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New paradigm of research on the condition of spent nuclear fuel in accident and dumping sites

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Abstract. The new technologies of operative monitoring of spent nuclear fuel (SNF) condition at the bottom seas and search for the lost objects with SNF at the bottom using Kr-85, are developed. The development of the ship low-level background complex (Kr-85, tritium etc.) for radionuclide anomalies searching in the sea is launched also. The purpose of a carried out stage of researches is the development new paradigm of sea radioecology based on a preliminary experimental research of kinetics of release fission products (^{85}Kr and ^{137}Cs) from SNF, with the subsequent realization expedition (including preliminary radionuclide measurements on a vessel!) and modeling researches of ^{85}Kr transport in near-bottom layer for minimization of points of sampling (stations and working horizons) on the hydrological data of expedition area (with use of a fractal formalism). Final result of the project is the conservative estimation of risks to the population and environment at its pollution in case of release of a radioactivity from dumped ship reactor and NPS with the not unloaded SNF.

1. INTRODUCTION

Recent international conferences (ECORAD 2004, Aix-en-Provence; Int. Conf., Nice.2005 etc.) ascertained basic lack of our knowledge about the processes of change of the condition of spent nuclear fuel (SNF) at sea bottom. It was marked, that a set of studies is devoted to modeling the SNF condition; however, practically there are neither experimental works nor expedition researches aimed to establishing the start point of destruction of containments with SNF.

For the first time the data on ^{85}Kr and ^{137}Cs release from SNF are obtained at corrosion of one of them in sea water. The main conclusion is the proof of hypotheses correctness put forward by us in the beginning of 90s about earlier ^{85}Kr releasing (its release is an order more than ^{137}Cs release), than other fission products (FP) at loss of fuel rod case tightness during corrosion of SNF at the sea bottom [6, 7]. The purpose of a carried out stage of researches is the development of a new paradigm of sea radioecology based on a preliminary experimental research of kinetics of FP release (^{85}Kr and ^{137}Cs) from SNF, with the subsequent realization expedition (including preliminary radionuclide measurements on a vessel) and radionuclide transport modeling using fractal theory.

For the first time we present the results of the long-term experimental researches (1800 days) of the process of fission products (FP) of ^{235}U release from SNF into the sea water and development of methodology of search of anomalies (plume) of distribution of gaseous fission products, released from SNF in zones of accident and dumping.

2. METHODS

2.1 Development of shipboard technology of ^{85}Kr and ^{3}H measuring

2.1.1 ^{85}Kr technology

It was designed the ship multi-counter low-level background set up for operative measurements of radionuclide noble gases extracted from sea water, including: 1. Nine proportional counters with internal filling. 2. Vacuum system for gas clearing (extracted from 150 l sea water) and counter filling. 3.Anticoincidence veto from plastic scintillators. It may be established in ship laboratory (best place is hold) inside passive shielding from low activity materials. The vacuum system may be installed near the extractor on the desk. Ship multi-counter set up parameters:

– counter length – 5 cm; - diameter – 1 cm; - operating volt. over the range 600 to 1000 V;
– gas counter internal pres. – 600 mm Hg; - coefficient of gas amplif. stability better 1%

2.1.1.1 The description of ^{85}Kr measurement. The Kr is extracted from gas flux in low temperature U-formed trap with charcoal which is preliminary heated at temperature 100–150°C for Rn removing. Then Kr is passed into another low temperature trap (−100°C) with Xe portion that is gas carrier and counter working gas at the same time. For clearing gas sample (Kr+Xe) of possible impurity of oxygen, nitrogen, etc. clearing at temperature 900°C titan getter is used. Further gas sample is pumped by Toepler pump into gas counter with methane addition (10%). After filling the counter is put into the registration system and Kr activity is measured. Counter works in regime of limited proportionality with the coefficient of gas amplification $\sim 10^4$. The calibration is done with ^{55}Fe source (5.6 keV).

2.1.2 Tritium technology.

For operative work on ^{3}H measurement in the sea the shipboard models of installations were created, which were tested on several R/V ("Ak. Kurchatov"- 1971, "Professor Bogorov" - 1977,"Ak. Lavrent'ev" - 1985, etc). A ship variant of counting gas (CH_4) preparation system and equipment for tritium measurement by the gas proportional counter consists of: 1- reactor of synthesis of methane, 2 - heaters, 3 - vacuum valves, 4 - vacuum pump, 5 - membrane compressor, 6 - receiver, 7 - circulating pump, 8 – counter (volume 4 l), 9 - a.-c. protective counter, 10 - passive shield. Optimum arrangement of counters in shielding, selection of working gas of the protective counter, selection of discrimination levels have provided background speed of counting 3.8 count/min. at pressure 2.5 tam in the counter. It allows us during the measurement of 1000 minutes to detect tritium concentration of 1.7 TE with the probability of 0.975. Arrangement of electrolyzes installations on the vessel for preliminary hundredfold enrichment of water sample by tritium is provided. Installation is tested in the same cruises.

2.2 Modeling

Model of existential structure of distribution of a passive impurity is developed taking into account local heterogeneities in concentration. The impurity is spread under the influence of turbulent movements of sea waters. The structure of distribution of an impurity in space and time is defined by existential structure of a field of the turbulent currents transferring an impurity. The model is based on the combinatorial recursive sequence describing local structure of turbulence taking into account fluctuation of turbulent energy dissipation. We suppose that the dissipation of turbulent energy in a turbulent current is a random function of co-ordinates and time, a velocity of a current fluctuating together with a field. Fluctuations depend on singularities of large-scale movement - from Reynolds's defining number of cascades in hierarchy of curls of a various size number on which the dissipation kinetic energy in warmth is transmitted. Turbulence distribution in time and space is extremely nonuniform and has explicitly expressed alternating character. The logic design of the model consists in the following [4].

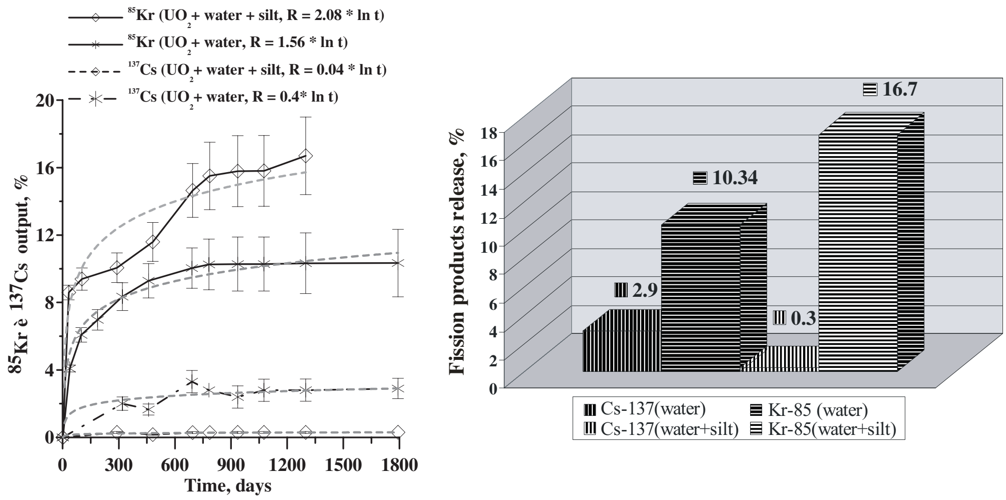

Figure 1. ^{85}Kr and ^{137}Cs release data for 1792 days (except for ^{85}Kr release data from the samples with water and silt, this data is given for 1299 days). Temperature from 1150 to 1792 days was +15 °C. In spite of this change the output kinetics was the same.

Let ε_r be dissipation of the turbulent energy which is taking place in the area by size r. Thus ε_r is a stream of kinetic energy from curls of size r to curls of a smaller size - this stream will be valid dissipation when these curls have an order of the Kolmogorov's scale of length η. It is supposed, that the curl of size r collapses in 2^d curls of equal size. There is a dimension of considered space $r/2$. Here d is a dimension of considered space. Besides, it is presumed, that the energy stream to these smaller curls occurs nongomogeneously. The elementary nontrivial choice is such: the part of energy of turbulence is transmitted by magnitude $p_{\varepsilon 1}$ to one half 2^d of new smaller curls, and the remaining part equal $p_{\varepsilon 2} = 1 - p_{\varepsilon 1}$ is transmitted to the other half of curls. This process repeats with constant $p_{\varepsilon 1} < 1$ further and further, till the curls reach order η. In the report it is given one-dimensional version of cascade model of curls which are split up in two new cascades. The kinetic energy stream to smaller scales is described by unequal shares $p_{\varepsilon 1}$ and $p_{\varepsilon 2}$. This cascade breaks when curls reach a size of Kolmogorov's scale η. Multifractal distribution corresponds to the generalized two-scale Cantor set with $l_1 = l_2 = 1/2$. The given model describes well the alternating structure of turbulence observed in various existential scales in ocean and atmosphere [4]. According to this model the concentration of an impurity will faster decrease in more turbulent areas of a liquid, and the concentration of an impurity is distributed under the logarithmically normal law. This law of distribution of concentration is received in other theoretical models of local heterogeneities of turbulent movements. Natural observations at ocean confirm the logarithmically normal law of distribution of these parameters [3, 5]. Thus, the offered model gives the law of distribution of impurity concentration that is consistent with the known theoretical and experimental representations (notions) so it can serve the acknowledgement of its adequate character.

3. RESULTS

3.1 Experimental data obtained in "hot" laboratory

Experimental data to SNF corrosion in sea water research are presented in fig. 1.

The received results for FP release from small samples (0.2 and 0.3 G of $^{235}UO_2$) in 2002-2007 have shown:

- kinetics of release of gaseous FP (^{85}Kr) from SNF strongly differs from kinetics of release of other FP (^{137}Cs). ^{85}Kr release rate dozens of times exceeds those for ^{137}Cs. Thus, *it is proved, that* ^{85}Kr

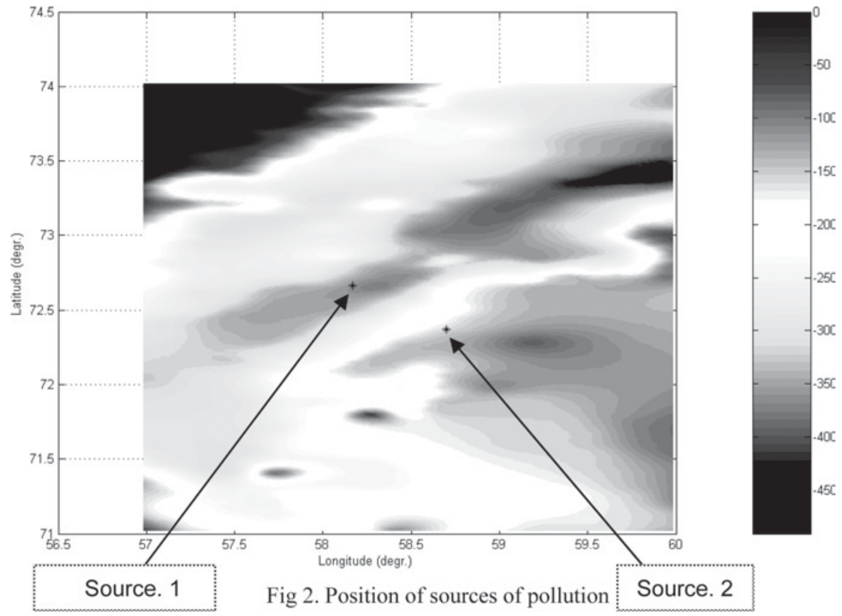

Figure 2. Position of sources of pollution.

is the best indicator of the beginning of fuel rods cladding failure and the following corrosion process $^{235}UO_2$.

- time of ^{85}Kr-output from SNF fragments (16,7% from total saved up ^{85}Kr for 1300 days of corrosion in sea water) *allow us to propose reliable and rather inexpensive methodology of periodic* (*once for 2–3 years*) monitoring of SNF condition on the sea-bottom in an accident and dumping zone.

The received experimental curves of total ^{85}Kr and ^{137}Cs release from time are well enough approximated by the following empirical dependences:

$$R^{85}Kr = 2,08 \ln t \text{ (water silt)}; \qquad R^{137}Cs = 0,04 \ln t \text{ (water silt)};$$
$$R^{85}Kr = 1,56 \ln t \text{ (water)}; \qquad R^{137}Cs = 0,4 \ln t \text{ (water)};$$

where: $R^{85}Kr$ and $R^{137}Cs$ are an output of the corresponding radionuclide in percentage of its accumulation in fuel at the moment of measurement; t - time from the start moment of fuel interaction with sea water (sea water with the additive of silt), in days.

Dependences of rate of ^{85}Kr and ^{137}Cs release out of SNF at its interaction with sea water during any moment of time are defined by the first derivation of a total output function of time. Thus, dependences of rate of radionuclide output out of SNF from time of the start moment of fuel interaction with sea water (sea water with silt) can be described by the following formulas:

$$V^{85}Kr = 2,08\ t^{-1} \text{ (water + silt)}; \qquad V^{137}Cs = 0,04\ t^{-1} \text{ (water + silt)};$$
$$V^{85}Kr = 1,56\ t^{-1} \text{ (water)}; \qquad V^{137}Cs = 0,4\ t^{-1} \text{ (water)};$$

where: $V^{85}Kr$ and $V^{137}Cs$ – are rate of an output of corresponding radionuclide; t - start moment of time of fuel interaction with sea water (sea water with silt).

Thus, it is possible to assume, that in case of the start moment of mass cladding damage of fuel rods out of active zones of atomic ice-breaker "Lenin", which dumped in the Kara Sea, the top limit rate of radionuclide release of SNF can be estimated by the dependences resulted for example in Fig. 2 [6].

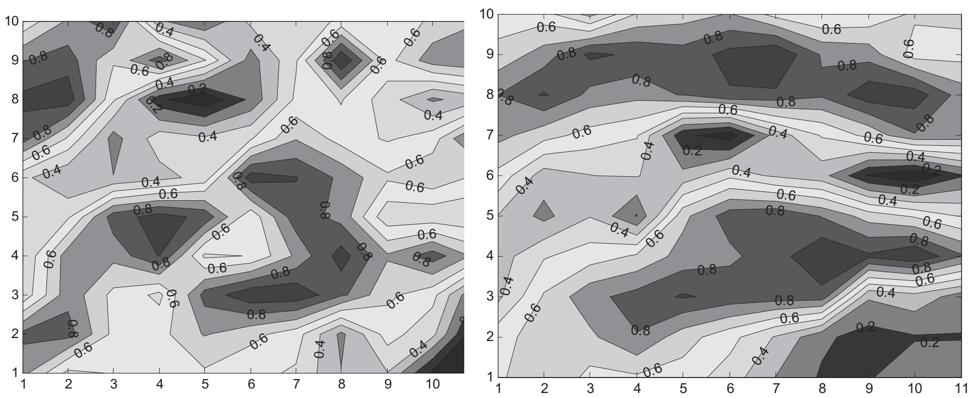

Figure 3. Estimation of monthly average values of an impurity concentration for August and September.

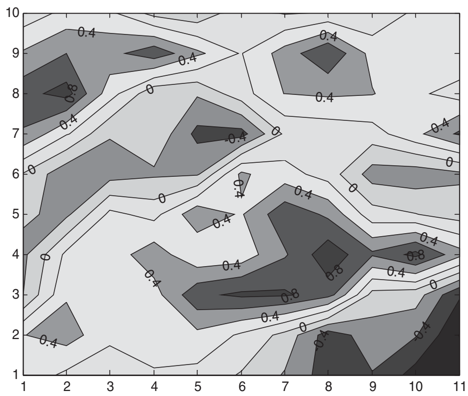

Figure 4. Difference between September and August values of an impurity concentration monthly average.

3.2 Results of radionuclide transport modeling from the source in the Novozemlya hollow

The developed imitating model has been applied to an estimation of distribution of a radioactive impurity in the allocated area of Kara Sea. Two sources of pollution were considered. Their position is presented by points in fig. 2. Data and a field of the speeds, calculated in AANII/AARI [2] on a uniform grid of a stereographic projection with step of 7.5 miles have been used bathymetric data.

For points monthly average files of horizontal speeds (sm /sec) near to a bottom for August and September, accordingly have been calculated. On the horizons nearest to a bottom speeds of currents have been calculated. (Monthly average vectors of benthonic currents for August and September).

Estimations monthly average values of an impurity concentration on the developed model are resulted for August and September in fig. 3, and the difference between them in fig. 4. From these results follows that it is necessary to expect the greatest concentration in areas with the minimum of current speeds. Variability of a site in time of areas with high concentration is the high. It is necessary to consider not only horizontal currents, but shift currents in a benthonic layer.

Thus, the model of the description of existential distribution of an impurity in a turbulent torch near to the sunken radioactive sources on the basis of empirically confirmed multifractal's distribution models in the field dissipation's turbulent energy is developed. On the basis of the known dependences checked up by natural supervision between values dissipation's turbulent energy and size of relative turbulent diffusion the combinatory model of distribution of an impurity in a turbulent torch is constructed, and with its use the distribution analysis impurity in a turbulent torch is carried out, and the imitating model is developed. Obvious expressions for distribution in the field of concentration of an impurity are received. They have sedated an appearance.

On the basis of concepts fractal's diffusion front interpretation of visible loss uniformity in structure of a turbulent torch on some distance from a source is given.

Numerical calculations for a benthonic layer of area of Kara Sea are spent. It is shown, that in the conditions of concrete area at low the information on dynamics of sea currents can draw qualitative conclusions on relative increase in concentration of an impurity in areas with small turbulence near to a radioactive source.

At a following stage of work on physical bases creation of studying of the mechanism of sea environment pollution at hypothetical accident in southern Primorski Krai (for example, at the huge high water leading to washout on a bottom of adjacent bights where coastal storehouses of the spent nuclear fuel are located) use of the developed models for reception of a conservative estimation is supposed.

4. CONCLUSION

Final result of the Project is the estimation of risks to the population and environment at its pollution in case of release of radioactivity from the dumped ship reactor and NPS with the not unloaded SNF. In the Arctic Region such objects can be SNF of reactor of the nuclear icebreaker "Lenin" dumped in the Kara Sea and reactors with SNF NPS K-159, sunken at the Kola Bay entry of the Barents Sea in August 2003.

The *new approach to the decision of the designated fundamental problem having also the important humanitarian value,* consists that for the first time attempt of a scientific substantiation and development of methodology and corresponding high-sensitivity techniques for early detection of release of gaseous FP (^{85}Kr, ^{3}H etc.) will be made, at contact of SNF to sea water in dumpsites NPS reactors, with the following radionuclide release into waters of Arctic (and other seas). This project will promote *reception of new knowledge of sedimentation and contamination processes of marine environment near to a bottom.* On the basis of 3D numerical modeling with use of an available base of hydrological data, and also the data received with use tritium during of planned expeditions, *"residence time " of gaseous FP in dump site* will be established.

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