Nickel-63 (a pure beta-emitter with a half-life of 100 years) is one of the most promising radionuclides that can be used in miniature autonomous electric power sources with a service life of above 30 years (nuclear batteries) working on the betavoltaic effect. This effect is analogous to the photoelectric effect, with the difference that electron-hole pairs are produced in a semiconductor with p-n-transition under the action of beta-particles rather than optical radiation.

In addition to $^{63}$Ni, among all variety of radionuclides only tritium $^3$H (half-life 12.3 years; $E_{\text{max}} = 18.6$ keV; $E_{\text{av}} = 5.7$ keV) and promethium $^{147}$Pm (half-life 2.62 years; $E_{\text{max}} = 230$ keV; $E_{\text{av}} = 65$ keV) can be considered as candidates for the betavoltaic converter. In this case the service life will be 5-10 years for the tritium battery and even shorter for the promethium battery. What is more, the maximum energy of the promethium beta-particles ($E_{\text{max}} = 230$ keV) exceeds the threshold of radiation damage for semiconductor converters, which is undesirable.

All other beta-emitters ($^{85}$K, $^{90}$Sr and many others) are unsuitable for any of several reasons:
- accompanying gamma-radiation;
- strong bremsstrahlung, which requires the use of radiation protection;
- the average beta-spectrum energy considerably exceeding the threshold of radiation damage for semiconductor converters, which can reduce the service life of nuclear batteries down to a few months (for silicon $E_{\text{th}} = 145$ keV; for gallium arsenide $E_{\text{th}} = 170$ keV).

Pure alpha-emitters, such as plutonium-238, are unsuitable for betavoltaic conversion because of fast failure of the p-n junction in semiconductors under
the action of alpha-particles. Although $^{238}$Pu, as a source of energy, has unique energetic characteristics (specific energy 0.55 W/g; $T_{1/2} = 87.6$ years), its use in autonomous sources of energy is reasonable where the thermal cycle (thermoelectricity, thermionic emission, thermophotovoltaic converters, etc.) is used to convert the energy of radioactive decay into electricity.

The maximum energy of beta-particles in the $^{63}$Ni emission spectrum is 65 keV, which is much lower than the threshold of radiation damage in the semiconductors intended for use – silicon and gallium arsenide. This must assure the stability of the electric parameters of nickel nuclear batteries throughout the whole service life (at least 30 years).

The average energy of the $^{63}$Ni beta-spectrum is so small ($E_{av} = 17$ keV) that neither bremsstrahlung, nor beta radiation goes beyond the battery case. The absence of dosimetric problems enables the use of batteries of this kind for a wide variety of implantable biostimulators as well as in many other areas of technology (microelectronics, space engineering, telecommunications, environmental monitoring, etc.).

The use of $^{63}$Ni is hindered by the small volume of its production and the high cost of the product. Nickel-63 is produced by neutron irradiation of targets containing nickel-62 as a starting isotope. To increase the specific activity (per unit mass) the targets are enriched as much as possible in nickel-62 and irradiated in ultrahigh-flux nuclear reactors. The centrifugal technology used presently in Russia for separation of stable isotopes makes it possible to prepare enriched isotopes of more than two tens chemical elements [1, 2] including nickel [3]. The volatile compound, nickel tetrafluorophosphine $\text{Ni}(\text{PF}_3)_4$, is used as a working substance in separating the nickel isotopes [4]. The nuclear-physical characteristics of nickel-62 and nickel-63 [6] do not allow one to obtain the nickel-63 content in the in-reactor irradiated target more than 30% of the initial amount of starting nickel-62. The real economic reasons restrict this figure to 15-17%. Eventually, we have the not very efficient, but expensive, process of irradiating the very expensive target. This process is a-priori incapable of producing the highly-enriched nickel-63 at reasonable cost.

The problem can be solved if the target nickel is charged into the channels of industrial and/or power reactors of the RBMK and ADE type with medium-level neutron fluxes of $2\times10^{13} - 2\times10^{14}$. The inexpensive use of these reactors in comparison with ultrahigh-flux neutron sources allows a long-term campaign with a charge of many kilograms of nickel enriched in nickel-62 (but not considered as "highly-enriched"). The centrifugal technology makes possible large-scale separation of nickel isotopes similar to production of depleted zink to be added to the coolant of power reactors [5]. The post-irradiation enrichment of the irradiated target in nickel-63 with the use the centrifugal separation technology provides the basis for the production of nickel-63 in desired amounts with a required concentration.

The characteristics of the sequence of the basic technologies: fabrication of a nickel-62-enriched target + irradiation in a nuclear reactor + enrichment of nickel-63 have been calculated. The calculations show a possibility to solve the above problem. The isotopic concentrations of products in one variant of
Table.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>58</th>
<th>60</th>
<th>61</th>
<th>62</th>
<th>64</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal neutron capture cross-section, barn</td>
<td>4.6</td>
<td>2.9</td>
<td>2.5</td>
<td>14.5</td>
<td>1.52</td>
</tr>
<tr>
<td>Resonance integral, barn</td>
<td>2.2</td>
<td>1.5</td>
<td>1.5</td>
<td>6.6</td>
<td>0.98</td>
</tr>
</tbody>
</table>

The process are presented in the figure.

The target nickel containing mainly nickel-62, but not highly-enriched, is produced in the course of the first centrifugal separation; in this case the lightest isotope (nickel-58) content is minimized. This is rather essential, because nickel-58 has the great "harmful" neutron capture cross-section (see Table and [6]) and transforms via electron capture to stable Co-59. In turn Co-59, as a starting isotope, produces Co-60 which is the gamma-emitting pollutant in the target.

The exposure of a target having a nickel-62 content at a level of 80% in a reactor with a neutron flux of \(10^{14}\) \(\text{nxcm}^{-2}\text{s}^{-1}\), for example during two years, makes it possible to build up above 6% nickel-63. It should be noted that nickel-63 has an appreciable neutron capture cross-section (24 barns), which is why nickel-63 is burned in the reactor producing nickel-64. This process
limits the maximum attainable specific activity of nickel-63. Because of burning, the nickel-64 content of the irradiated product is higher than in the initial target. The small amount of nickel-58 in the target leads to the production of nickel-59 in amounts of only small fraction of percent. As for cobalt-60, some fast neutron induced reactions of target isotopes are responsible for its formation in addition to the chain considered above. Prior to the process of enrichment in nickel-63 the thorough radiochemical cleaning of the irradiated nickel targets is required to prevent the second centrifugal cascade from contamination with cobalt-60. Then the volatile compound must be synthesized from nickel and directed to centrifuging. As a result of centrifuging, a product with a nickel-63 content of 80% and higher can be obtained in the useful fraction. The degree of enrichment in nickel-63 and the degree of its recovery are determined by the relation between expenditures on enrichment and irradiation of the target.

Another, light, fraction consists at the most of nickel-62 and contains some residual amount of nickel-63. After transforming the light fraction to metal this material is directed again to irradiation. By this means no radioactive waste is accumulated in the given production scheme.

The implementation of large-scale nickel-63 production based on the scheme considered will make it possible to create a radionuclide source of electric energy of milliwatt range with a service life of 30-50 years. This is a basis for operation of various autonomous devices which cannot be realized without development of microelectronics, for example, in the area of long-term environmental monitoring.

References