice all their entire kinetic energy is absorbed in the fuel itself (the leakage with bremsstrahlung is $\leq$ 1%). The energy absorbed inside the isotopic element is equal to the total energy of radioactive decay, and the specific heat release with sufficient accuracy can be taken as being in volume.

The most difficult problem is the determination of the specific or full heat release from γ -radiation, since the path of γ -quanta is commensurable with dimensions of the isotopic element. For such isotopes as ^{137}Cs and ^{60}Co, this gives rise to a noticeable

leakage of the energy. The calculation of the distribution of the specific heat release which appears as a result of the interaction of γ -radiation with substance at different points of the thermal unit, is connected with the numerical integration by several variables. Such a calculation is made for the thermal element on ^{137}Cs in the form of the equivalent sphere.

The experimental check of the heat release was conducted with the help of calorimetric installations developed by us in conformity with the working conditions in a hot chamber. The obtained results were coordinated sufficiently well with the calculated.

Biological Protection

In the development and creation of isotopic sources of electrical energy which contain kilocurie quantities of radioactive fuel, the problem of the calculation of biological protection is very important. One of the features of such a calculation is the fact that the dimensions of the isotopic unit, on the one hand, are commensurable with the mean free path of γ-quanta in the material of fuel (in connection with which it is necessary to consider the effect of self-absorption in the source); on the other hand, its dimensions are much less than the distance at which the allowed value of the dose rate is limited. Therefore, in point approximation it is possible to assume that

$$P = \frac{Q}{R^2} F$$

Here $F = \sum K_{\gamma i} f_i e^{-\mu d} B(\mu d)$; in the case of bremsstrahlung

$F = \int \frac{I(E)}{E} K_j(E) f(E) e^{-\mu d} B(\mu d) dE$, where K_j - differential γ-constant; B — buildup factor; f - the self-absorption factor; μ - linear attenuation factor.

Energy Conversion Radioactive Decay

One of the important problems which must be solved when developing the isotopic energy sources is the conversion of energy of radioactive decay into electrical energy. In the range of power from one to several hundreds of watts the most acceptable method for conversion is thermoelectric, which combines high reliability, acceptable effectiveness and prolonged service life.

Thermo-emmission and machine methods can be used in the producing of generators of high power (kilowatt range); the development of atomic batteries is advantageous (in separate cases) in the producing of sources of electrical power supply in the micromilliwatt power range.

Today the thermoelectric method is the most developed: obtained are low-temperature semiconductor materials (up to 300°C), which possess sufficiently high effectiveness (5-8%), and mean temperature (300-700°C) and high-temperature materials (more than 700°C). The combination of different materials in the form of cascade elements now makes it possible in experimental models to achieve an effectiveness of conversion of 12-15%.

In the producing of isotopic thermoelectric generators there have been developed engineering methods of the calculation of single and multistage converters, and the radiation stability of thermoelectric materials in the field of neutron and γ-radiation has been studied. The high concentration of recombination centers gives rise to a considerable reduction in the lifetime of the electron-hole pairs. As a result of this the influence of the ionizing effect of γ-quanta on properties of thermoelectric materials ($\alpha, \sigma, \varkappa, z$) proves to be unimportant. The estimate of the quantity of defects which appear in the transit

of neutrons (10^6-10^7 neutrons/cm^2·s) for a long period of time (10^5 h) shows that even without allowing for the effect of annealing the quantity of the radiation defects is two or three orders less than admixed.

Selection of the Thermal Circuit and Calculation of Thermoelectric Generator

The isotopic thermoelectric generator is a system with an internal source of heat release, the temperature field in which is determined basically by processes of thermal conductivity or radiation inside the system and convective and radiant or contact heat exchange with the environment.

The isotopic thermoelectric generator (Fig. 2) is a heat source - radioisotopic unit 5, on part of the surface of which elements of the thermionic converter 3 or structural connections 4 are located; the remaining surface is surrounded by heat insulation 6. Heat transfers are connected with structural elements of the generator 2, which scatter the unconverted heat through the system of heat discharge 1 into the surrounding space.

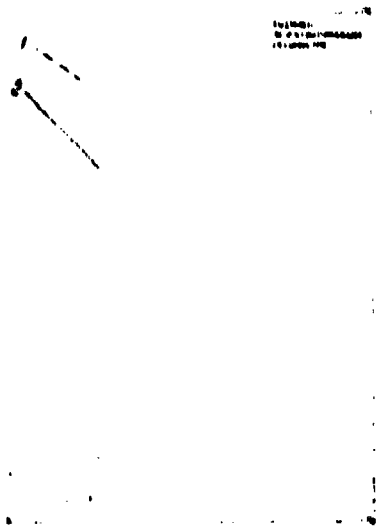

Fig. 2. Diagram of an isotopic thermoelectric generator.

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The basic problem in the selection and calculation of a thermal circuit is the provision for maximum heat flux to the thermionic converter and the producing on it of a temperature drop which corresponds to the optimum temperature range for the selected simiconductor material. Special attention is given to the producing of a uniform heat flux through elements of the thermionic converter and to a decrease in the parasitic thermal resistances on its path. The abundance of the factors which influence the thermophysical characteristics of the generator gives rise to the necessity for their detailed calculation and experimental investigation.

Isotopic Thermoelectric Generators
^{144}Ce, ^{137}Cs, ^{90}Sr, ^{238}Pu, and
$^{242}Cm(^{210}Po)$

From an analysis of properties of isotopes ^{144}Ce and ^{90}Sr, the degree of the development of technology and the production potentialities of the obtaining of these isotopes, it is possible to draw the conclusion about the advisability of the producing on their basis of isotopic thermoelectric source generators of the power supply of automatic radiometeorological stations (APMC) [ARMS] and other devices of ground-based purpose, whereupon generators based on ^{144}Ce can have a service life of 0.5-1 years and generators based on ^{90}Sr - 1-10 years.

From calculations of the radiation yield and heat release in an isotopic element, it is evident that the basic contribution in them is given by daughter isotopes (^{144}Pr and ^{90}Y), and selected as the chemical form of fuel are $CeMoO_4$ and $SrTiO_4$. The thermal converter is made from low-temperature alloys on the basis of $Bi_2Te_3 + Bi_2Se_3$ and $Bi_2Se_3 + Sb_2Te_3$, which possess

the best thermoelectric properties in the range of temperatures of 200-600°K at very high reliability. The system of the switching of elements is fulfilled in the form of a single battery, which substantially simplifies the technology of assembly of the generator and replacement of the battery. The heat insulation is completed in the form of a system of screens, which ensure the minimum heat losses and make it possible to use a sufficiently simple circuit of the control of the heat discharge when using relative short-lived ^{144}Ce isotopes (Fig. 3).

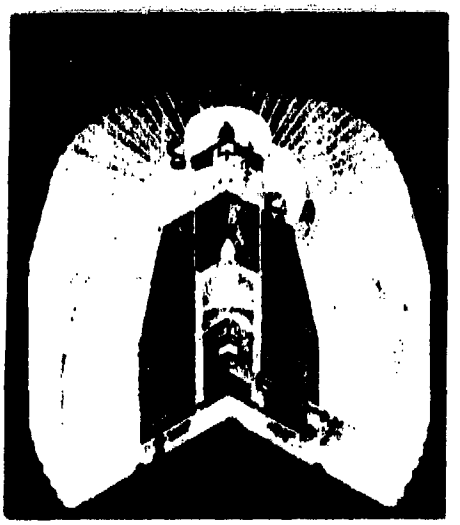

Fig. 3. Generator "Beta-1" on the basis of ^{144}Ce.

On the basis of ^{90}Sr, during 1963-1970 a series of isotopic thermoelectric generators were developed: "Beta-2", "Beta-3", "Beta-C" (Fig. 4) for the power supply of various types of ARMS, the installations "Ether" and "Penguin" for the power supply of light radio beacons and magnetic variation stations (Figs. 5 and 6), and the dual-purpose installation "Angara" for the supply of thermal and electrical energy of a high-mountain station of cosmic rays.

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Fig. 4. Generator "Beta-C" on the basis of ^{90}Sr.

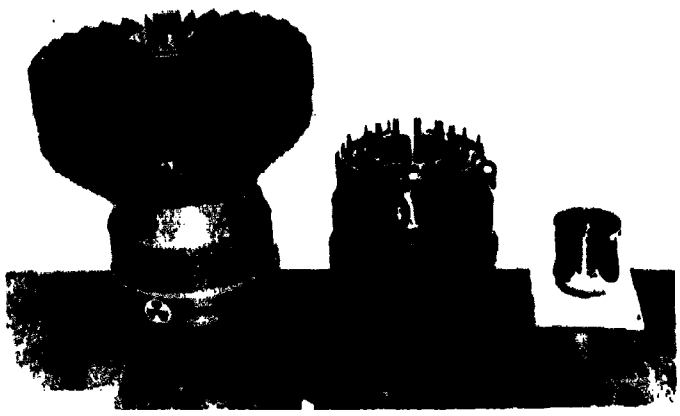

Fig. 5. Generator "Penguin" on the basis of ^{90}Sr.

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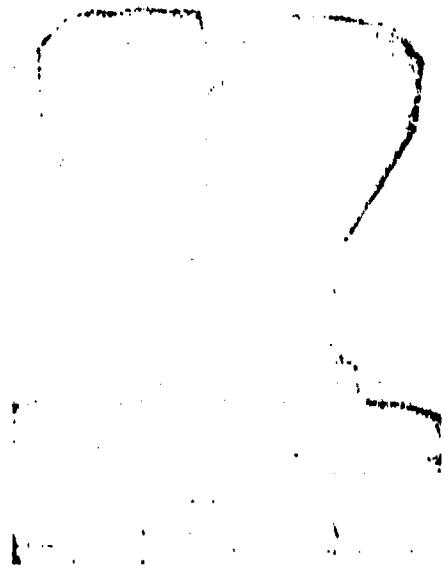

Fig. 6. Generator "Ether" on the basis of ^{90}Sr.

The selection of ^{137}Cs for the producing of sources of electrical power at various hydrographic stations is conditioned primarily by the presence of its industrial production in large quantities, the sufficiently long half-life, and the possibility in operating conditions of using sea water as a biological protection. Among the deficiencies of ^{137}Cs it is possible to note the presence of associating γ-radiation (^{137}Cs - 660 keV, ^{134}Cs - 1.367 MeV) and the relatively low value of the specific heat release (0.123 W/cm^3). The working form of the fuel in the installations of the purpose indicated above is accepted in the form of cesium-lead-silicate glass as being a less soluble compound of cesium.

The distinctive feature of the fuel on the basis of ^{137}Cs is that 70% of energy of radioactive decay is included in the kinetic energy of γ-quanta, which caused the necessity for the introduction of a massive thermal element. The location of this element inside the effective heat insulation makes it possible to create the directed heat flux in the direction of the thermionic converter and raise its density 5-6 times.

The development of isotopic sources of electrical energy on the basis of ^{238}Pu was started in connection with the necessity for producing portable power sources of different purpose.

Requirements of minimum weight and overall dimensions and also the low background of the associating neutron and γ-radiation lead to the necessities for the use of isotopic fuel on the basis of α-radioactive isotopes. The most fully indicated requirements are satisfied by ^{238}Pu, which is actually a single α-radioactive isotope with sufficiently large $T_{1/2}$ (86.4 years) and low level of the associating radiation.

The producing of portable generators of the MIG-67 type (Fig. 7) on the basis of ^{238}Pu required the solution of a number of the problems both in the region of the producing of an isotopic element and in the region of a micromodule thermionic converter. Limited dimensions of the generator substantially affect the heat losses through the structural elements: with small lengths of strong couplings the heat losses through them become commensurable with losses through the heat insulation, and therefore preference is given to the location of the isotopic (thermal) element of low power either directly on the thermopile or between two thermopiles.

The unique properties of ^{242}Cm and ^{210}Po (high specific energy release, sufficiently low intensity of γ-radiation), and also the potential possibility of obtaining them in considerable quantities open up wide prospects for the use of ^{242}Cm and ^{210}Po for producing sources of electrical energy with high specific power. The development of isotopic generators on radioactive fuel with a high specific energy capacity made it possible to create isotopic thermoelectric generator on cascade converters with an efficiency equal to 8-10% in the range of temperatures of 300-850°K (Fig. 8).

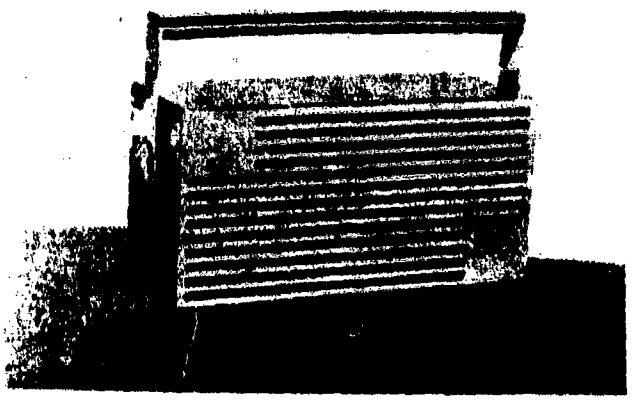

Fig. 7. Generator MIG-67 on the basis of ^{238}Pu.

Fig. 8. Generator using cascade converters.

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The positive experience of the development and operation of isotopic generators in different regions of the country and increasing possibilities of the obtaining of radioactive isotopes in large quantities make it possible to hope that isotopic power in the USSR will obtain further extensive development.