

MODERN TECHNIQUES FOR CONTAMINATED SITES REMEDATION

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ABSTRACT

Remediation of radionuclide contaminated sites requires a combined technique for immobilizing and removal of radioactive matters captured by topsoil.

To this end, new immobilizing agents based on interpolyelectrolyte complexes (IPEC) have been developed by joint efforts of VNIINM and MSU. The IPEC treatment produces a water-insoluble but air- and moisture- permeable protective coat, which is resistant to water and wind erosion. The water erosion resistance has been tested under field conditions. The polymeric agents are found to aggregate the soil structure and to increase the water resistance by 3-10 times.

For wind erosion tests a laboratory wind tunnel was used. The erosion intensity and limiting air velocity studies with and without the protective coat show that in the wind velocity range from 0 to 40 m/s the treatment with 1.0 l/m² of the aqueous solution containing 2% wt. of IPEC provides a dust suppression factor of $10 - 7.5 \cdot 10^6$ as a function of the treatment technique.

The effect of the soil composition and properties on the soil-radionuclide interaction, the contamination features and factors influencing the radioactivity distribution throughout phases and fractions have been studied.

Polymer structureformers were experimentally tested using the confinement technique developed. IPEC with trade mark "MM-1" was used at the Chernobyl NPP site in 1986-1990 on a commercial scale. Also, polymers developed can be helpful in reducing highly-saline sand transfer from dried-up bottom of the area of the Aral See ecological calamity. Tests carried out under the Polyarny Ural since 1995 suggest that the polymers are useful for remediation of forest-tundra soils.

As a result of a comprehensive analysis of the Bryansk region soils contaminated due to the Chernobyl accident and relying on the Cs-137 distribution measurements a soil decontamination technique has been developed, described and experimentally tested. The decontamination factor is found to range from 2.0 to 4.5 depending on the decontamination procedure.

APPROACH

A great deal of studies evidences that topsoil (5-8 cm long) collects the radioactivity bulk (95 %). [1]. Considering large contaminated areas and long decay periods (10-25 years for Cs and Sr) it is felt that the contaminated soil remediation remains urgent in Russia now and in sight.

A wide variety of ecological systems (forests, bogs, water bodies) makes the development of a unified remediation technology difficult. A combined remediation procedure is needed to optimize the efficiency and to minimize the costs.

As a starting point for research the following grounds have been taken:

1. Decontamination of large areas is time consuming.
2. Long-term soil conservation including the radioactivity immobilization is needed.
3. Routine approach to site remediation involves removal of contaminated media to offsite locations, decontamination and return of the cleaned soil to be reused. The contaminated soil residue is treated as solid radioactive waste.

The final objective of the conception under consideration is the return of remediated soils to national economy.

Three-stages for combined soil remediation technique have been developed.

The first stage, immediately after an accident, involves the radioactivity immobilization by IPEC-based aggregating agents which are currently the most efficient ones [2-4]. An appropriate effort is directed to develop a procedure for their application and to study their resistance to water and wind.

The second stage (perhaps, within a few years) deals with the soil decontamination. Work is proceeding on the development of a decontamination procedure based on gravity separation to remove the radioactivity from fine-grained soils of various compactness.

The third stage completes the immobilization by applying a polymeric agent and perennial herb seeds at a time. After a while the turf layer developed serves as a natural protective cover against water and wind erosion.[5,6]

The paper summarizes basic data of our research into the radionuclide contaminated soil remediation technique.

RADINUCLIDE CONTAMINATED SOIL PROTECTION AGAINST WATER AND WIND EROSION

Concepts and Methods of Immobilizing Radionuclides on Topsoil

Radionuclide Mobility and Immobilization

Nuclear accidental releases generate a need for stabilizing radioactive matters on the surface of equipment, buildings and soils. The paper considers the immobilization of radioactivity captured by grass cover-free soil. In this case, there exists a risk of a secondary radioactivity transfer by wind and water streams. Since just after an accident radioactive matters are disposed at the extreme top of soils, the immobilization procedure is similar to that used for preventing erosion. The most efficient immobilization procedure involves the soil aggregation by high molecular compounds (HMC) including IPEC. The latter results from interplay between oppositely charged polyions, and the active polycationic groups interacting electrostatically with the kaolinite surface of negative charge. The negative charge can be caused by the partial dissociation of silanol groups Si-OH on the superficial irregularities.

During two-three years after an accident radionuclides are redistributed within topsoil: they penetrate through pores and cracks, migrate with water streams, are sorbed by soil components and plants. An important point is that with time radionuclides are primarily grouped within fine soil particles of 1-5 mkm in size. The Chernobyl's and other studies confirm this observation [1,5,7-10].

On the one hand, the immobilization efficiency depends on the proper mobility of radionuclides but, on the other hand, it is governed by their fixation with polymeric compounds.

Factors influencing the natural radionuclide mobility and migration can be classified into three groups: properties of a radionuclide; soil characteristics; weather and climatic conditions.

IPEC as Aggregators of Dispersed Systems

When eliminating the consequences of nuclear accidents a prompt fallout immobilization is a vital issue the resolution of which is responsible for environmental contamination and for the level to which the public may be exposed to radioactivity.

In the last decade in Russia a new generation of highly active polymeric aggregators or binders based on interpolyelectrolite complexes (IPEC) [1-4,12] has been developed. A new polymeric class – MM-1, MT-1, MN-1 - has been tested under

laboratory and field conditions by VNIINM and MSU. The polymers are characterized by ease of their preparation and application. They are prepared by mixing original components to get a diluted aqueous 1-2 wt. % IPEC solution (10 ton per ha) and applied by means of any standard spraying equipment available: sprinklers, hydraulic planters, helicopters. It is important that the final product requires no solvent other than water. The polymeric product applied on topsoil produces a soil-polymer crust 3-5 mm thick consisting of water-resistant aggregates, which are firmly attached to each other by the polymer. The MM1-based crust photomicrographs show sand particles of different size joined by thin polymeric layers, fine particles being clung to larger ones [2,3]. It is essential that there is no continuous film. The soil particles are fixed only at their juncture or by polymeric bridges. Owing to this the IPEC-treated soil remains penetrable to moisture and air.

Thus, the IPEC-soil interaction stabilizes topsoil and thereby prevents from dust generation and loss by wind and water. Besides, the crust developed is water insoluble, but air- and moisture-penetrable, contrary to that based on latex, and contributes to vegetation. The effect of IPEC on the wind and water resistance is reported.

IPEC Protective Properties

IPEC Influence on the Soil Water -Resistance

Laboratory tests and field observations at Chernobyl (1986-1993) and near Vladimir (spring-summer 1998, with pure soils similar in composition and erosion risk to Chernobyl's ones) were performed. The soils were treated with 1.0, 2.0 and 4.0 l/m² IPEC water solution. The treatment was found to contribute to the dry soil aggregation:

the weighted mean diameter of an aggregate ($D = \frac{\sum D_i(\%)_i}{100\%}$) is 1.4 -18 times that of IPEC-untreated soil (control).

Table I The Effect of IPEC on the Soil Aggregation and Water Resistance

Date	Aggregation									
	Period	Period	Control	Control		1.0 l/m ²		2.0 l/m ²		4.0 l/m ²
1-4 June	15	15	8,56	0,47	10,75	2,54	12,17	3,62	12,84	4,4
15 July	35	50	2,89	0,49	4,18	0,84	5,09	1,3	7,74	2,98
21-24 July	10	60	2,8	0,49	4,2	1,2	4,7	1,4	6,26	2,12
9-12 September	50	110	2,7	0,38	4,43	1,25	4,47	1,38	4,34	1,23

The treated soil showed a still further increase in water resistance (by 10 times in the early days and by 3.5 times 120 days later) as well as a 1.7-fold lower content of fine aggregates chiefly susceptible to being lost, with a consequent doubling in the value of V (Fig. 1).

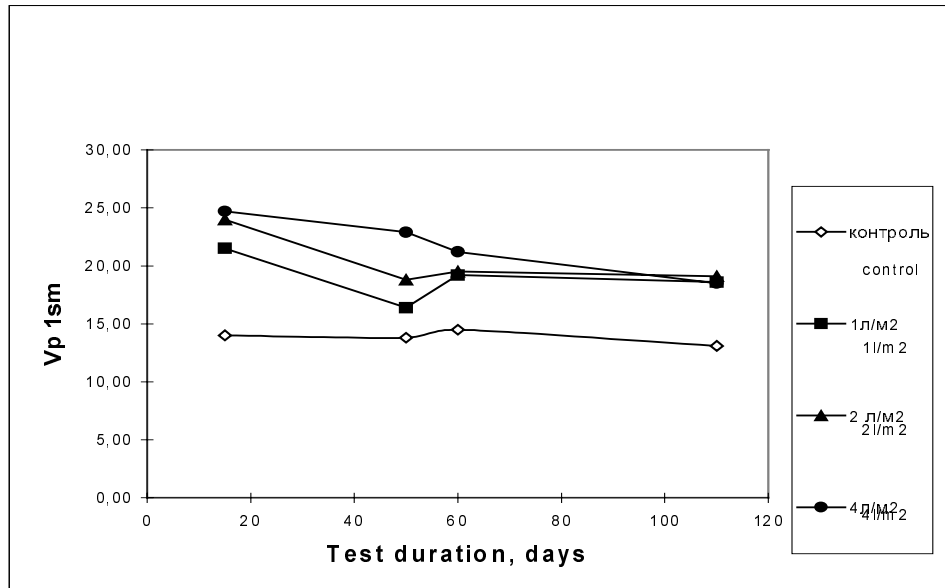


Fig. 1. Soil erosion resistance dynamics.

The IPEC treatment improved the soil erosion resistance. Within 60 days after the treatment the ratio V_{pIPEC} / V_{pcontr} became stable at ~ 1.5 and remained unaffected from this point on. The soil erosion resistance peaked in the early days of treatment with 4 l/m² IPEC while the best coat performance (expressed in terms of an increase in the erosion resistance per unit polymer consumption) was noted for IPEC doses ranging from 1.5 to 2.5 l/m² (Fig. 2).

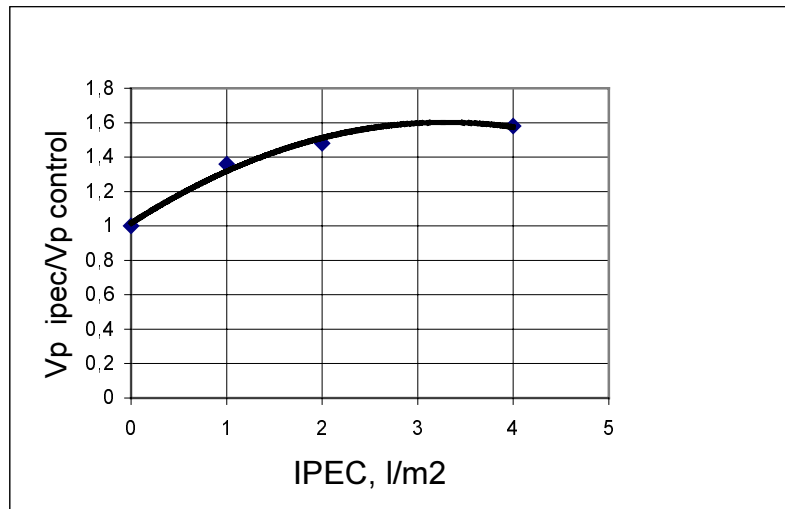


Fig. 2. Erosion resistance versus IPEC doses.

MM1 Protective Efficiency

The efficiency of MM1 in preventing against wind erosion was experimentally tested using a laboratory wind tunnel. [3,12]. The limiting air stream velocity above which soil is lost and the soil loss-air velocity relationship were used as a basic guideline.

A wind tunnel container was filled with a soil of measured humidity, structure, compactness, grain size, humus content and quality, polymeric concentration on its surface. The sample was allowed to stay at room temperature, placed into the wind tunnel and exposed to an air stream at a velocity varying from 8 to 40 m/s. The density of the samples was 1.50 g/cm³. Dry turf-podzolic samples treated with pure water and KNO₃ were used as controls. The mass transfer was determined from the amount of the soil lost and from the airborne particle concentration (using a laser particle size analyzer) [2,3,12].

The resulting data are listed in Table II.

Table

II. Efficiency of Immobilizing Polymeric Agents

Treatment	IPEC, %	U _{crit.} , m/s	(q _{cp} Δv), kg/m ² s	K (relative to an agent)		
				None	Water	KNO ₃
None	0	5,4	329,902	1	0,000001	0,000004
Water	0	5,9	0,00039	840904	1,00	3,1
KNO ₃	0	6,3	0,0012	274002	0,33	1,0
IPEC	0,02	5,6	0,00079	417416	0,50	1,5
IPEC	0,064	5,9	0,00133	247312	0,29	0,9
IPEC	0,2	6,4	0,00053	622069	0,74	2,3
IPEC	0,64	16,5	0,00036	914292	1,09	3,3
IPEC	2	33,2	4,4E-05	7531032	8,96	27,5

The limiting velocities obtained (Table) gave an insight into the nature of the relation between the crust mechanical strength and the polymer concentration. At <0.064% IPEC its efficiency was similar to that of water or KNO₃ but with increasing IPEC concentrations the limiting air stream velocities and hence the crust mechanical strength smoothly increased. The ratio of the loss for an IPEC-treated sample to that for water (KNO₃)-treated one was used in evaluating the polymer effect on the soil loss. The equation:

$$K = \frac{q_{cp}^{K.}}{q_{cp}^{H.}} = \frac{\frac{1}{\Delta v} \int_{v_1}^{v_2} q^{K.}(v) dv}{\frac{1}{\Delta v} \int_{v_1}^{v_2} q^{H.}(v) dv} = \frac{\int_{v_1}^{v_2} q^{K.}(v) dv}{\int_{v_1}^{v_2} q^{H.}(v) dv},$$

[1]

where $q_{cp.}^k$ is the untreated soil loss, $q_{cp.}^n$ is the IPEC treated soil loss served as an IPEC performance criterion, the polymeric efficiency factor being given by :

$$q_{cp.} = \frac{1}{\Delta v} \int_{v_1}^{v_2} q(v) dv,$$

[2]

At a concentration of 2% the MM1 mixture was 9 times as efficient as water ($K = 8.96$) and provided the mechanical strength good enough to resist a 33 m/s air stream. Even at 40 m/s the MM1- produced coat remained intact.

The generation of aerosols 0.3 - 10 mkm in size is shown in Fig. 3 .

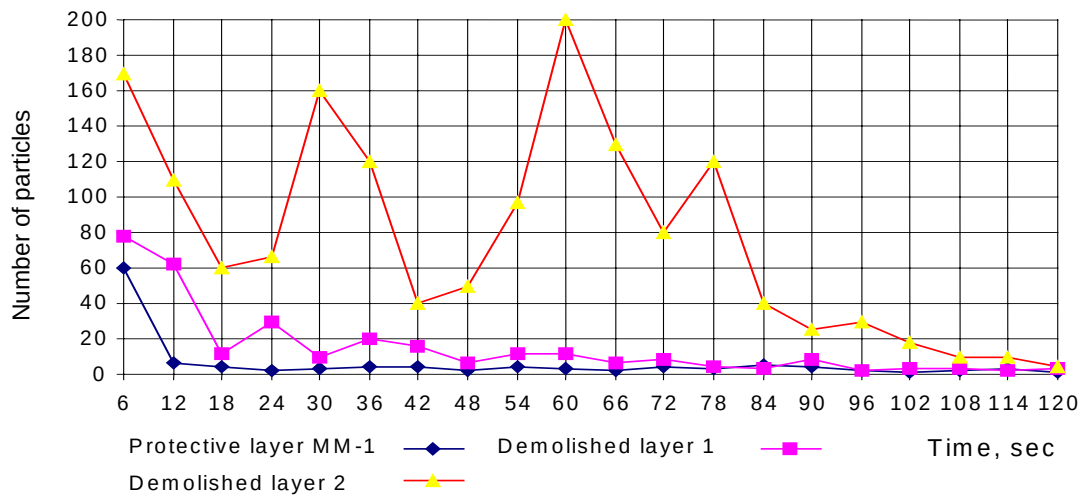


Fig. 3. Aerosols generation at air speed 25.0 m/s.

RADIONUCLIDE CONTAMINATED SOIL DECONTAMINATION TECHNOLOGY

A distinctive feature of soil as a radionuclide collector allows the use of dressing for its cleanup. The procedure involves the separation of radionuclides from fine-grained fractions (primary and secondary minerals and organics) of various compactness. Thus, as much as 95% of radionuclides can be removed and concentrated in a small volume (no more than 10-15% of the starting material). It should be noted that foreign decontamination procedures are intended for nonagricultural lands while in Russia the major part of decontaminated soils is a ploughland.

The radionuclide removal efficiency depends heavily on the soil type, the organics content, structure and properties and on the contamination type. Our experience shows that the most important factors in this respect are:

- chemical and mineralogical soil composition;
- physical and mechanical soil composition;
- level, type and distribution of radionuclides throughout soil fractions.

Contaminated Soil Study

Contamination characterization:

Type: radioactive fallout following the Chernobyl accident;

Major radionuclides: Cs137. Sr90

Level: 15 Ci/km or 6-7 Bk/g

Table III. Soil Characterization.

Type:	turf, podsol, sandy
Organics content:	8-10 wt %
PH:	5.6
Clay particles	4%
Humus	~1.5%
Grain size :	> 0.05 mm - 75.6%
	0.05 mm -0.005 mm - 23.9%
	<0.005 mm - 0.5%

TABLE IV. Sample Measurement, Site 1.

Fraction, mm		Content % wt.	Cs-137, Bk/g	Activity, %	Particle density, g/cm ³
Sample		100,0	6,11	100,0	2,450
Mineral bulk	Organic-mineral portion				
1,0 - 0,630	1,0 - 0,630	2,6 1,9	1,51 29,5	0,65 9,32	2,513 1,134
0,630 - 0,315	0,630 - 0,315	15,1 1,7	1,2 34,5	2,95 9,57	2,640 1,656
0,315 - 0,160	0,315 - 0,160	51,2 1,5	1,04 37,6	9,04 9,27	2,503 1,873
0,160 - 0,063	0,160 - 0,063	17,3 2,4	2,05 42,7	5,93 16,61	2,613 1,905
sludge portion less 0,063		6,3	35,5	36,66	2,100
"Clean soil"	"Contaminated Soil"	86,2 13,8	1,28 36,0	18,57 81,43	

TABLE V. Sample Measurement, Site 2.

Fraction, mm		Content % wt.	Cs-137, Bk/g	Activity, %	Particle density, g/cm ³
Sample		100,0	6,11	100,0	2,450
mineral bulk	organic-mineral portion				
1,0 - 0,630	1,0 - 0,630	1,9 1,2	1,01 21,5	0,3 4,4	2,58 1,03
0,630 - 0,315	0,630 - 0,315	11,4 1,3	1,14 26,9	2,1 6,0	2,60 1,56
0,315 - 0,160	0,315 - 0,160	57,2 1,1	1,47 34,5	13,6 7,4	2,61 1,77
0,160 - 0,063	0,160 - 0,063	17,1 1,2	2,45 48,4	6,7 10,1	2,60 1,90
sludge portion less 0,063		7,6	40,0	49,0	2,20
"Clean soil"	"Contaminated Soil"	87,6 12,4	1,6 37,1	22,1 77,9	

Tables shows the analytical results obtained for Chernobyl-contaminated soils sampled from the Bryansk region. The results confirm that the soil consists of three principal components different in composition and radioactivity distribution:

1. The **mineral bulk** is composed of wind-blown silt particles 1.0-0.063 mm in size with an average density of 2.60 g/cm³. X-ray examination evidences α -quartz SiO₂ as a mineral. The organic matters present cause its low density as compared to that of quartz. The sand component amounts for 84% and contains as much as 20% of the total radioactivity.
2. The **sludge portion** (6-8%) consists of fine sandy, clay and organic particles <0.063 mm in size with a density of 1.6-2.0 g/cm³ similar to that of clay. The sludge incorporates 30-40% of radioactivity.
3. The **organic-mineral portion** (8-10%) is composed of organic substances with sand and clay impurities and incorporates as much as 30-40% of radioactivity.

The soil under study is characterized by a high radioactivity (50% of the total level) of the organic-mineral component.

The decontamination efficiency depends on the content of fine particles and the level to which they can be separated from the soil bulk. Gravity separation shows much promise in classifying mineral particles into sizes and densities. If necessary, other procedures (floatation, washing, etc.) can be used as supplementary ones. The dressing procedure has also been successfully tested on a large scale at Novosybkovo (Bryansk region) and Khalch (Gomel region, Belorussia).

Decontamination Process

The decontamination procedure is based on classifying soils hydraulically and separating fine-grained particles in an aqueous medium, the water circulated around a closed path and solid waste compacted. The personnel is protected against radioactive dust and aerosols by a special system of dust collectors and discharges. Besides, the aqueous medium gives no way of generating dust.

A thin contaminated layer 5 -10 cm thick is removed with a special device to be coarsely screened and further disintegrated into small particles. The pulp is separated. The mineral portion is also classified into sizes. The fine-grained portion is concentrated, compacted and discharged while the remainder is returned to a field.

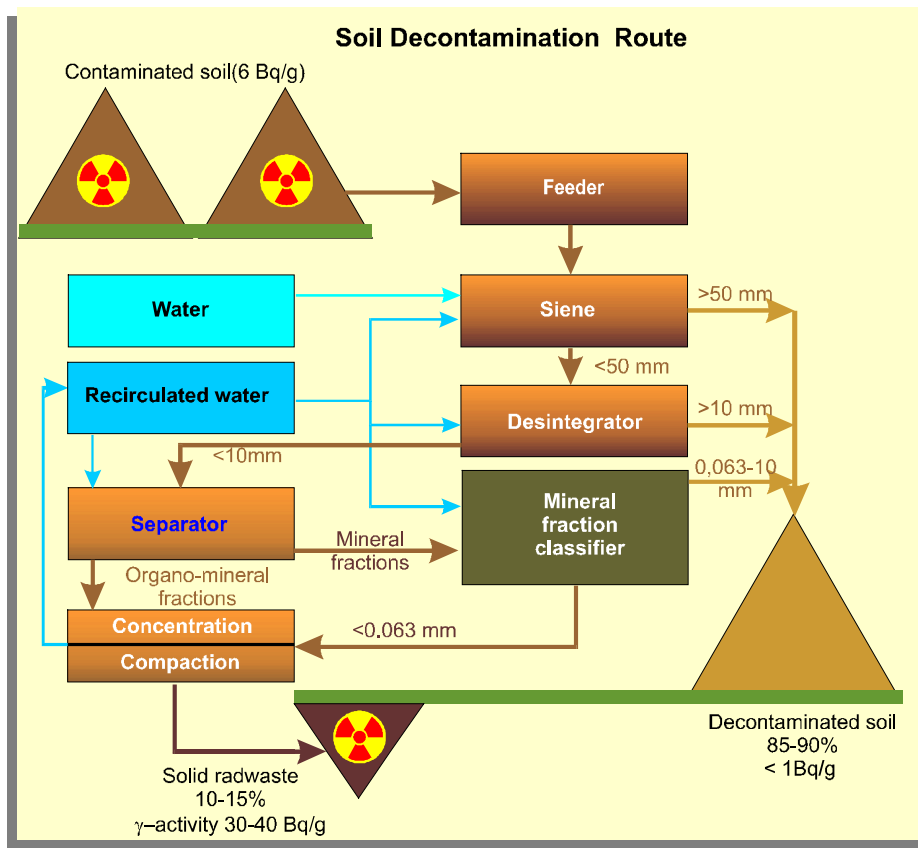


Fig. 4. Schematic diagram of the soil decontamination process.

The resulting decontamination factor ranges from 4.5 to 2.0 (Table 6) as a function of the extent to which the sludge and organic-mineral particles are removed. The cleaned soil radioactivity can be as low as 1.3 - 1.6 Bq/g.

Table VI. Decontamination efficiency

Portions separated	Decontamination factor ($K_d = A_{II} / A_K$)
Sludge (<0.06 mm)	2.0
Sludge + organics	4.5

A - the Cs-137 content prior to decontamination

A - the Cs-137 content after decontamination

CONCLUSIONS

1. Interpolyelectrolyte complexes - based agents and a procedure have been developed for preventing contaminated soil erosion.
2. The performance of the polymeric agents has been specified, tested and compared with polymeric alternatives.
3. The polymeric agents developed are recommended for wide application in the event of industrial or ecological emergency.
4. A new soil decontamination procedure has been developed, researched and tested on Chernobyl-contaminated soils.

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