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RADIATION POLLUTION OF ATMOSPHERE DURING METSAMOR NUCLEAR POWER STATION OPERATION AND RADIATION MONITORING ON THE TERRITORY OF AZERBAIJAN REPUBLIC BY USING DRONES

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Abstract: There have been presented in paper the results of analysis of emission of the short half-life gaseous isotopes from out Metsamor Nuclear Power Station and of the process of spread of long half-life isotopes ($T\frac{1}{2} > 10$ hours) on the territory of Azerbaijan Republic. There had been developed a method of application of drones for radioprospecting on the territory of Azerbaijan Republic and offered to use drones in Chemical Troops of Armed Forces.

Keywords: Metsamor nuclear power station, nuclear reactor, radioactive waste, radionuclide, drone, radiation monitoring.

1. Introduction

One of the major aspects of nuclear safety is ecological security. Currently, "Metsamor" NPS is regarded to be a poisonous infrastructure for region's ecology. The NPS significantly lags behind in terms of nuclear safety and one of the factors that increase the risk for the region is its non-persistence. According to the estimations of radioactive pollution, severe accident that can occur in "Metsamor" NPS will lead to radioactive pollution in regional states under any weather conditions. Radioactive substances are released to the environment during the daily operation of the reactor and as a result, Kura and Araz rivers are polluted. Radionuclides enter the territory of Azerbaijan in a short time through the surface and underground water. Therefore, the environment exposes to radioactive pollution [1].

National leader Haydar Aliyev emphasized this issue during the visit of the former secretary general of International Atomic Energy Agency (IAEA), Mohammad Al Baradei in 2002 and noted that, "Metsamor" NPS must cease its activity because of the end of its operational life and the damage to environment. At the same time, he asked for taking into account of this issue in the IAEA. But, the life of reactor is extended by 12-13 years as a result of certain support to Armenia.

Emission of the gaseous isotopes from out nuclear reactor and its characteristics

Any NPS in operation have a strong influence on the environment, and form radioactive wastes (RW). There are three main types of current nuclear reactors: the reactor working on natural uranium graphite-gas scheme, enriched uranium - heavy water scheme and enriched uranium-boiling water scheme. "Metsamor" NPS works on enriched uranium - heavy water scheme, and these type reactors (WWER–440) significantly pollute the environment [2]. Although reactors work on close-cycle, sometimes the density of coating is violated. Moreover, it is necessary to change the water in the first cycle of the reactors. Radioactive isotopes of rare elements and ¹³¹I are thrown to the atmosphere and ³H to the rivers taking into account the issuable parts. ³H, which is the radioactive isotope of hydrogen, is formed in the third stage of reaction [3].

The reactor (WWER–440) operating in "Metsamor" NPS is high pressurized water reactor and the thermal energy, released from heat-producing elements in active zone, is given to water, which is under high pressure in the first cycle, in this reactor [4]. 160 Pa high pressure is formed in order to prevent boiling of water in the first cycle at the temperature above 300^oC. Water energy in the first cycle is converted into water in the second cycle, as well as the water in the second cycle into vapour at 330^oC temperature through heat exchanger and it makes turbine move in 12-60 Pa. The water in the first cycle is functioning as heat carrier and neutron moderator. Water – water energetic reactors are the most used reactors in the world (more than half of the industrial reactors operating in the world) [5].

The total activity of fission materials increases by million times in the working process of this reactor. As ¹³¹I and ⁸⁵Kr separate, they have high activity. As half-life of ⁸⁵Kr is 10 years, it accumulates in the atmosphere. Total activity of fuel, enriched 3% by ²³⁵U, is $6 \cdot 10^{11}$ Bq (16 Ku) in the complete filling of the reactor (it is uranium more than 30 tones). The activity of the products, formed in the nuclear fission process, will be $4 \cdot 10^{19}$ Bq (10⁹ Ku) after one year use. 10^{19} Bq (300 mln. Ku) fission products will be formed in NPS during the production of 1GW/year electric energy [6].

There are formed nearly 300 different radionuclides from uranium fuel by nuclear fission in "Metsamor" NPS, and more than 30 of them can enter the atmosphere: ¹²⁹I ($T_{1/2} = 16$ mln. years), ¹⁴C ($T_{1/2} = 5730$ years), ¹³⁷Cs ($T_{1/2} = 30$ years), ³H ($T_{1/2} = 12,3$ years), ⁸⁵Kr ($T_{1/2} = 10,76$ years), ¹³¹I ($T_{1/2} = 8,05$ days), ¹³³Xe ($T_{1/2} = 5,27$ days), ¹³³I ($T_{1/2} = 20,3$ days), ⁴¹Ar ($T_{1/2} = 1,83$ hours), ⁸⁷Kr ($T_{1/2} = 76$ min.), ¹³⁸Xe ($T_{1/2} = 17,5$ min.), ¹⁶N ($T_{1/2} = 7,35$ sec.).

There are three types of radioactive wastes during the operation of NPS: solid, liquid, gaseous. WWER-440 type reactors form 40000 Ku gaseous radioactive wastes during a year. Most of them are captured by filters or they easily decay by losing their radioactivity. Most parts of radioactivity of gaseous wastes decay within a few hours and days by generating with short half-life radionuclides and without damaging the environment. Besides the usual gaseous wastes, a few radionuclides - corroded reactor products as well as nuclear fission fragments of uranium - ⁵¹Cr, ⁵⁴Mg, ⁶⁰Co, ⁹⁵Nb, ¹⁰⁶Ru, ¹⁴⁴Ce, etc. are sometimes thrown to atmosphere from NPS.

The source of radioactivity in reactors is considered induced (formed under the neutron influence) radioactivity. The main products of the activity of heat carriers are Ne, Ar, F, Cl, Na, K, Mn, Co, Fe, Cu, Zn, Ag, ³H, C, etc. elements.

Heat carrier of the first cycle (contour) always keeps some expanded gases during the use of reactor. The source of these gases is: water radiolysis; corrosion of constructive materials; gas compensators of the capacity; feeding heat carriers; irregular heat-dissipating elements; chemical additives for the maintenance of water regime; nuclear reactions; radiolysis and thermoradiolysis of ionites of purification system of the first cycle.

Water molecules and mixtures, which are solved in it, decompose, are excited and ionized under the different ionizing radiation influence. There occurs water radiolysis and H, OH, H₂, H₂O, O₂, HO₂ are formed as a result of it. Most of them have oxidative or recovery properties depending on mixtures in heat carriers, temperature and pressure in the first cycle, etc.

There occurs a few gases in the working process of nuclear reactor – nuclear reaction products: ${}^{14}C$ – in the activation of oxygen and nitrogen which are in the fuel as a mixture, *during* triple fission in fuel; ${}^{3}H$ – during triple fission and through the activation of Li, B, D, which are in the heat carrier as a mixture; ${}^{18}F$, ${}^{20}F$ – in the activation of nuclear of water and sodium; ${}^{23}Ne$ – in the activation of nuclear of sodium; ${}^{41}Ar$ - in the activation of ${}^{40}K$ in sodium and ${}^{40}Ar$ in water and air.

The character of gaseous radioactive wastes depends on the type of reactor and behavior system with these wastes. In radioactive inert (ineffective) gases (RIG) (Kr, Xe radionuclides), gaseous forms of ³H and ³H steam, ⁴¹Ar, ¹⁴C, ¹³N, ¹⁶N activating gases, haloids, and solid radioactive substances (fission and activation products).

Their total volume is 25 cm³ for per 1 MW within a day (at normal pressure and room temperature). They fall to heat carrier during the violation of hermetization of heat dissipating elements. Reactive inert gases (RIG) can enter the environment from reactor through the percolation of water from the first cycle (contour). Their total activity is 0,3 TBq/MW during a year (table 2 and 3), ¹³³Xe occupies the main place in the activity of these wastes (table 4).

T	able	1

				mun n		or gase	ous nuc	nues				
The name of nuclides	³ H	¹⁴ C	⁴¹ Ar	^{83m} Kr	⁸⁵ Kr	^{85m} Kr	⁸⁷ Kr	⁸⁸ Kr	⁸⁹ Kr	¹³³ Xe	^{133m} Xe	¹³⁵ Xe
T ¹ /2	12,26 years	5730 years	1,83 hours	1,86 hours	10,76 years	4,4 hours	76 min.	2,8 hours	3,18 min.	5,27 days	2,3 days	9,14 hours

Half-life time of gaseous nuclides

The name of nuclides	^{135m} Xe	¹³⁷ Xe	¹³⁸ Xe	¹³⁹ Xe	¹⁴⁰ Xe	¹³¹ I	¹³² I	¹³³ I	¹³⁴ I	¹³⁵ I
T ¹ ⁄2	15,6 min.	3,9 min.	17,5 min.	43 sec.	16 sec.	8,05 days	2,26 hours	20,3 days	52 min.	6,68 hours

Table 2

Emission of radiation gaseous nuclides in atmosphere

The type of reactor	Activity, MBq/MW per year					
The type of reactor	RIG, TBq	iodine	Aerosols (without iodine)			
WWER-440	0,11 - 0,28	0,74 – 9,25	0,37 – 11			

Table 3

The average amount of radioactive gaseous wastes thrown from reactor. (GBq of per produced electrical energy GW)

The type of reactor	RIG*	¹³¹ I	LHR**	³ H
WWER-440	220	1,7	4,5	5,9

* Radioactive inert gases

** Long half-life radionuclides

Formation scale of ⁸⁵Kr is larger by a few levels than the other radionuclides - 375 Ku-MW during a year. The amount of ⁸⁵Kr increases in atmosphere year after year. Currently, ⁸⁵Kr composition in the atmosphere is more 1 000 000 times than the period up to the beginning of the nuclear era.

Table 4

	Relative composition, %
⁴¹ Ar	0,2
⁸⁵ Kr	6
^{85m} Kr	5,4
⁸⁷ Kr	1
⁸⁸ Kr	2,2
¹³³ Xe	72
^{133m} Xe	_
135Xe	13,2
^{135m} Xe	_
The other isotopes of Xe	_
Total RIG	100
¹³¹ I	58,9
¹³³ I	31,9
¹³⁵ I	8,3
Total	100

The amount of RIG and iodine in gaseous wastes in "Metsamor" NPS

 14 C is generated in nuclear reactor as a result of 14 N(n, p) 14 C reaction (activation of nitrogen in fuel as a mixture), as well as during triple fission: 17 O(n, α) 14 C (during activation, in oxidized fuel and heat moderator) and 13 C(n, γ) 14 C, which are considered practically more important than others.

 14 C accumulates in the biosphere in large quantities by replacing carbon in organic compound. Carbon turns to nitrogen during the fission and organic molecules decompose. It is considered that, the accumulation of 14 C is the reason for slower growth of trees. Currently, the amount of 14 C in the atmosphere has increased by 25% than the period up to nuclear era.

Tritium ³H exists in gaseous state and oxidative form of H^3HO and ³H₂O, as well as, it can be component of more complex organic and inorganic compounds. Nearly 0,37–0,74 GBq/MW (electric power of reactor) ³H are formed in heat-dissipating elements in the account of triple fission in a reactor during a year. ³H turns to He when it decomposes and it emits β -rays. This transmutation is extremely dangerous for the living organisms, so the genetic apparatus of cells are damaged in this case.

Some radionuclides of iodine are generated during fission reaction, as well as decay of fission products. More radiation doses are ¹²⁹I, ¹³¹I, ¹³²I, ¹³³I, ¹³⁴I and ¹³⁵I radionuclides. Radionuclides of iodine can enter the environment from the coating, which hermetization is violated, of heat-dissipating elements and the first cycle. Nuclide of iodine ¹³¹I is considered watering source. Long half-life ¹²⁹I is not determined in the environment around NPS and its wastes are less than the wastes of radionuclides of other iodine. ¹³¹I exists in either aerosol or gaseous state. Average wastes of ¹³¹I from reactor are 74–185 and 1,85–22,2 MBq/MW (electric power of reactor). Radioactive iodine leads to hormonal changes and obesity in humans.

One part of fission products, fission products of radioactive inert gases and nuclides with induced activity form aerosols and as a result, they enter the environment through airflow. Tens of nuclides in the composition of aerosols thrown from reactor are identified and ¹³¹I, ⁸⁹Sr, ⁹⁰Sr, ⁹¹Sr, ¹⁰³Ru, ¹³⁷Cs, ¹⁴¹Ce, ¹⁴⁴Ce, ⁵¹Cr, ⁵⁴Mn, ⁵⁵Mn, ⁵⁹Fe, ⁵⁸Co, ⁶⁰Co, ⁹⁵Zr, ^{110m}Ag, ²²Na, ²⁴Na, ⁸⁸Rb, ⁹⁹Mo are such type nuclides.

Total composition of solid fraction of wastes thrown from reactor is approximately 2,2 MBq/GW (electrical power of reactor) within a year.

Spreading long half-life radionuclides on the territory of Azerbaijan Republic

Let's consider the spread of gaseous radionuclides released to the eastern part of the territory of Azerbaijan Republic and Nakhchivan Autonomous Republic during the operation of reactor in "Metsamor" NSP. The atmosphere convection is equal 2 m/sec airflow. Let's consider the spreading process of long half-life ($T\frac{1}{2} > 10$ hour) radionuclides entering the territory of Azerbaijan Republic.

The distance from "Metsamor" NPS to Nakhchivan Autonomous Republic is $60\div75$ km, but to Absheron peninsula is ≈ 600 km. Then, long half-life radionuclides will reach to the territory of NAR with atmospheric convection airflow within ≈ 10 hours, but to the eastern part (Baku) of Azerbaijan during 84 hours or 3.5 days.

The average amount of gaseous radionuclides thrown from the reactor is 4,5 GBq for per 1 GW electric energy of NPS for long half-life radionuclides (table 3).

Radionuclides spread on the territory of NAR: ¹²⁹I, ¹⁴C, ¹³⁷Cs, ³H, ⁸⁵Kr, ¹³¹I, ¹³³Xe, ¹³⁵Xe, ¹³³Xe, ¹³³Mze, ¹³⁵I.

Radionuclides spread on the territory of eastern part of Azerbaijan Republic: ¹²⁹I, ¹⁴C, ¹³⁷Cs, ³H, ⁸⁵Kr, ¹³¹I, ¹³³Xe, ¹³³Xe, ¹³³Mze, ¹³³I.

So, the conducted analyses show that, above-mentioned radionuclides constantly enter the environment and spread along the direction of the wind on the territory of Azerbaijan during the use of reactor in "Metsamor" NPS.

Drones

As the problems related to Nuclear Power Stations (NPS) in various countries of the world are a great importance among the increasing ecological threats to humanity, weak safety regulation of "Metsamor" NPS is considered to be one of the main factors that increase the threats for region. It makes unavoidable the spread of radioactive sediments, caused by the accident that can occur in "Metsamor" NPS, on the territory of Azerbaijan Republic [2]. Therefore, it makes necessary to use not only dosimetric devices operating in a stationary position, but also drones on a large scale and in continuous mode in the areas bordering with Armenia. For this purpose, in the article, it has been developed the method of radiation monitoring through drones by Chemical Troops of Armed Forces on the territory of Azerbaijan Republic.

Currently, the tendency is observed in the field of improvement of the forces involved to eliminate consequences for the heaviness of results and the possibility of occurrence of accident in technogenic objects, including NPS. Mainly it is related to radiation accidents that may less likely to occur and many financial, human and other resources are needed during the accident. The standard scheme of the existence of large forces is considered non-optimal for the study of radiation condition in these cases and especially for the elimination of its consequence in the initial phase of the accident. Therefore, the application of highly effective radiation monitoring devices and specially trained staff is more appropriate.

In the modern life, the preparation of the devices for radiation monitoring is objectively needed in the areas that radiation accidents occur. It is needed to put the radiation monitoring devices that enable to carry out spectrometric and dosimetric measurements in drones in high radiation area, too. It allows carrying out remote aero-gamma-spectrometric shooting:

- working opportunity of the operator in the high radiation area without being exposed to radiation;
- the opportunity of examination of large areas in a short time;

- the examination in impassible areas, as well as, the determination of the parameters of radioactive wastes stream.

MD4-1000 type drone is prepared on "quadrocopter" scheme and defined to do wider-scale tasks. There are remote control and autonomous flying high technological platform on it [7]. Drone consists of comprehensive navigation system based on GPS and it allows to carry loads up to 1.2 kg.

Manually management of drone is carried out through the remote control panel. Surface base station is used in order to obtain telemetric data transmitted from drones. Received signals are decoded and prepared for the use as working platform in usual Notebook.

It has been used hanging spectrometric and dosimetric equipment for conducting radiation measurements. There are a spectrometer and two Geiger-Müller counter based on 31×31 mm sized NaI(Tl) crystal inside the equipment corps. The evaluation of dose power is carried out on the basis of spectrometric data in the range from 10^{-7} Sv/hour to 10^{-4} Sv/hour of radiation, but the measurements are carried out by two Geiger-Müller counter in the range of $10^{-4} \div 10^{-1}$ Sv/hour.

Telemetric data are transmitted to surface base station from drone (pic. 1).



Pic. 1. Management of drone

The development of the data after the flight includes the following:

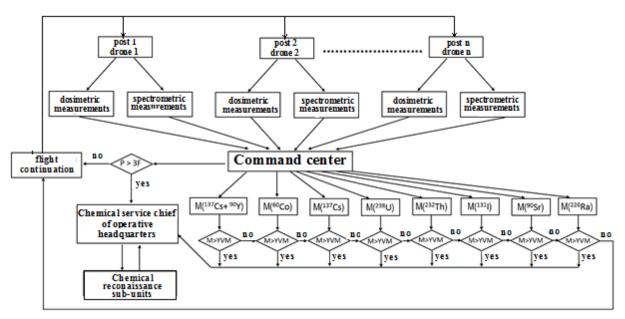
- calculation of spectra from the memory of measuring block;
- preparation of the sum of spectrum according to its measuring points;
- determination of average dose power in the group of measuring points on spectrum;
- determination and deduction of background of natural radionuclides;
- determination algorithm of the density of surface pollution;
- search algorithm of the state of the point source and determination of its activity.

Radiation monitoring method by using drones

In this section, it is explained the method of radiation monitoring carried out through drones by Chemical Troops of Armed Forces on the territory of Azerbaijan Republic.

The analysis of threats of "Metsamor" NPS for the region is fully explained in the previous articles and the formulas, obtained during the investigations, allow the evaluation of radiation dose, taking into account the direction and speed of air flow, as well as landscape of the area and the distance from the center of "Metsamor" NPS on the territory of Azerbaijan Republic, as well as Nakhchivan Autonomous Republic [8].

Stationary radiation control posts of Ministry of Ecology and Natural Resources are functioning in different regions of the republic in order to control the spread of radioactive sediments and the number of them is not enough [9]. It has been explained the method of application of below mentioned drone on a large scale and in continuous mode in the areas bordering with Armenia (pic.2):



Pic. 2. Application algorithm of radiation monitoring methods by using drones.

- Drones fly in defined route in the areas bordering with Armenia and monitor radiation background; and they are determined in the areas bordering with Armenia, especially in the east direction from "Metsamor" NPP;

- The flight of drone is defined in the height of 10-15 m (away from settlements); it is related with that, when the level of radioactive wastes, caused by the accident in "Metsamor" NPS, is 10% in atmosphere, the radiation level in the areas bordering with Armenia becomes approximately 400-600 mkR/hour (4-6 mkSv/hour) in the earth surface [2,8] and although radiation decreases by 25 times in this height, spectrometric and dosimetric devices in the drones can define sufficiently effective radioactive pollution;

- The results of the measurements of radiation background are transmitted to Chemical service of Operative headquarters by mobile communication on continuous real-time mode and they are processed [10]. Therefore, it is necessary to have mobile communication tools for the transmission of data more quickly;

– The chief of Chemical service of Operative headquarters is immediately informed when radiation background is high in any point (R > B), and radiation monitoring groups (from chemical departments) are sent there. Here, natural radiation background is as B=30 mk/hour (0,3 mkSv/hour) in open air [11].

Here, A is amount (concentration of activity) Bq/l; PA- permissible amount; B - natural radiation background.

Note: The chief of Chemical service of Operative headquarters is informed when PA is high, but flight is continued in defined route when PA is low.

2. Conclusions

- 1. 3 H, 14 C, 41 Ar, 83m Kr, 85 Kr, 85m Kr, 87 Kr, 88 Kr, 89 Kr, 133 Xe, 135m Xe, 135m Xe, 135m Xe, 137 Xe, 138 Xe, 139 Xe, 140 Xe, 131 I, 132 I, 134 I, 135 I radionuclides enter the environment and ecologically damage it during the activity of "Metsamor" NPS reactor (T¹/₂ ≥ 16 sec.).
- 2. Long half-life ¹²⁹I, ¹⁴C, ¹³⁷Cs, ³H, ¹³¹I, ¹³³I, ¹³³Xe, ¹³³MXe, ¹³³I, ¹³³I, ⁸⁵Kr radionuclides enter (overcome 75 km distance for 10 hours) the territory of Azerbaijan (firstly, Nakhchivan AR) on convection airflow of atmosphere (2m/sec.) and spread. According to the current

meteorological conditions as ⁴¹Ar, ^{83m}Kr, ^{85m}Kr, ⁸⁷Kr, ⁸⁸Kr, ⁸⁹Kr, ¹³⁵Xe, ^{135m}Xe, ¹³⁷Xe, ¹³⁸Xe, ¹³⁹Xe, ¹³⁹Xe, ¹⁴⁰Xe, ¹³²I, ¹³⁵I radionuclides are short half-life, the possibility of their entry into the country is 0.

3. It is proposed the use of drones in order to control the spread of radioactive sediments, caused by the accident that can occur in "Metsamor" NPS, on the territory of Azerbaijan Republic. Therefore, in paper the method of radiation spectrometric and dosimetric monitoring has been developed.

Offer

It is offered to use the radiation monitoring devices, which enable to carry out spectrometric and dosimetric measurement, in unarmed drones in Chemical Troops of the Armed Forces in order not to involve the financial, human and other resources during the accident that can occur in technogenic dangerous areas, as well as to fulfil the tasks more efficiently and control the spread of radioactive sediments.

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ВЫБРОС РАДИОАКТИВНЫХ ИЗОТОПОВ В АТМОСФЕРУ В РЕЗУЛЬТАТЕ ЭКСПЛУАТАЦИИ МЕЦАМОРСКОЙ АЭС И КОНТРОЛЬ РАДИОАКТИВНОГО ЗАГРЯЗНЕНИЯ НА ТЕРРИТОРИИ АЗЕРБАЙДЖАНСКОЙ РЕСПУБЛИКИ С ПОМОЩЬЮ БПЛА

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Резюме: В статье представлены результаты анализа характеристик короткоживущих газообразных радионуклидов, выбрасываемых из реактора Мецаморской АЭС и процесса распространения

долгоживущих радионуклидов (Т½ > 10 часов) на территории Азербайджанской Республики. Разработана методика использования БПЛА с целью проведения радиационной разведки на территории Азербайджанской Республики и предложено применение БПЛА в Химических Войсках Вооруженных Сил.

Ключевые слова: Мецаморская АЭС, ядерный реактор, радиоактивные выбросы, радионуклид, беспилотный летательный аппарат, радиационная разведка.

"METSAMOR" AES-in ST SMARI NƏT CƏS NDƏ ATMOSFERƏ ATILAN RAD OAKT V ZOTOPLAR VƏ PUA VAS TƏS LƏ AZƏRBAYCAN RESPUBL KASI ƏRAZ S NDƏ RAD OAKT V ÇÖKÜNTÜLƏR N YAYILMASINA NƏZARƏT

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Xülasə: Məqalədə "Metsamor" AES-in reaktordan atmosferə atılan qazaoxşar qısamüddərli radionuklidlərin qısa xarakteristikaları və Azərbaycan Respublikası ərazisinə daxil olan uzunmüddətli (T½ > 10 saat) radionuklidlərin yayılması prosesi analiz edilib. Azərbaycan Respublikasının ərazisində radioaktiv çöküntülərin yayılmasına nəzarət məqsədilə PUA-ın istifadə metodikası işlənilmişdir və Silahlı Qüvvələrinin Kimya Qoşunlarında məsafədən spektrometrik və dozimetrik ölçmələri aparmağa məqsədilə qəbul olunması təklif olunur.

Açar sözlər: Metsamor AES, nüvə reaktoru, radioaktiv tullantı, radionuklid, Pilotsuz uçuş aparatı, radiasiya kəşfiyyatı.