



EUROPEAN
COMMISSION

Community research

COMET

(Contract Number: Fission-2012-3.4.1-604794)

DELIVERABLE IRA-Particle Behaviour-D2

Characterization of fuel particles (FP) and associated dissolution kinetics based on FP from a Red Forest radioactive waste trench

Author(s): V. Kashparov, B. Salbu, S. Levchuk, V. Protsak, I. Maloshtan, C. Courbet, H. Nguyen,

Date of issue of this report: 13/09/2016

Start date of project: 01/06/2013

Duration: 48 Months



DISTRIBUTION LIST

Name	Number of copies	Comments
André Jouve, COMET, EC Project Officer	1	Electronically
Hildegarde Vandenhove, COMET Co-ordinator (WP-1), SCK•CEN	1	Electronically (pdf file)
Contributors V. Kashparov , UIAR of NUBiP S. Levchuk, UIAR of NUBiP V.Protsak, UIAR of NUBiP I.Maloshtan, UIAR of NUBiP B. Salbu, NMBU C. Courbet, IRSN H. Nguyen, IRSN	1 per contributor	Electronically (pdf file)
COMET Executive Committee members: WP-1: H. Vandenhove, SCK•CEN WP-2: M. Muikku, STUK WP-3: Å. Sjøvik, NRPA WP-4: C. Adam-Guillermin, IRSN WP-5: B. Howard, NERC	1 per member	Electronically (pdf file)
COMET Management Committee: H. Vandenhove, SCK•CEN T. Ikäheimonen, STUK Å. Sjøvik, NRPA J. Garnier-Laplace, IRSN B. Howard, NERC A. Real, CIEMAT M. Steiner, BfS C. Bradshaw, SU B. Salbu, NMBU B. Michalik, GIG V. Kashparov, UIAR S. Gashack, Chernobyl Centre K. Nanba, Fukushima University P. Masqué, Universitat Autònoma de Barcelona K.O. Buesseler, Woods Hole Oceanographic Institute J. Nishikawa, Tokai University M. Christl, ETH, Zurich R. García-Tenorio, University of Seville P. Roos, DTU D. Child, ANSTO	1 per member	Electronically (pdf file)

[COMET]

2/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, _____
characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016

COMET Steering Committee	1 per member	Electronically (pdf file)
COMET Wiki site	1 per member	
ALLIANCE	1 per member	

[COMET]

3/19

Project co-funded by the European Commission under the Seventh Euratom Framework Programme for Nuclear Research & Training Activities		
Dissemination Level		
PU	Public	
RE	Restricted to a group specified by the partners of the [COMET] project	
CO	Confidential, only for partners of the [COMET] project	CO

[COMET]

4/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, _____
 characterization of FP matrix.
 COMET Particle Behaviour Group (Initial Research Activity)
 Dissemination level: CO
 Date of issue of this report: 08/09/2016

Table of Contents

Executive Summary	6
Introduction.....	7
Methods and Results.....	12
Radiochemical determination of physico-chemical forms of the radionuclides	13
Assessment of the radionuclide fractions associated with FP of various types	13
Fraction of the chemically extra-stable fuel particles.....	15
Fraction of the UO ₂ fuel particles.....	15
Conclusion	17
References.....	18

[COMET]

5/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, _____
characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016

Executive Summary

Contamination of the near exclusion zone of the Chernobyl nuclear power plant (ChNPP) was initially mainly composed of refractory radionuclides associated with spent nuclear UO₂ fuel particles (FP). The model parameters of weathering and of the FP dissolution in soils situated in the radioactive waste trench and in sediments as well as in the drained areas describe the mobility and bioavailability of particle associated radionuclides. Refinement of these parameters by systematization of existing data and by obtaining novel data on the long-term environmental behaviour of Chernobyl fuel radioactive particles (solubility in the soil, migration to ground water and transfer of radionuclides to plants) is very important for assessment of environmental impact and for radiation protection of humans and the environment.

In 2015, the efficiency of the models on Chernobyl fuel particles dissolution, initiated 15 years ago, and the reliability of the predictions of the dynamics of radionuclides leaching, have been verified in an experimental site situated in the so-called Red Forest within the Chernobyl exclusion zone. At this site radioactive waste was disposed into shallow sub-surface storages i.e., waste trenches in an area about 2.5 kilometers west from the Chernobyl Nuclear Power Plant - ChNPP. The main results include identification of the radionuclide source term: the description of the physical and chemical properties of the fuel particles encountered in the waste trench, and a model of fuel particles dissolution and subsequent radionuclide leaching into the soil solution inside the waste storage site under natural conditions. The theoretical/modeling estimations of 2000-2001 of the exchangeable fraction of radionuclide activity in soils coincide with the experimental results obtained in 2015. The data show that the mobile form of ⁹⁰Sr in the trench has reached the maximal value.

The results will be prepared for publication by NUBiP, IRSN and NMBU and submitted to Journal of Environmental Radioactivity.

[COMET]

6/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, _____
characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016

Introduction

Following the Chernobyl accident in 1986, the near zone (30-km) of the reactor was contaminated with refractory radionuclides, which were deposited mainly in a matrix of fine-dispersed irradiated nuclear fuel – fuel particles (FP) [1, 2, 3]. Extensive work associated with soil decontamination/cleanup was initiated just after the accident. The work included removal and burying the upper 10-cm soil layer. For this purpose, temporary storages of radioactive waste (TSRW) - the near-surface radioactive waste burial (trenches) - were utilized. In the first post-accidental years, during very short time, more than 800 trenches in the 30-km zone were created without proper hydro-isolation. According to the inspections carried out in 1996, radioactivity in TSRW in the 30-km Chernobyl zone amounted in 2000 to $1.6 \cdot 10^{15}$ Bq [3]. Some trenches are presently periodically flooded by ground water, which can promote the migration of radionuclides from these trenches [4, 5, 6]. This demonstrates the importance of developing a strategy of management for the TSRW in the Chernobyl zone.

The study site is located 2.5 km South-West of the ChNPP in the so-called TSRW “Red Forest”. In 1987 in the course of the clean-up work, the radioactive materials such as topsoil layer (>90 % activity) and contaminated tree trunks from the dead pines killed by extreme radiation levels in 1986, had been bulldozed and buried *in situ* in a few meters deep trenches. Afterwards, the whole territory of the Red Forest was covered with a 30-50 cm clean sand layer. Young pine and birch trees, bushes and grass were planted to prevent the wind resuspension of sand.

A particular waste burial (trench #22), namely Chernobyl Pilot Site (CPS), was selected in 1998 by the joint French-Ukrainian scientist team (the Institute for Radiation Protection and Nuclear Safety (IRSN) from France, the Ukrainian Institute of Agricultural Radiology (UIAR) and the Institute of Geological Sciences (IGS) from Ukraine) for radiological characterization and monitoring studies as illustrated in Fig. 1. [7, 8, 9]. The burial represents an about 70 m long, about 8-10 m wide and about 2-2.5 m deep unlined trench. Radiometric results made it possible to determine the spatial distribution of ^{137}Cs activity in the trench and to estimate the total activity levels of ^{137}Cs , ^{90}Sr and $^{238,239,240}\text{Pu}$; $A_0^{137}\text{Cs}=800\pm360$ GBq, $A_0^{90}\text{Sr}=400\pm160$ GBq, $^{238-240}\text{Pu}=6.7\pm3.2$ GBq related to 1986. The source term of radionuclide migration to the geo-environment is a heterogeneous mixture of contaminated organic materials and soils containing micron-sized FP. The depth to the groundwater table is about 2-3 m. During 28 years disposal, the radionuclides have been leached from the trench due to precipitation (average annual rainfall is 550-650 mm), and have been penetrating the underlying unsaturated soil and the aquifer. As a result, the ^{90}Sr and $^{238-240}\text{Pu}$ activity concentration in groundwater in the upper part of the aquifer in the vicinity of the trench were in the range $n \times 100$ -10,000 Bq/l and $0.n$ - 500 mBq/l, respectively, and the radiostrontium and plutonium plume has spread some 10 m downstream from the source [4, 5, 6].

To predict the radionuclide migration from the trench into groundwater and vegetation, it is not sufficient to know how the radionuclides are distributed inside the storage trench and what are their total amounts. We have to know the long-term dynamics of the mobile forms of particle associated radionuclides, which is determined by the fuel particles weathering rate and dissolution. Thus, it is necessary to develop and to parameterize a model

[COMET]

7/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, _____
characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016

describing the FP dissolution. Following particle weathering and FP dissolution, associated radionuclides are mobilized and transferred into soil solution and can subsequently be involved into abiogenic and biogenic migration.

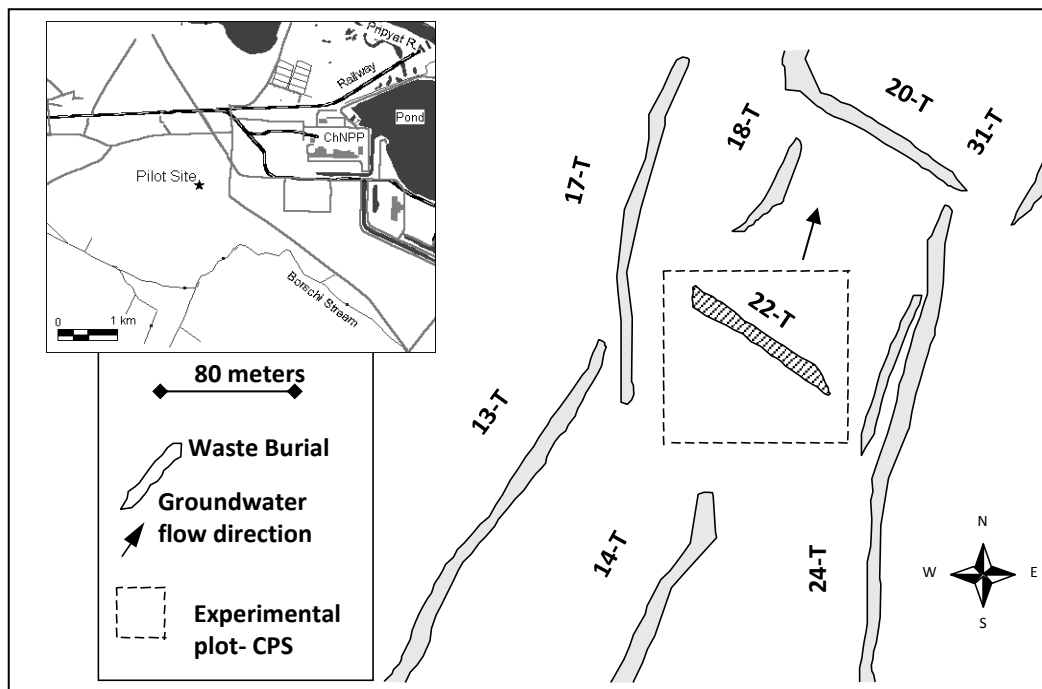


Fig.1. Map of the Chernobyl Pilot Site

About 3-4 tons of spent fuel was released from the reactor, ranging from sub micrometer sized particles to fragments. The large ^{90}Sr and Pu mass in the initial fallout in 1986 was predominantly present as fine-dispersed nuclear fuel particles of various level of transformation, with low mobility and bioavailability. The ratio between the activities of refractory radionuclides (i.e., ^{90}Sr , $^{154,155}\text{Eu}$, $^{238-241}\text{Pu}$ and ^{241}Am) in the fuel component of Chernobyl radioactive fallout corresponded to that of the nuclear fuel at the time of the accident [1,2,3]. The FP matrix is subjected to dissolution under natural conditions, and the radionuclides are leached into the soil solution and subsequently involved into ecosystem transfer processes. The FP dissolution rate in soils of the near zone is determined by the physico-chemical characteristics of the particles e.g., composition, particle size, morphology, crystalline structure, oxidation state such as ratio of uranium oxides $\text{UO}_2/\text{U}_3\text{O}_8$, and by pH (acidity) of the soil solution and by the presence of oxygen [10,11,12].

Fuel particles (separate grains and crystallites of uranium oxide with median diameter of 4-6 μm) and their conglomerates in the radioactive fallout can be divided into 3 types (Fig.2) [11]:

- ✓ Chemically extra-stable particles (hypothetically U-Zr-O, formed as a result of high-temperature and high pressure annealing (>1900 C) of UO_2 in presence of zirconium or carbon in construction materials (e.g.,

[COMET]

zirkaloid, moderator) and without air [11]). These particles were formed in the first moment of the accident on 26.04.86 and were deposited within the narrow western trace passing through the experimental site Red Forest;

- ✓ Non-oxidized chemically stable fuel particles (UO_2) associated with the first release (26.04.86), formed as a result of mechanical destruction of the nuclear fuel [13]. These particles created the narrow western trace of fallout, passing through the experimental site Red Forest. The leaching of the fission products from the particles during the accident was minimal. This fact is confirmed by constant ratio of fission products;
- ✓ Chemically low stability particles (UO_{2+x}), formed as a result of the fire i.e., oxidization of U in the nuclear fuel in the period 26.04.86-5.05.86 [14]. These particles created the Northern and Southern traces of fuel fallout.

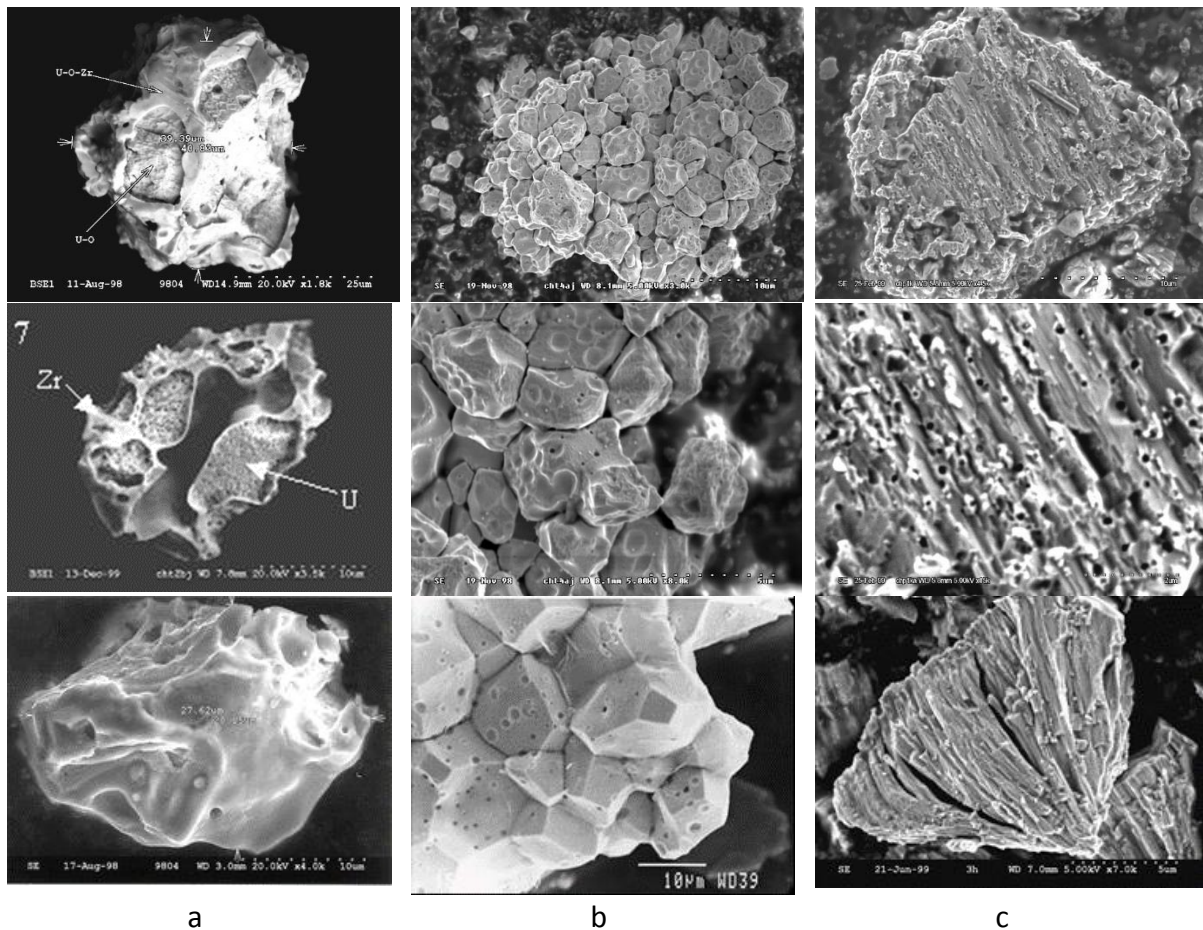


Fig.2. SEM images of fuel particles from trench no. 22: (a) ZrU_yO_x fuel particles; (b) UO_2 fuel particles; (c) UO_{2+x} fuel particles [15, 16].

[COMET]

9/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, _____
 characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016

Previous investigations using synchrotron based radiation x-ray micro-analytical techniques such as μ -XANES and μ -XRD/XRF, showed that fuel particles released during the initial explosion (west of the reactor) were not oxidized; contained a UO_2 core with layers of inert and apparently reduced forms of U associated with Zr or carbide (13). In contrast, particles released during the reactor fire (north of the reactor) were characterized by a UO_2 core surrounded by oxidized U ($\text{U}_2\text{O}_5/\text{U}_3\text{O}_8$ and possibly other intermediate forms). Although the source was the same (spent UO_2 fuel), differences in the release scenario; explosion (high temperature, high pressure and no oxygen) and the fire (median temperature, normal pressure and the presence of air) influenced the particle characteristics (elemental composition, crystallographic structures and oxidation states of U in fuel particles), influencing weathering kinetics, mobility and soil-to-vegetation transfer coefficients of radionuclides.

A superposition of all three types of the fuel particles is observed in the initial fallout in various points of 30-km ChNPP zone. Their partial contributions vary depending on direction and distance from ChNPP. Classification of the fuel particles into three types is hypothetical because of difficulties in the determination of the borders between various types of the particles (e.g., oxidization degree of uranium and the zirconium content in particles can vary in a wide range) [11, 13]. Based on the fact that the narrow western fuel trace of the radioactive fallout passing through the Red forest site was mainly formed at the initial moment of the accident 26.04.86, it can be assumed that the trace predominantly contained UO_2 and U-Zr-O fuel particles. Furthermore, as wind direction changed significantly during 10 days of the subsequent fire, and the Red Forest is located at a short distance from the source of release, particles with oxidized uranium (UO_{2+x}) and condensed ^{90}Sr are expected to be presented in the fallout in this area. Condensation particles are formed when volatile radionuclides deposit on available particle surfaces (e.g., fly ash).

Process of dissolution of the fuel particles of various geneses (U-Zr-O, UO_2 and UO_{2+x} particles) can be described as a sum of the first-order kinetic equations [10, 11, 12]:

$$\Delta FP(t) = \sum_{i=1}^4 \Delta FP_i(0) \cdot \exp(-k_i \cdot t) \quad (1)$$

$$\sum_{i=1}^4 \Delta FP_i(0) = 1$$

where: $\Delta FP_i(0)$ is the initial fraction (part of the activity) of the i-type particles (genesis: $i=1$ for **U-Zr-O** particles, $i=2$ for **UO_2** particles, $i=3$ for **UO_{2+x}** particles and $i=4$ for condensed particles of ^{90}Sr outside FP);

k_i – transformation constant of the particles of i-type (year^{-1});

t – time after the beginning of the dissolution process (years).

As a result of studies of the fuel particles dissolution kinetics in natural and simulated conditions, the relative (%) fractions of various FP types in Chernobyl radioactive fallout were

[COMET]

10/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, _____
 characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016

estimated for a first time [10, 11]. Using values of the fuel particles transformation constants ($k_2=0,018\pm 0,001 \text{ year}^{-1}$ and $k_3=0,28\pm 0,05 \text{ year}^{-1}$ at $\text{pH}=5$) and the value of ^{90}Sr activity fraction in fuel particles ($\Delta\text{FP}(t)=0,64\pm 0,13$ for $t=15$ years), fractions of activity of UO_2 and UO_{2+x} particles for $t=0$ were thus obtained.

It was found that in TSRW Red Forest (2.5 kilometers South-West of the ChNPP) ^{90}Sr and $^{154,155}\text{Eu}$, $^{238-241}\text{Pu}$ and ^{241}Am in initial radioactive contamination of the trench (at $t=0$) were present in four different physico-chemical forms [16, 17]:

1. **U-Zr-O** particles: $\Delta\text{FP}_1(0)=0,20\pm 0,10$ of ^{90}Sr total activity and $\Delta\text{FP}_1(0)=0,22\pm 0,10$ of $^{154,155}\text{Eu}$, $^{238-241}\text{Pu}$ and ^{241}Am total activity ($k_1\approx 0$)
2. **UO_2** particles: $\Delta\text{FP}_2(0)=0,57\pm 0,15$ of ^{90}Sr and $^{154,155}\text{Eu}$, $^{238-241}\text{Pu}$ and ^{241}Am total activity ($k_2=0,018\pm 0,001 \text{ year}^{-1}$ for $\text{pH}=5$)
3. **UO_{2+x}** particles: $\Delta\text{FP}_3(0)=0,21\pm 0,15$ of ^{90}Sr , $^{154,155}\text{Eu}$, $^{238-241}\text{Pu}$ and ^{241}Am total activity ($k_3=0,28\pm 0,05 \text{ year}^{-1}$ for $\text{pH}=5$)
4. Condensed particles of ^{90}Sr outside FP: $\Delta\text{FP}_4(0)=0,02\pm 0,01$ of ^{90}Sr total activity ($k_4=\infty$). $\Delta\text{FP}_4(0)=0$ for $^{154,155}\text{Eu}$, $^{238-241}\text{Pu}$ and ^{241}Am .

In order to characterize the trench source term, the parameters of the FP dissolution kinetics in the trench were estimated and the model of the ^{90}Sr mobile forms dynamics was created in 2001. Thus, we can calculate the relative dynamics of the ^{90}Sr FP-associated fraction $\Delta\text{FP}(t)$ and that leached from FP into mobile forms fraction of strontium activity $A_{\text{Sr}}(t)$ in the trench (Feil! Fant ikke referanse kilden.3):

$$\Delta\text{FP}(t) = (0,20\pm 0,10) + (0,57\pm 0,15) * \exp(-(0,018\pm 0,001) * t) + (0,21\pm 0,15) * \exp(-(0,28\pm 0,05) * t);$$

$$A_{\text{Sr}}(t) / (A_{\text{Sr-total}}(0) * \exp(-\lambda * t)) = (1 - \Delta\text{FP}(t))$$

The results illustrated that presently, 30 years after the accident; the activity of the mobile forms of ^{90}Sr in the trench had reached the maximum value.

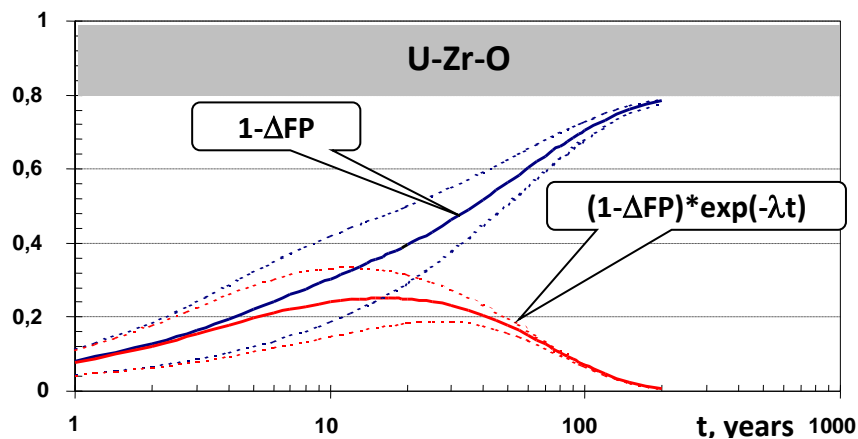


Fig.3. Relative (%) dynamics of ^{90}Sr contents in the mobile forms in the trench (without release due the migrations from the trench)

Methods and Results

Soil sampling in the trench was carried out in 2015 at the depths up to 3 m using the standard agrochemical sampler with a cylindrical working part ($\varnothing=6$ cm, $h=10$ cm). Taking into account the specific heterogeneity and spatial distribution of the radionuclides in the trench [4, 9], the samples were collected in 5 points along the trench axis near the sampling points used in 2000-2001 as illustrated in **Feil! Fant ikke referansekinden.** [17] from depth 0.5-1.0 m, 1.0-1.5 m, 1.5-2.0 m and 2.0-2.5 m. The composite representative sample was prepared from the 20 single samples (120 g DW) from different sampling points and different depth of the trench 22 for analysis. The composite representative sample was thoroughly homogenized. Relative standard deviation of the specific activity of ^{137}Cs in any 5 subsamples (120 g DW) of the composite sample was less than 20 %).

[COMET]

12/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, _____
 characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016

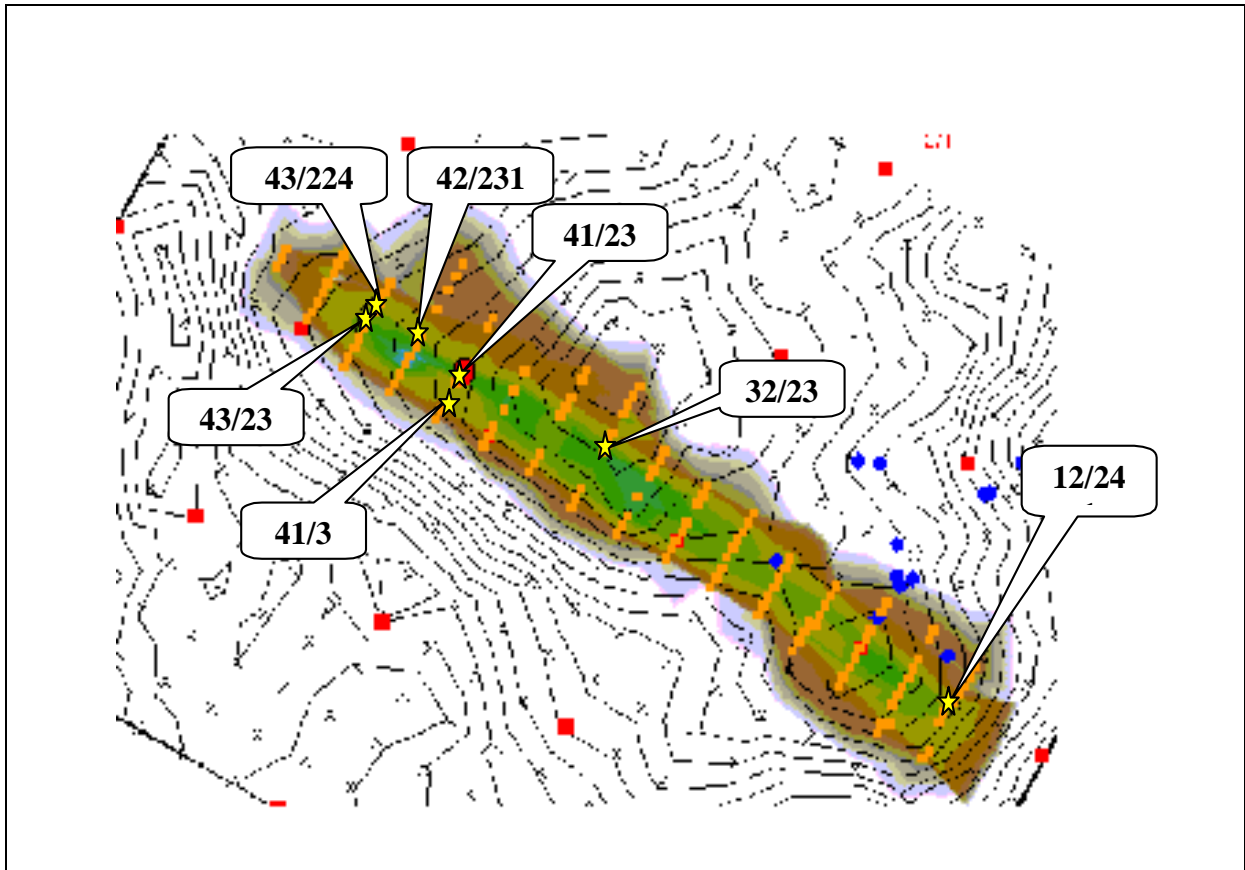


Fig. 4. Location of the sampling points in the trench #22

Radiochemical determination of physico-chemical forms of the radionuclides

Gamma-spectrometry of the composite sample of the soil (120 g) and solution samples was performed using a low-background gamma-spectrometer with multichannel analyzer ASPEC-927 (software GammaVision 32) equipped with passive shield and a semi-conducting detector of pure germanium GEM-30185, (EG&G ORTEC, USA) for determination ^{137}Cs , ^{154}Eu and ^{241}Am activity.

The activity of ^{90}Sr in the composite sample of the soil (10 subsamples with mass 10 g) after radiochemical separation from solutions was measured using scintillation beta-gamma-spectrometer SEB-01-150 or SEB-01-70 (Research and Production Enterprise “AKP” - “Atom Komplex Prylad”, Ukraine).

Assessment of the radionuclide fractions associated with FP of various types

For estimating the radionuclide (^{90}Sr , ^{154}Eu and ^{241}Am) fractions associated with FP of various types in the 120 g the composite sample of soil from the trench no.22, we applied

[COMET]

sequential extractions with injection of a radioactive spike. This method was developed for analyses of the radioactive contaminated topsoil, nuclear fuel and radioactive waste [10, 11, 12, 16, 17]. In the composite sample of soil from the trench no. 22 radionuclide (^{90}Sr , ^{137}Cs , ^{154}Eu and ^{241}Am) activity concentrations were measured by means of gamma and beta spectrometry (Table 1). Each sub-sample of the composite sample of soil (about 20 g) was measured 5 times by means of beta spectrometry and mixed after each measurement in order to reduce the effect of heterogeneity of contamination.

In the first stage, extractions by 2M NH_4Ac (exchangeable forms of the radionuclides) and 6M HNO_3 heated to 98°C during 2 hrs according to the standard radiochemical method (total content of the radionuclides in soil, including those in the UO_{2+x} and UO_2 particles) were carried out (Table 1).

Afterwards, a salt extraction of radionuclides in 2M NH_4Ac was performed. Each sample with added solution (1:10) was mixed intensively during 1 hr and then the mixtures were left for 1 day. Then, the standard radiochemical method was applied for determining the radionuclide activity concentration in solution.

Table 1. - Activity concentration of radionuclides in initial composite soil sample, extractions and in residue after extractions

Radionuclides	Activity concentration, kBq/kg			
	Initial composite soil representative sample (A_{total})	Acetate extractions by 2M NH_4Ac (A_{soil})	Acid extraction (6M HNO_3 heated to 98°C) ($A_{\text{UO}_2+\text{fixed of soil}}$)	Residue after extractions ($A_{\text{stable - U-Zr-O}}$)
^{90}Sr	118±14	31±6	38±8	38±8
^{137}Cs	264±26	38±8	-	19±2
^{154}Eu	0.76±0.15	0.13±0.03	0.37±0.07	0.22±0.03
^{241}Am	5.9±1.2	1.2±0.2	4.0±0.8	1.9±0.2

After filtration of the solution, hydroxides of high-valence metals (Fe, Al, Ti, Mn, Th, U) were extracted by adding ammonia without carbon into the solution. Then, after acidification, the solutions were left for 3 weeks in order to reach a balance between ^{90}Sr and ^{90}Y . At the end of this period, a stable isotope of yttrium was added to the solution and extracted by ammonia without carbon. Obtained precipitate was incinerated to Y_2O_3 and 1 day later analyzed by the beta-spectrometer.

[COMET]

14/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, _____
 characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016

Fraction of the chemically extra-stable fuel particles

For estimation of the fraction of chemically extra-stable particles (**U-Zr-O, $\Delta FP_1(t=29.4 \text{ year})$**) activity concentration of ^{90}Sr , ^{154}Eu and ^{241}Am in residue after extractions of the composite soil sample by 6 M HNO_3 heated to 98°C during 2 hrs were measured by means gamma and beta spectrometry (Table 1). Prolongation of the redissolved in 6M HNO_3 period up to 4 hrs did not result in any significant increase of the radionuclides (^{90}Sr , ^{137}Cs , ^{154}Eu and ^{241}Am) leaching into solution. Thus, it was assumed that the fraction of radionuclides leached from soil to solution by means of standard radiochemical methods (A_{stable}) should be rather small. Insoluble contaminated sediment components obtained after two stages extraction with 6M HNO_3 were attributed to chemically extra-stable fuel particles. Therefore, the activity fraction of extra-stable particles **$\Delta FP_1(t=29.4 \text{ year})$** is expressed as **$\Delta FP_1(t=29.4 \text{ year}) = A_{\text{stable-U-Zr-O}} / A_{\text{total}}$** (Table 1,2).

We assume that these inert particles consist of UZrO . X-ray microanalysis of such particles after extractions of the composite soil sample by 6 M HNO_3 heated to 98°C during 2 hrs should confirm this assumption at NMBU. It seems that such inert fuel particles hardly can be dissolved in soil under natural conditions and that the radionuclides associated with these particles will not have any significant influence on the radiological situation, although such entities can be retained by filtering organisms. However, the presence of these particles should be taken into account when we measure the total radionuclides activity in samples or when we develop technologies to deal with the radioactive waste containing such particles.

Table 2. Fraction of radionuclides present in different physico-chemical forms/FP types

Radionuclides	U-Zr-O particles: $\Delta FP_1(t=29.4 \text{ year})$	UO_2 particles: $\Delta FP_2(t=29.4 \text{ year})$	$1-\Delta FP_1-\Delta FP_2$
^{90}Sr	0.32±0.08	0.28±0.07	0.40±0.10
^{154}Eu	0.29±0.07	0.25±0.08	0.46±0.11
^{241}Am	0.32±0.07	0.23±0.07	0.45±0.10

Fraction of the UO_2 fuel particles

The UO_{2+x} particles have been completely dissolved in 29 years (radionuclides moved into the soil solution [16, 17]:

$$\Delta FP_3(t=29.4 \text{ year}) = \Delta FP_3(0) \cdot \exp(-k_3 \cdot t) = (0,21 \pm 0,15) \cdot \exp(-(0,28 \pm 0,05) \cdot 29.4) < 0,0005$$

To estimate the fraction of non-dissolved fuel particles (**UO_2 particles: $\Delta FP_2(t=29.4 \text{ year})$**) in waste material at the time of analysis, the method used was developed by UIAR and applied earlier for contaminated upper layer of soil [10, 11, 12, 16, 17]. The fraction of non-dissolved UO_2 fuel particles (ΔFP_2) in waste was determined using the data of the

[COMET]

exchangeable (A_{soil}) and the total soluble part of activity ($A_{\text{UO}_2+\text{ fixed of soil}}$) of ^{90}Sr , ^{154}Eu and ^{241}Am in the composite sample of soil (Table 1). By comparing the activity concentrations in soil of exchangeable forms of ^{90}Sr of Chernobyl fallout with that injected as water-soluble form (^{85}Sr) determination of the activity fraction associated with the matrix of non-dissolved UO_2 fuel particles could be attained. The exchangeable form of ^{85}Sr in the composite sample of soil from the trench no. 22 was 85 % (in 2000-2001 and 2015). At each stage, balance calculations of ^{90}Sr , ^{137}Cs , ^{154}Eu and ^{241}Am activities in the extract and solid residues were carried out by means of gamma and beta spectrometry.

$\Delta\text{FP}_2(t=29.4 \text{ year})$ were calculated as (Table 1, 2):

$$\text{For } ^{90}\text{Sr } \Delta\text{FP}_2^{\text{Sr}}(t=29.4 \text{ year})=(A^{\text{Sr}}_{\text{UO}_2+\text{ fixed of soil}}- A^{\text{Sr}}_{\text{soil}}\cdot\mathbf{15/85})/A^{\text{Sr}}_{\text{total}}=0.28\pm 0.07$$

Were $\mathbf{15/85}$ =activity of fixed form of ^{85}Sr /activity of exchangeable form of ^{85}Sr in soil

$$\text{For } ^{154}\text{Eu } \Delta\text{FP}_2^{\text{Eu}}(t=29.4 \text{ year})=(A^{\text{Sr}}_{\text{UO}_2+\text{ fixed of soil}}- A^{\text{Sr}}_{\text{soil}}\cdot\mathbf{15/85})/173/A^{\text{Eu}}_{\text{total}}=0.25\pm 0.08$$

Were 173=activity ^{90}Sr /activity ^{154}Eu in Chernobyl FP for $t=29.4$ year [3]

$$\text{For } ^{241}\text{Am } \Delta\text{FP}_2^{\text{Am}}(t=29.4 \text{ year})=(A^{\text{Sr}}_{\text{UO}_2+\text{ fixed of soil}}- A^{\text{Sr}}_{\text{soil}}\cdot\mathbf{15/85})/24/A^{\text{Am}}_{\text{total}}=0.23\pm 0.07$$

Were 24= activity ^{90}Sr /activity ^{241}Am in Chernobyl FP for $t=29.4$ year [3]

Results of the described experiments showed that in 2001-2002 fraction of ^{90}Sr activity associated with the FP of various types was $64\pm 13\%$ ($20\pm 10\%$ activity in the chemically extra-stable U-Zr-O fuel particles) of its initial activity in the trench [16, 17]. An alternative approach to estimate the FP-associated fractions of radionuclides in the trench RAW involved sedimentation of a sample sest in heavy liquid and calculation of the activity balance after isolation of FP. According to the results $\approx 65\text{-}70\%$ of the activity of the mentioned radionuclides were associated with the FP matrix, which is in a good agreement with the above estimate [16, 17]. The relative (%) fraction of the ^{90}Sr and ^{154}Eu activity leached from fuel particles in 2001-2002 was about 36 % (Fig.5)

The fraction of the ^{90}Sr , ^{154}Eu and ^{241}Am activity leached from fuel particles in 2015 ($1-\Delta\text{FP}_1-\Delta\text{FP}_2$) were 0.40 ± 0.10 , 0.46 ± 0.11 and 0.45 ± 0.10 , respectively (Table 2 and Fig.5). We have obtained similar values for different radionuclides, which are in good agreement with the theoretical predictions obtained for more than 15 years ago as seen in Fig.5 [16, 17].

In practice, the experimental activity ratio $^{90}\text{Sr}/^{154}\text{Eu}$ in the RAW of the trench was 67 (in 2000), which showed depletion of ^{90}Sr in the RAW. The aforementioned ratios, 72 and 67, allowed the radiostromium activity fraction that had migrated from the trench in the period from 1987 to 2000 to be evaluated $7\pm 5\%$ [16, 17]. This result is in good agreement with the fraction of leached ^{90}Sr derived from characterizing the radionuclide plume in the aquifer downstream from the trench considering radionuclides activity in different physico-chemical forms in 2015 (Table 2).

[COMET]

16/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, _____
 characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016

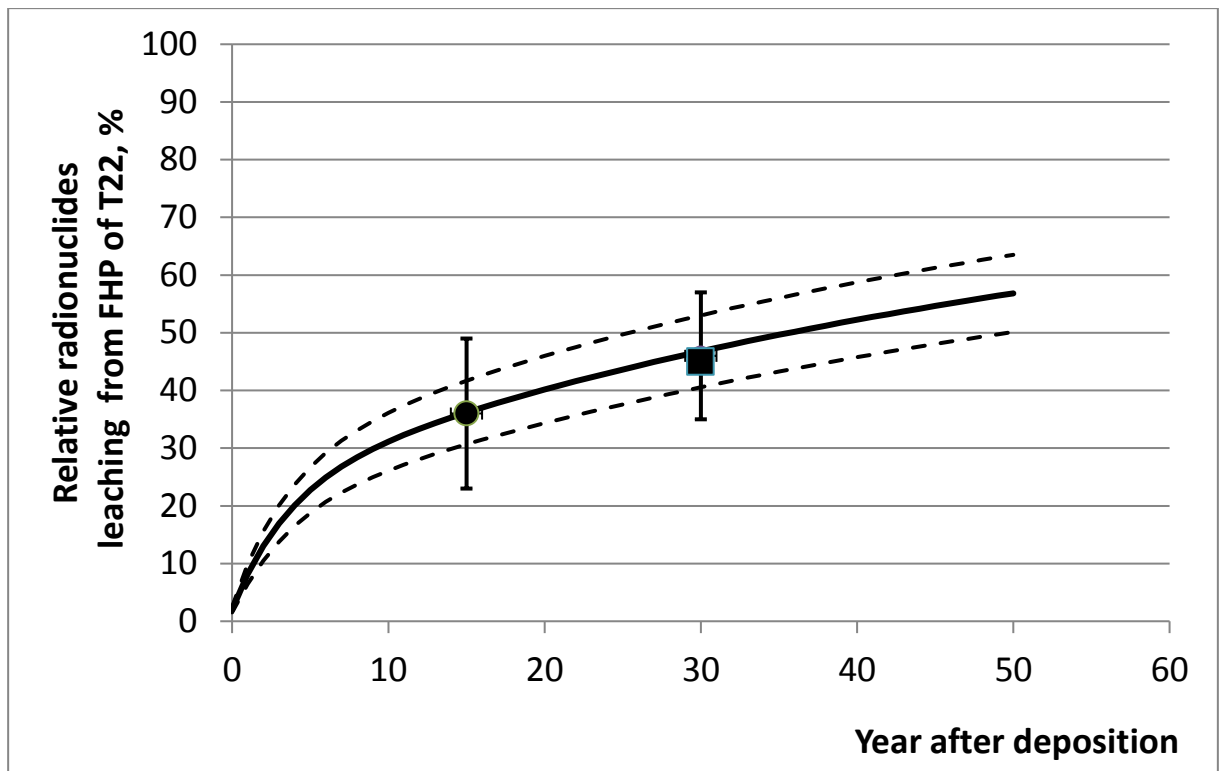


Fig.5 – Relative (%) fraction of ^{154}Eu and ^{241}Am activity of the leached from fuel particles in 2001-2002 and 2015 (solid line - theoretical dependence [16, 17])

Conclusion

In order to characterize the trench source term, the parameters of the FP dissolution kinetics in the radioactive waste trench were estimated and the model of radionuclides mobile forms dynamics was tested.

In the years 2015, the robustness of the models of Chernobyl fuel particles dissolution, elaborated 15 years ago, have been verified predicting the dynamics of ^{90}Sr , ^{154}Eu and ^{241}Am activity to fuel particles in the radioactive waste trench. The theoretical/modeling estimations of 2000-2001 of exchangeable fraction of radionuclides activity in soils coincide with the experimental results obtained in 2015. The results show that the activity of the mobile form of ^{90}Sr in the trench has presently reached the maximum value.

The results will be prepared for publication by NUBiP, IRSN and NMBU and the article submitted to Journal of Environmental Radioactivity.

[COMET]

17/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016

References

1. Kuriny V.D., Ivanov Yu.A., Kashparov V.A., Loschilov N.A., Protsak V.P., Yudin E.B., Zhurba M.A., Parshakov A.E. Particle Associated Chernobyl Fall-Out in the Local and Intermediate Zones. // *Annals of Nuclear Energy*. -1993, v.20, N.6, p.415-420.
2. Kashparov V.A., Lundin S.M., Khomutinin Yu.V., Kaminsky S.P., Levchuk S.E., Protsak V.P., Kadygrib A.M., Zvarich S.I., Yoschenko V.I., Tschiersch J. Soil contamination with ⁹⁰Sr in the near zone of the Chernobyl accident // *Journal of Environment Radioactivity*, v.56, № 3, 2001, p.285-298.
3. Kashparov V.A., Lundin S.M., Zvarich S.I., Yoschenko V.I., Levchuk S.E., Khomutinin Yu.V., Maloshtan I.N., Protsak V.P. Territory contamination with the radionuclides representing the fuel component of Chernobyl fallout // *The Science of The Total Environment*, vol.317, Issues 1-3, 2003, pp. 105-119.
4. Bugai D., Dewiere L., Kashparov V., Ahamdach N., Strontium-90 transport parameters from source term to aquifer in the Chernobyl Pilot Site. // *Radioprotection – Colloques*, 2002, vol. 37-C1. pp.11-16.
5. Dewiere L., Bugai D., Grenier C., Kashparov V., Ahamdach N. ⁹⁰Sr migration to the geo-sphere from a waste burial in the Chernobyl exclusion zone // *Journal of Environmental Radioactivity*, v.74, Issue 1-3, 2004, p.139-150.
6. Levchuk S., Kashparov V., Maloshtan I., Yoschenko V., Van Meir N. Migration of transuranic elements in groundwater from the near-surface radioactive waste site// *Applied Geochemistry*, v. 27, Issue 7, 2012, p. 1339–1347
7. Van Meir N., Bugai D., Kashparov V. The experimental platform in Chernobyl: an international research polygon in the exclusion zone for soil and groundwater contamination // *Radioactive Particles in the Environment, NATO Science for Peace and Security Series C: Environmental Security*, ed. by D.Oughton, V. Kashparov, Published by Springer, the Netherlands 2009, p. 197-208.
8. Bugai D., Kashparov V., Dewiére L., Khomutinin Yu., Levchuk S., Yoschenko V. Characterization of subsurface geometry and radioactivity distribution in the trench containing Chernobyl clean-up wastes // *Environmental Geology*, v.47, 2005, p.869-881.
9. Kashparov V., Yoschenko V., Levchuk S., Bugai D., Van Meir N., Simonucci C., Martin-Garin A. Radionuclide migration in the experimental polygon of the Red Forest waste site in the Chernobyl zone – Part 1: Characterization of the waste trench, fuel particle transformation processes in soils, biogenic fluxes and effects on biota // *Applied Geochemistry*, v. 27, Issue 7, 2012, p. 1348–1358
10. Kashparov V.A., Oughton D.H., Zvarich S.I., Protsak V.P., Levchuk S.E. Kinetics of fuel particle weathering and ⁹⁰Sr mobility in the Chernobyl 30-km exclusion zone // *Health Physics*. - 1999. - Vol.76, N.3. - P.251-259.
11. Kashparov V.A., Ahamdach N., Zvarich S.I., Yoschenko V.I., Maloshtan I.N., Dewiere L. Kinetics of dissolution of Chernobyl fuel particles in soil in natural conditions. // *Journal of Environmental Radioactivity*, v.72, Issue 3, 2004, p.335-353.
12. Kashparov V.A., Protsak V.P., Ahamdach N., Stammose D., Peres J.M., Yoschenko V.I., Zvarich S.I. Dissolution kinetics of particles of irradiated Chernobyl nuclear fuel: influence of pH and oxidation state on the release of radionuclides in contaminated soil of Chernobyl // *Journal of Nuclear Materials*, v. 279, 2000, p.225-233.

[COMET]

18/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016

-
13. Salbu B., Krekling T., Lind D.H., Oughton D.H., Drakopoulos M., Simionovichi A., Snigireva I., Snigirev A., Weitkamp T., Adams F., Janssens K., Kashparov V. High energy X-ray microscopy for characterization of fuel particles // Nuclear Instruments and Methods in Physics Research A 467-468, 2001, p.1249-1252.
 14. Kashparov V.A., Ivanov Yu.A., Zvarich S.I., Protsak V.P., Khomutinin Yu.V., Kurepin A.D., Pazukhin E.M. Formation of Hot Particles During the Chernobyl Nuclear Power Plant Accident. //Nuclear Technology. - 1996, v.114, N.1, p.246-253.
 15. Zhurba M., Kashparov V., Ahamdach N., Salbu B., Yoschenko V., Levchuk S. The “hot particles” data base //Radioactive Particles in the Environment, NATO Science for Peace and Security Series C: Environmental Security, ed. by D.Oughton, V. Kashparov, Published by Springer, the Netherlands 2009, p. 187-195.
 16. Kashparov V., Yoschenko V., Levchuk S., Bugai D., Van Meir N., Simonucci C., Martin-Garin A. Radionuclide migration in the experimental polygon of the Red Forest waste site in the Chernobyl zone – Part 1: Characterization of the waste trench, fuel particle transformation processes in soils, biogenic fluxes and effects on biota // Applied Geochemistry, v. 27, Issue 7, 2012, p. 1348–1358
 17. Kashparov V., Ahamdach N., Levchuk S., Yoschenko V., Fesenko S., Maloshtan I. Dissolution of particles of irradiated nuclear fuel in the temporary storages of radioactive waste in Chernobyl zone: sources for radionuclides migration //Radioactive Particles in the Environment, NATO Science for Peace and Security Series C: Environmental Security, ed. by D.Oughton, V. Kashparov, Published by Springer, the Netherlands 2009, p. 139-156.

[COMET]

19/19

(IRA- Particle Behaviour –D2) Set of parameters for FP dissolution in the radioactive waste trench, _____
characterization of FP matrix.

COMET Particle Behaviour Group (Initial Research Activity)

Dissemination level: CO

Date of issue of this report: 08/09/2016