STCU Project # 4207

Long-term prognosis of behavior of the fuel dust in Chernobyl Shelter

(proposal)

CONTENT

- **Objectives of new Project #4207**

- **Physical-chemical characteristics of the fuel particles**

- **Tasks of the Project #4207**

- **Interface with ISTC Project #3702** "Long-term behavior of corium after the accident (using the data of the Chernobyl NPP accident)"

Participating Institutions

- Ukrainian Institute of Agricultural Radiology of National Agricultural University of Ukraine

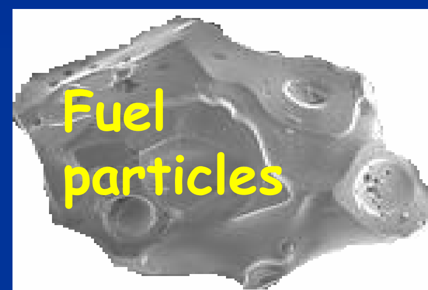
- Institute of Safety Problems of Nuclear Power Plants of National Academy of Sciences of Ukraine

Project duration	30 months;
Participating personnel	19 (13 “weapon” scientists);
Estimated cost	300,000 USD

What is a scope of research ?

Steam component

Noble gases, isotopes of I and Cs,
to some extent – isotopes of Sr and Ru



Condensation at the inert carriers

Processes leading to destruction and leaching the radionuclides from FP

Radioactive substances with unknown physical-chemical characteristics

Significance of the project

- Various authors estimate the fuel dust inventory in Shelter as 500 to 2000 kg. Approx 98 % of the irradiated nuclear fuel of the ChNPP unit 4 is presented by various FCMs
- Analysis of the available technologies shows that removal of FCMs from Shelter can take 50-100 years
- FCMs are subjected to destruction because of the external and internal factors. As a result, the new compounds and radioactive dust are formed
- Dust resuspension and transport in Shelter can be caused not only by the accidents, but by any actions implying the personnel translocations (even inspections of the rooms etc)
- Inhalation of the fuel dust is very dangerous in concern of the internal irradiation

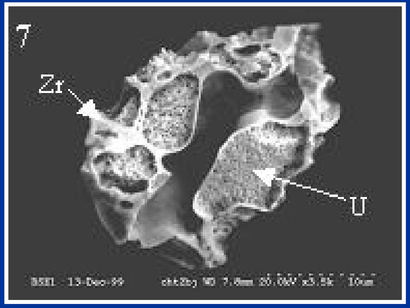
The project aims

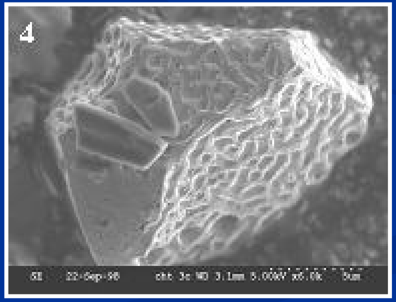
The project aim is creation of a model for the long-term (for 50-100 years) prognosis of the fuel dust behavior in Shelter. The model will describe

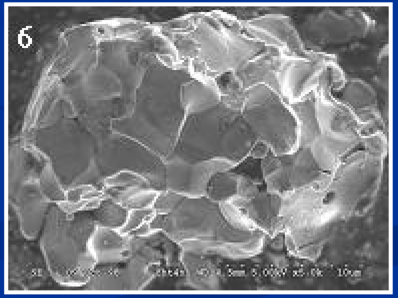
- Transformation of the existing fuel particles
- Formation of the new fuel particles from various FCMs in the Shelter conditions

Physical-chemical characteristics of the fuel particles and mechanisms of their formation during the Chernobyl accident

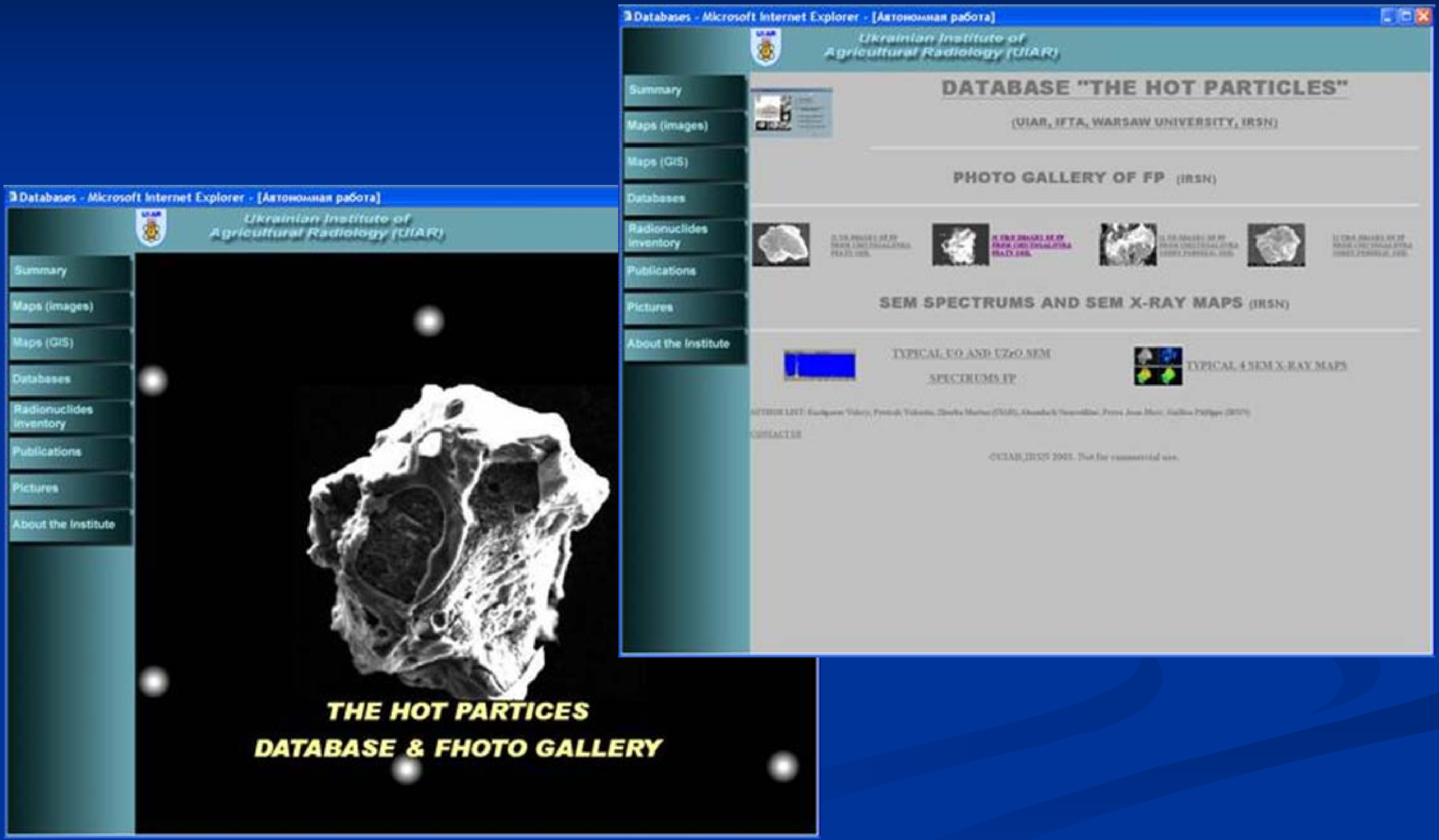
During 1987-1989, more than 1200 relatively large (size > 10 μm, activity > 100 Bq) Chernobyl hot particles were selected by scanning thin soil layers with a dosimeter, and about 500 additional hot particles were selected in the period 1989-1996.



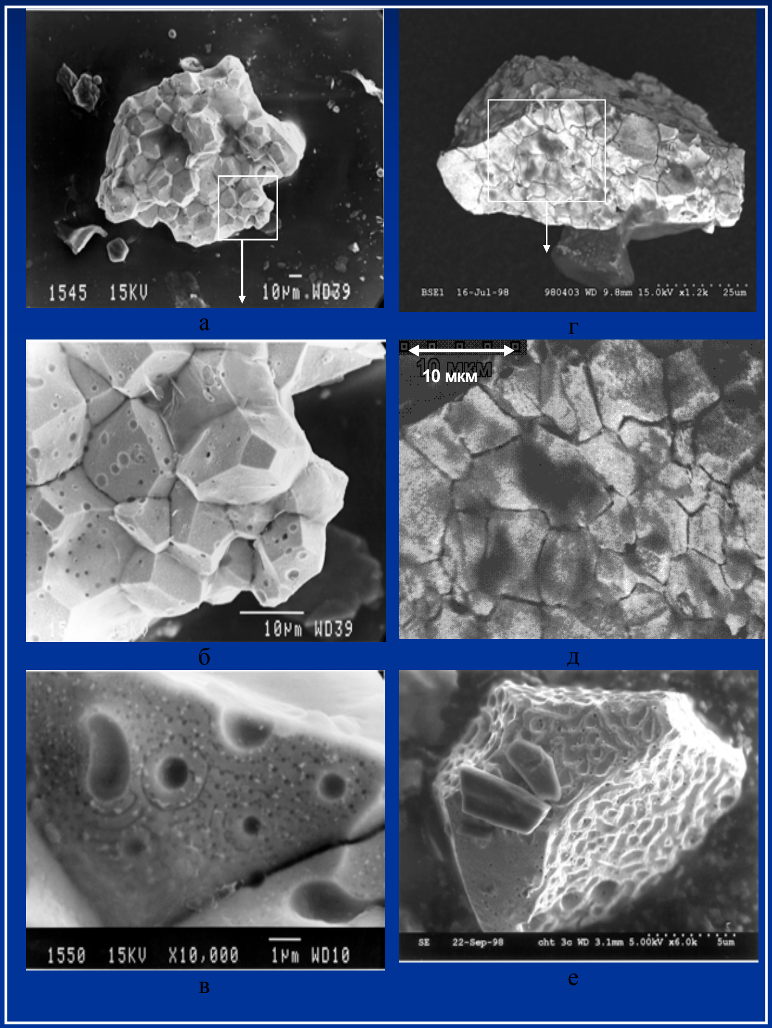


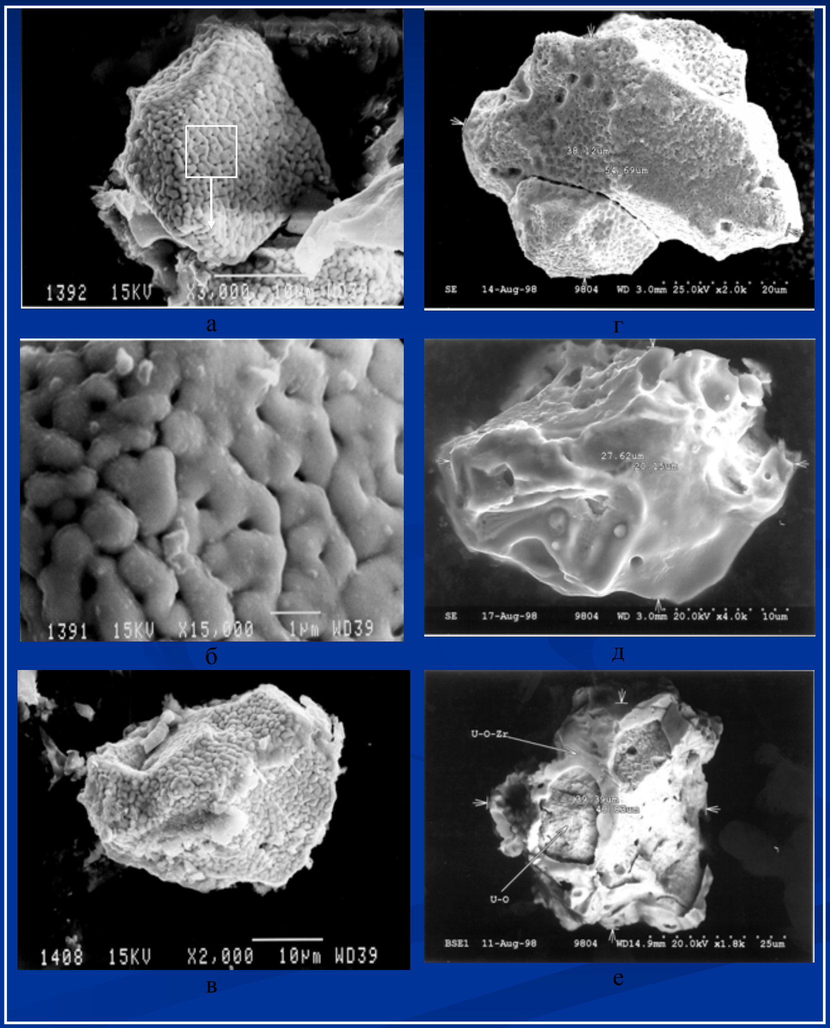


The Database “Hot Particles”

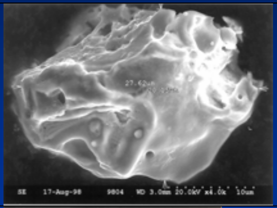
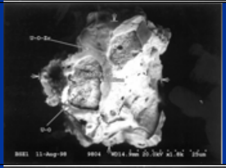
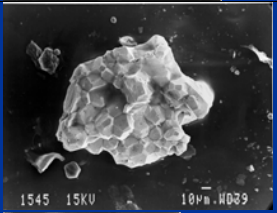
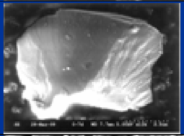
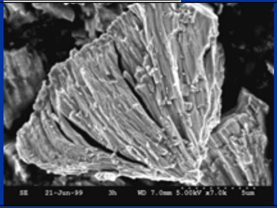


Typical fuel particles collected by UIAR (SEM by NLH, Norway, and IPSN, France)



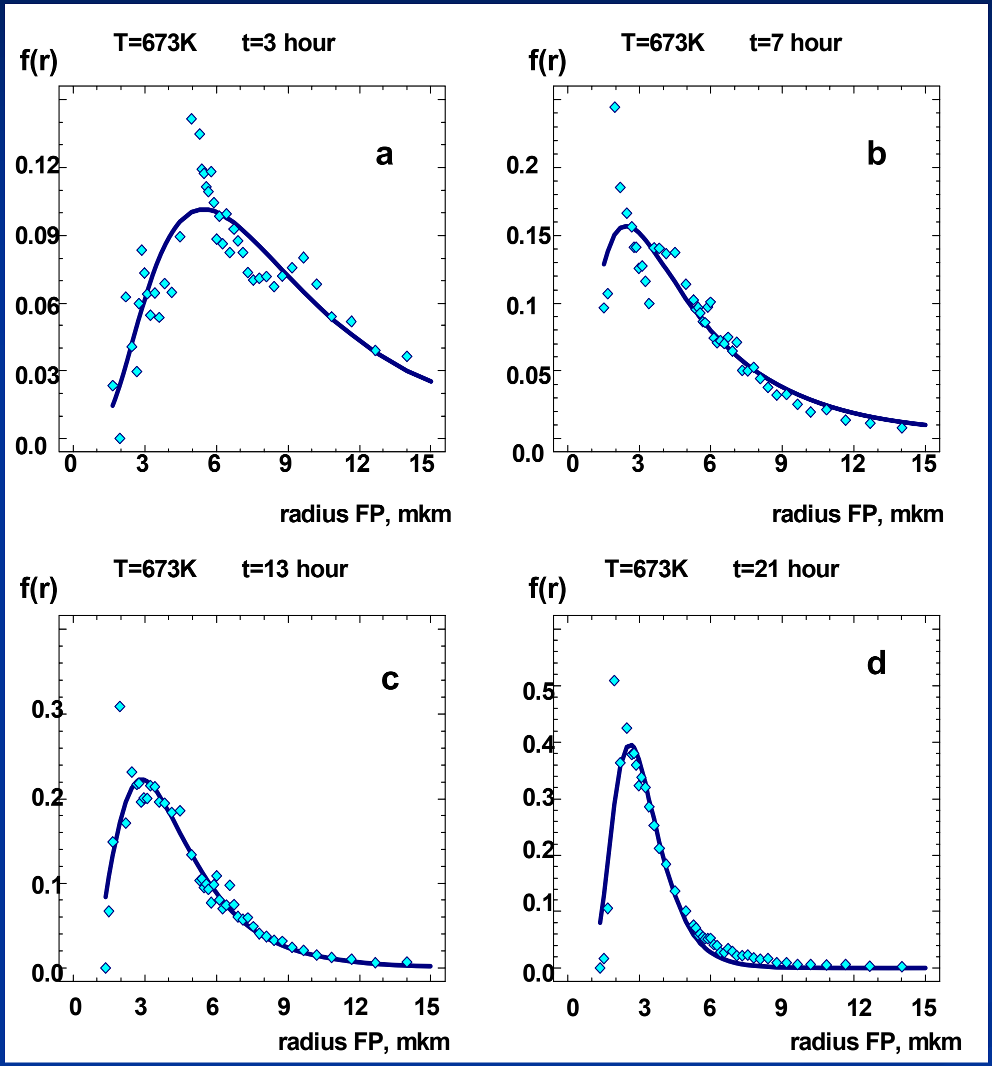


3 tyoes of FP according to their dissolution rates under natural conditions:

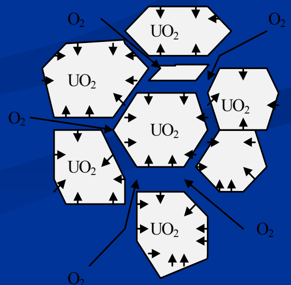
	**U-Zr-O** **Super stable**
	
	**UO_2** **Stable**
	
	**UO_{2+x}** **Non-stable**

- chemically extra-stable particles (U-Zr-O). These particles were formed at the first moment of the accident on 04/26/86 and were deposited within the narrow western trace;
- non-oxidized chemically stable fuel particles (UO2) of the first release (04/26/86), formed as a result of the mechanical destruction of nuclear fuel. These particles also were deposited along the narrow western trace of fallout;
- chemically low stable particles (UO2+x), formed as a result of oxidization of the nuclear fuel in the period 04/26/86-05/05/86. These particles were predominantly deposited in the northern and southern traces of fuel fallout.

For better understanding of the processes in the accidental unit and for estimation of the dispersal composition of the released fuel particles, the experiments have been performed which simulated the FP formation during the accident



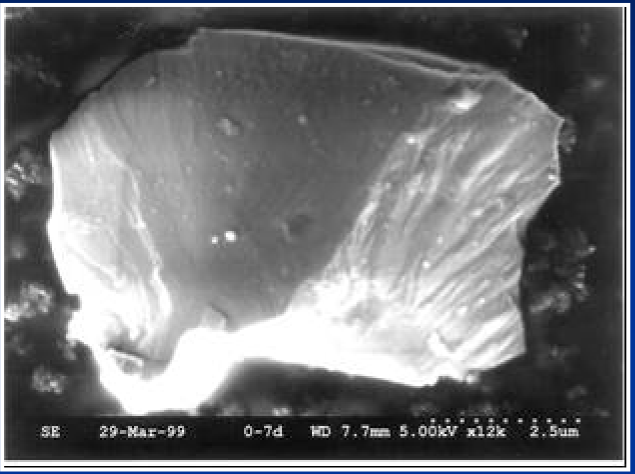
Parameters of FPs dispersal composition were obtained. Median diameter of FPs does not depend on the temperature of annealing and decreases to the fuel grain (crystallite) size (~6 μm) with prolongation of the annealing period. Taking into account the dispersal and matrix composition of the real Chernobyl FPs, one can infer that oxidization of the nuclear fuel was one of the principal processes leading to formation of FPs during the Chernobyl accident

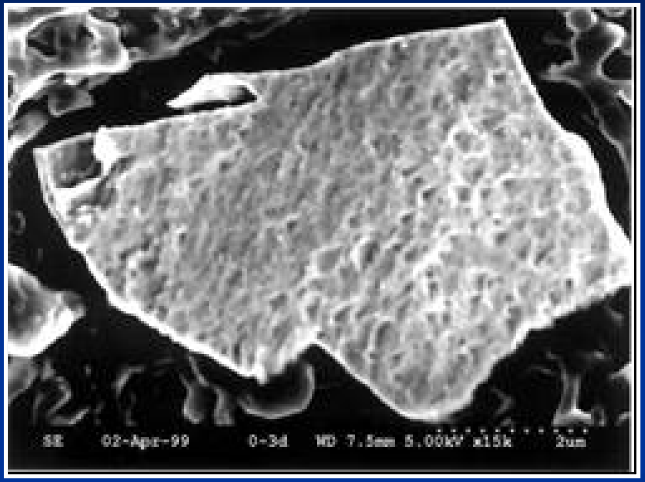


Fuel particles dissolution in the fluids at various pH

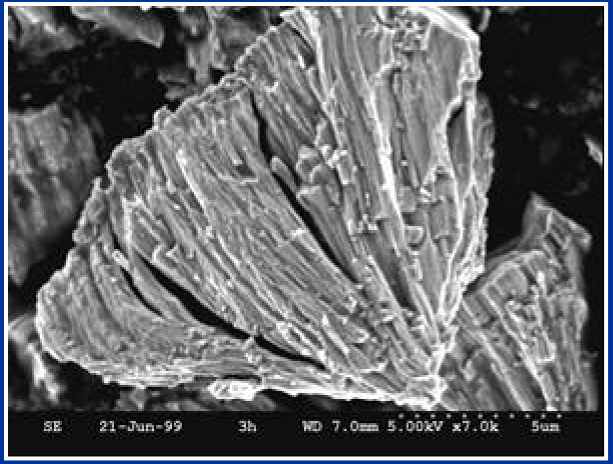
Before dissolution

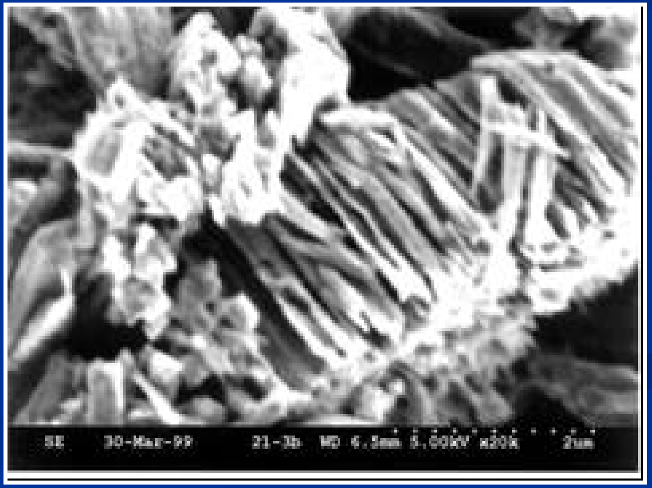
After dissolution





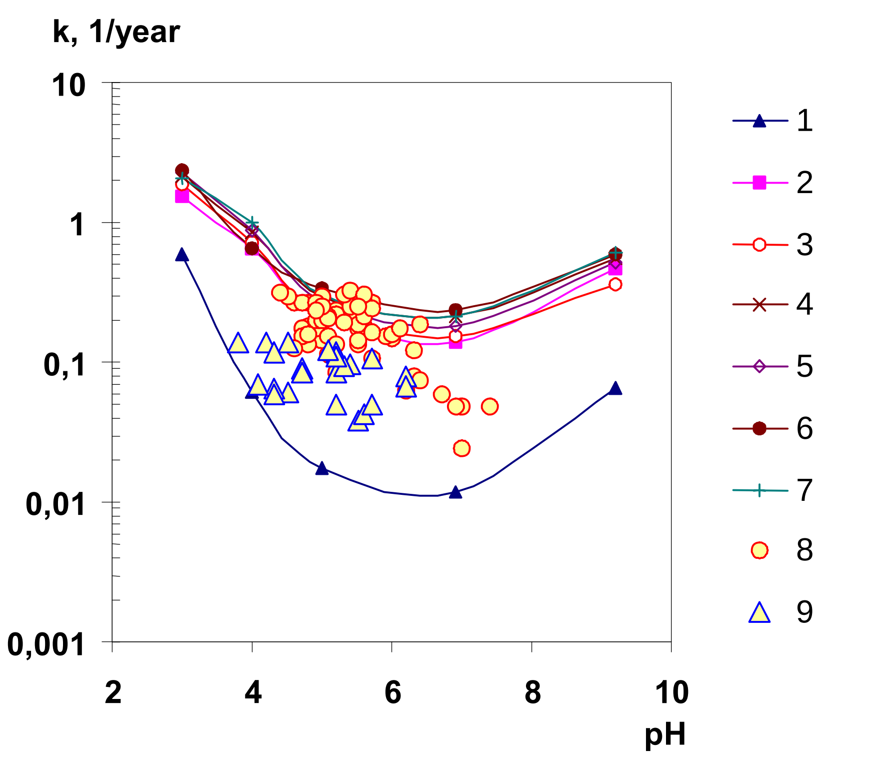
UO_2





UO_{2+x}

Chernobyl fuel particles dissolution kinetics



$dA/dt = -(k+\lambda)\cdot A$,
$\Delta FP = A(t)/A0 \cdot \exp(-\lambda t) = \exp(-kt)$,
A0 and A(t) – radionuclide initial activity in FP and activity remaining at the moment t after the beginning of dissolution, respectively;
k – FP transformation constant, $year^{-1}$;
λ - radioactive decay constant, $year^{-1}$

FP transformation constant vs pH of solution:

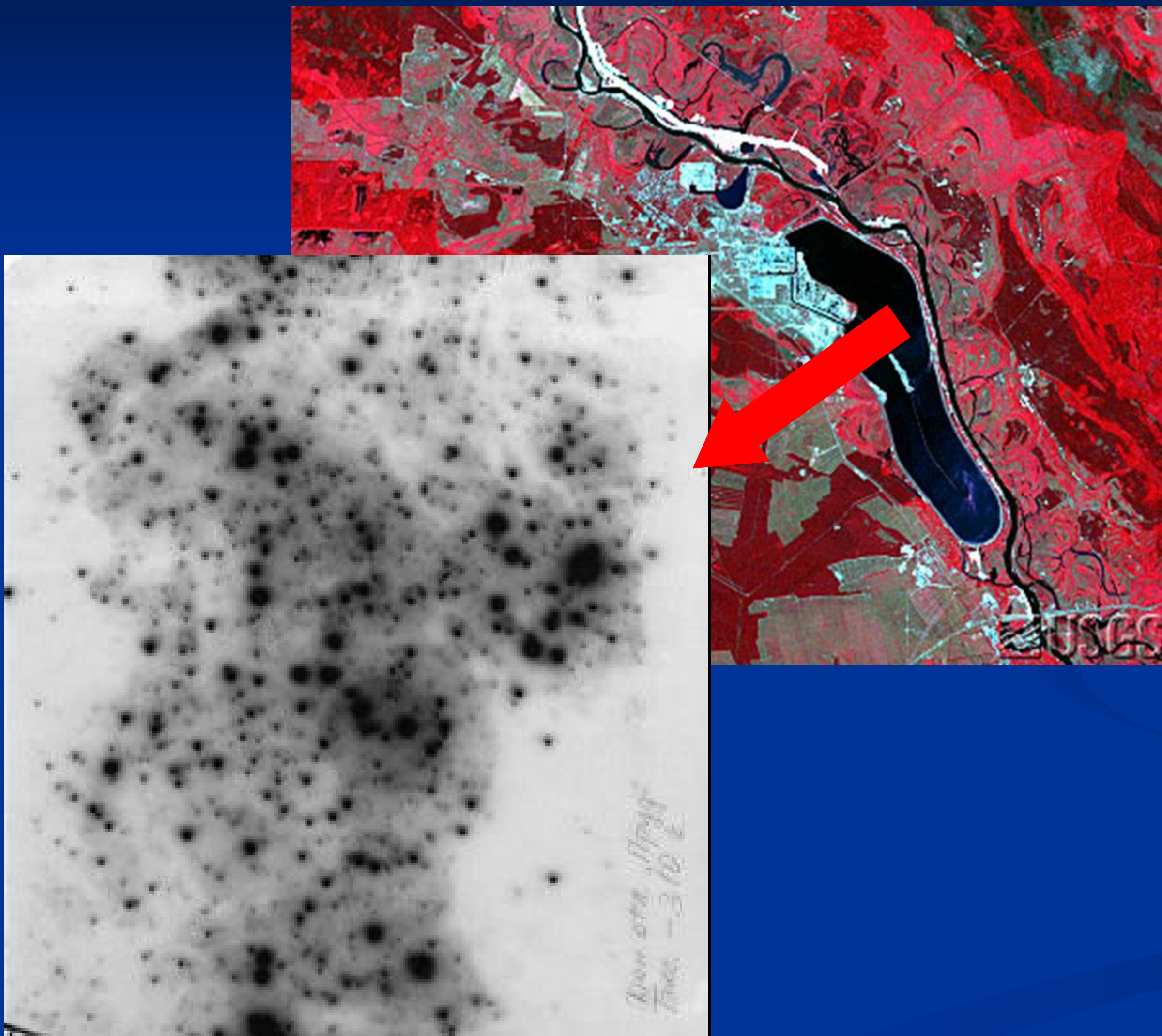
1 – non-annealed FPs; 2 – 1 hr annealing; 3 – 3 hr annealing; 4 – 5 hr annealing; 5 – 7 hr annealing; 6 – 13 hr annealing; 7 – 21 hr annealing;

and vs pH of soil:

8 – Chernobyl FPs from the Northern and Southern traces;

9 – Chernobyl FPs from the Western trace

FPs in the bottom sediments of the ChNPP cooling pond



- ^{90}Sr activities in the sediment samples were found equal if measured by autoradiography (i.e. in FPs) and by radiochemistry (total sample activity)

- In bottom sediments
$^{90}Sr/^{154}Eu=80\pm9$
- In the nuclear fuel (and FPs)
$^{90}Sr/^{154}Eu=76\pm5$

- ^{85}Sr exchangeable form
$98\pm1\%$
- ^{90}Sr exchangeable form

$0,7\pm0,1\%$

Our recent publications:

- *Kashparov V.A., Lundin S.M., Zvarich S.I., Yoschenko V.I., Levtchuk 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.*

- *Kashparov V.A. Hot Particles at Chernobyl //Environmental Science and Pollution Research, v.10 Special (1), 2003, pp.21-30.*

- *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.*

- *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.*

- *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.*

- *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, N1.- pp.246-253.*

The tasks planned
1st task

- Update the review of particles data by the recently obtained data and identify gaps in knowledge of their formation
- Examination of formation mechanisms of Chernobyl FP, physical-chemical characteristics & CFP classification.
- This will include analysis of the fuel samples & CFP, FCMs and sediments collected from the basement of the Shelter (in collaboration with Kurchatov's team of CHESS-2 Project #3702 "Long-term behavior of corium after the accident (using the data of the Chernobyl NPP accident")

2^{nd} task

- Research into the behavior of radioactive aerosols in Shelter: the release of radioactive aerosols from Shelter, their radionuclide and dispersal composition. Establishing the links between the lava and fuel sources and the aerosol formation.
- The aerosol sampling will be important for evaluation of the aerosol formation source.
- There will be sampling of the sub-micron aerosols since this is an important component of the aerosols.

These data will have the direct application to estimating the source term in Shelter during the new Shelter creation

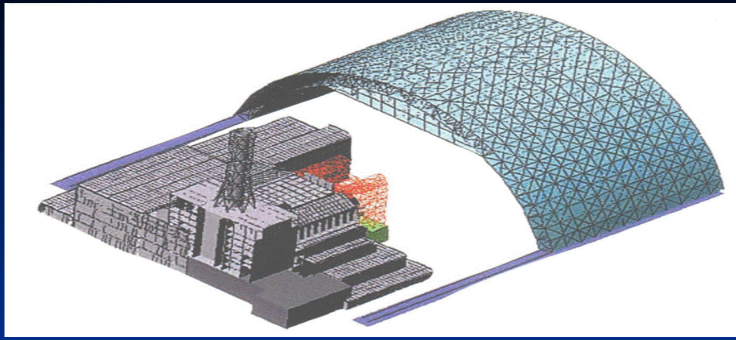
3rd task

- Experimental studies of CFP destruction rate as a function of particle characteristics: matrix composition, oxidation level, and as a function of the medium: humidity, temperature, pH.
- As the medium, we can use both actual water from Shelter and the fluids simulating it
- The medium pH will be alkaline (water in Shelter is in contact with concrete of the basement), with either acid (groundwater) or neutral (control) water for comparison.
- The fate of key radionuclides such as ^{90}Sr (as $SrCO_3$) and the leaching rates of U and Pu from the particles will be some of the essential information expected from this work.

4^{th} task

- Creation of the model predicting the long-term transformation of FPs in Shelter
- The model will describe the CFPs destruction, evolution of their dispersal composition and leaching of the radionuclides

5th task



- Creation of the long-term prognosis of behavior and transformation of the fuel dust as a most dangerous radioactive material in Shelter
- To complete this task we are going to utilize both the results of our project and the data by Russian team (Kurchatov's Institute) obtained within the frameworks of CHESS-2 ISTC Project #3702

AGREEMENT ABOUT THE SCIENTIFIC AND TECHICAL COLLABORATION BETWEEN THE PARTICIPANTS OF THE ISTC PROJECT #3702 (CHESS-2, RUSSIA) AND STCU PROJECT #4207 (UKRAINE)

Taking into account that the two projects are related by the common tasks and research methods, we intend to continue and to extend our collaboration by means of

- Coordination of the detail working plans at their preparation phase;
- Exchanging the current information, calculation results and experimental data during the Projects realization;
- Coordination of the presentations and published materials;
- Organizing twice a year the joint scientific meetings and invitation of the representative managers of the ISTC, STCU and interested institutions;
- Publishing the joint conclusions after the Projects completion.

Thank you very much for your attention