Study of Chernobyl "lava", corium and hot particles:

experience of V.G. Khlopin Radium Institute (KRI)

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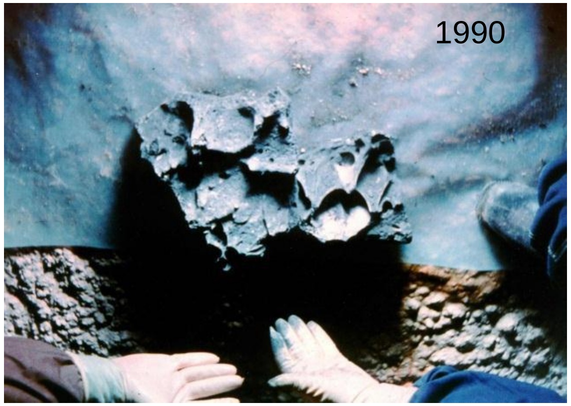


160 employees of V.G. Khlopin Radium Institute have been working in Chernobyl since 1986 till 1992

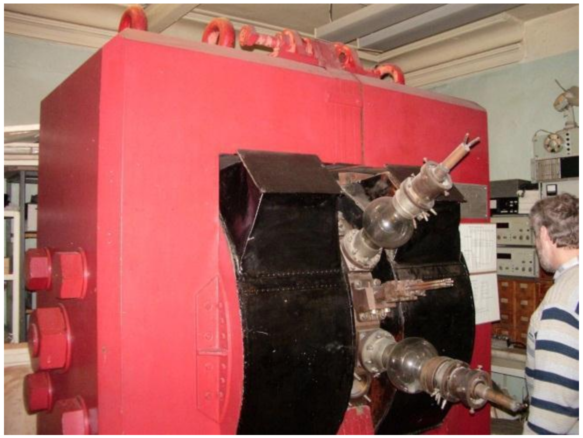
V.G. Khlopin Radium Institute (KRI)

study of radioactive materials since 1922

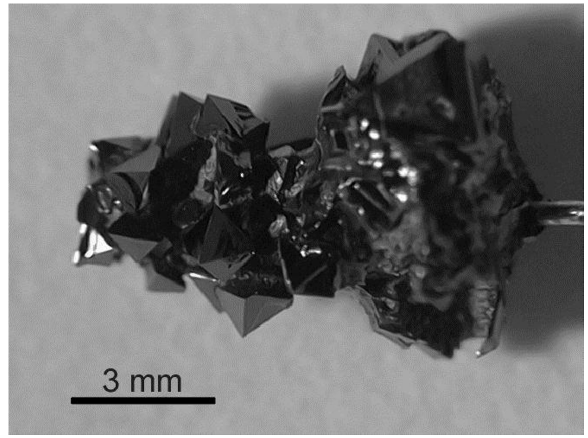
at present time KRI is a research institute of Russian State Corporation for Atomic Energy (ROSATOM)

- First European cyclotron built in 1937
- First sample of Soviet **Pu**, obtained in 1945
- Industrial Soviet technology of **Pu** extraction 1945-1949
- Chernobyl investigation 1986-1992 *(and present time)*
- *Unique collection of Chernobyl "lava", corium and hot particles available for international research and training*
- Hot-cell facility for research using any kind of spent nuclear fuel and liquid HLW
- Study of actinide-doped ceramics and HLW glasses
- Production of isotopes
- Radio-ecological monitoring
- *Applied training of international young scientists in the field of nuclear waste management*

V.G. Khlopin Radium Institute (KRI)

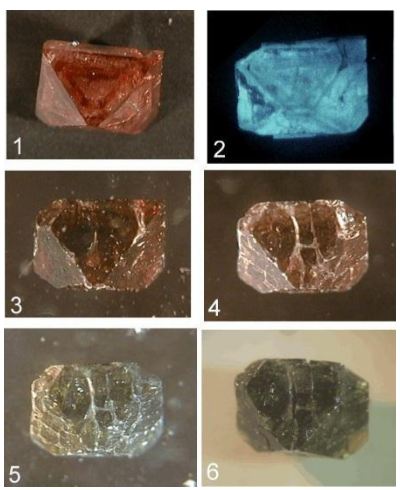


First European cyclotron (1937)

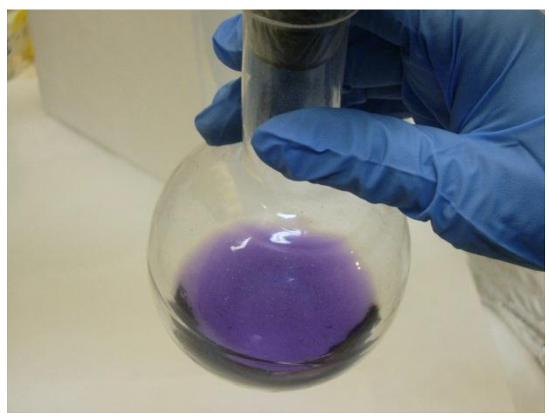


Crystals of NpO_2

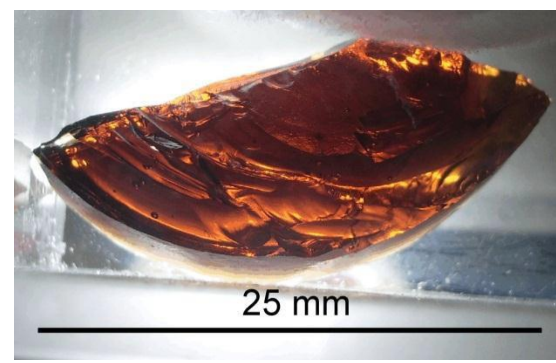
Hot-cell facility



Radiation damage effects in zircon crystal doped with Pu-238



Solution of $PuCl_3$



Borosilicate glass doped with Pu-238

Background
basic papers

1. Chernobyl: **The Soviet Report**. Nuclear News, Vol.29, #13, Oct. 1986.
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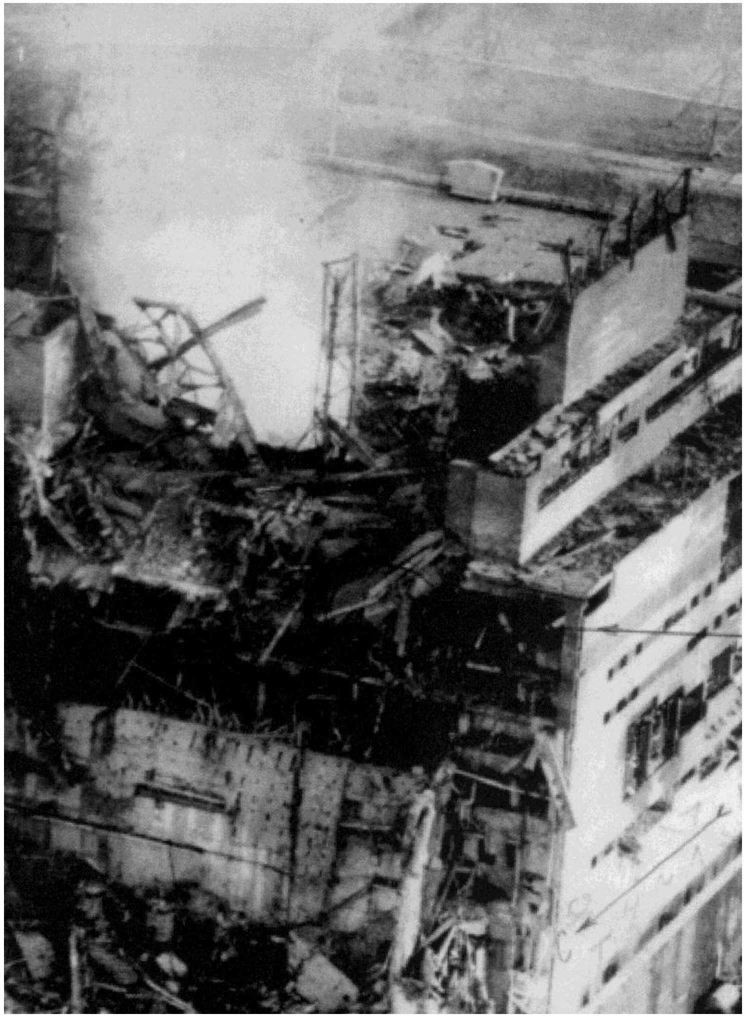
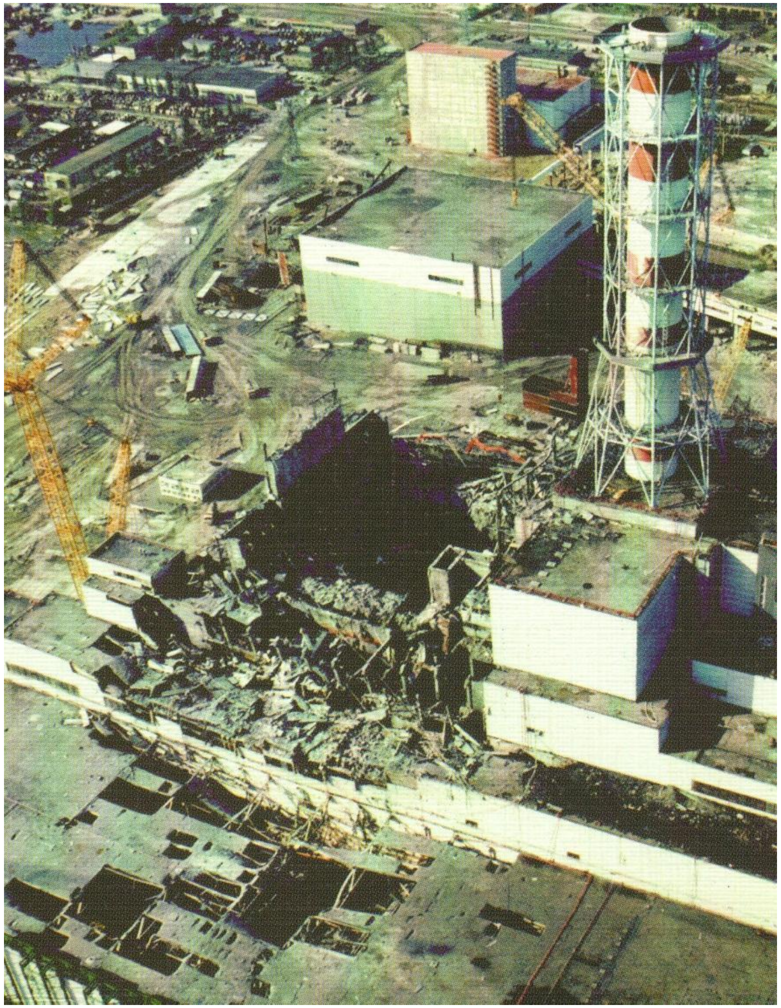
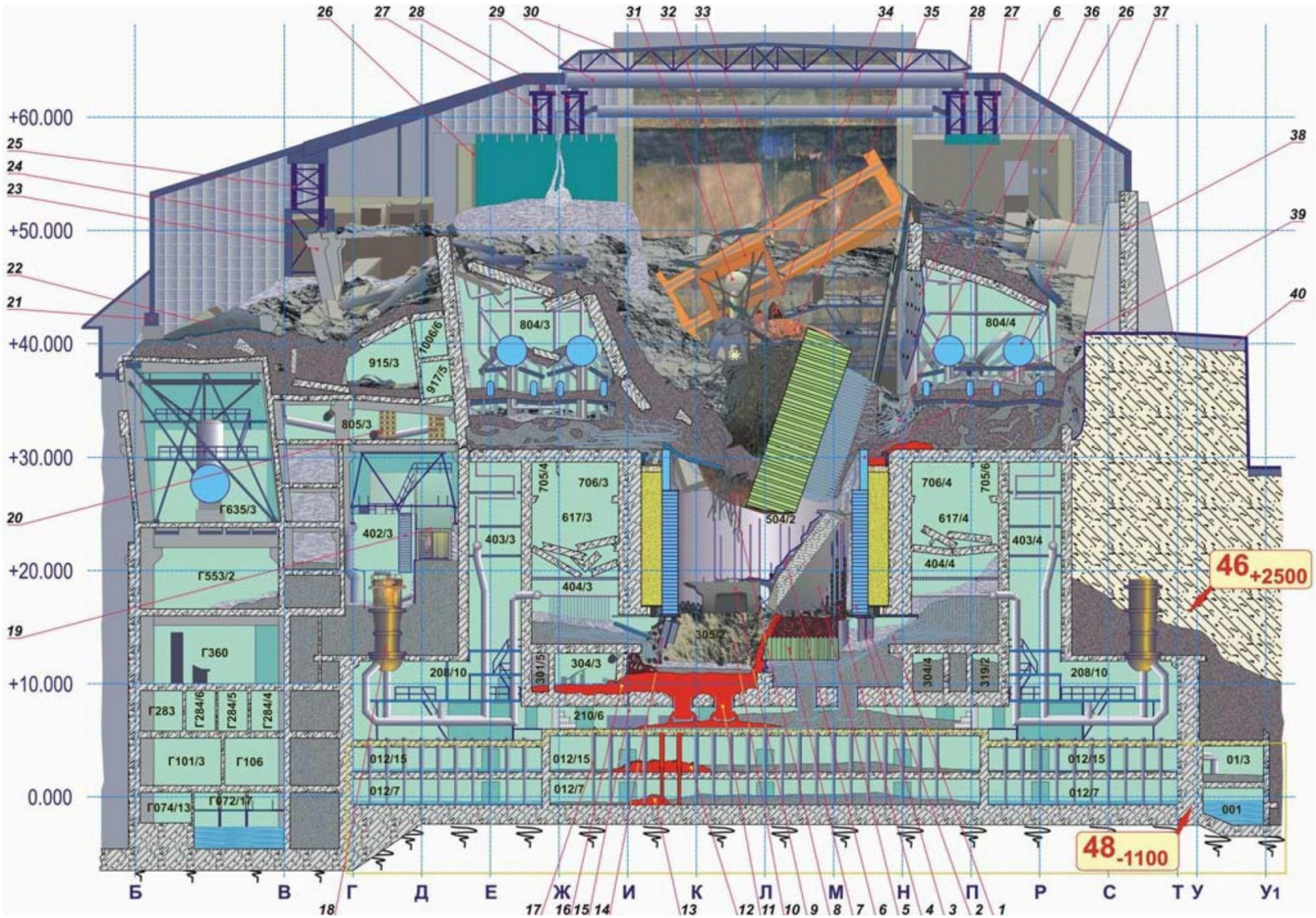
4. Лебедев И.А., Мясоедов Б.Ф., Павлоцкая Ф.И., Френкель В.Я. **Содержание плутония в почвах европейской части страны после аварии на Чернобыльской АЭС**. Атомная Энергия, т.72, вып.6, июнь 1992, с. 593-598. – *Lebedev I.A., Myasoedov B.F., Pavlotskaya F.I., Frenkel V.Ya.* ***Plutonium contents in the soils of European part of USSR after accident at Chernobyl NPP****. Atomic Energy, Vol.72, #6, June 1992, pp. 593-598 (in Russian).*
5. Киселев А.Н., Ненаглядов А.Ю., Сурин А.И., Чечеров К.П. **Экспериментальные исследования лавообразных топливосодержащих масс (ТСМ) на 4-м блоке ЧАЭС (по результатам исследований 1986-1991 годах)**. Препринт ИАЭ, Москва 1992 – *Kiselev A.N., Nenaglyadov A.Yu., Surin A.I., Checherov K.P.* ***Experimental study of lava-like fuel containing masses (FCM) at 4th Unit of ChNPP (based on results obtained in 1986-1991)****. Issue of IAE, Moscow 1992 (in Russian).*
6. Trotabas M., Blanc J-Y., Burakov B., Anderson E., Duco J. **Examination of Chernobyl samples. Impact on the accident scenario understanding**. Report DMT/92/309, SETIC/LECR-92/36, Report IPSN/93/02, Report RI-1-63/92, March 1993.
7. Pazukhin E.M., Fuel-containing lavas of the Chernobyl NPP 4th block – topography, physicalchemical properties, formation scenario. Radiochemistry **36**(2) (1994) 109-154.
8. Burakov B.E., Anderson E.B., Shabalev S.I., Strykanova E.E., Ushakov S.V., Trotabas M., Blanc J-Y., Winter P., Duco J. **The Behaviour of Nuclear Fuel in First Days of the Chernobyl Accident**. *Mat. Res. Soc. Symp. Proc. Scientific Basis for Nuclear Waste Management XX, Vol.465, 1997,1297-1308.*
9. Burakov B.E., Anderson E.B., Strykanova E.E. **Secondary Uranium Minerals on the Surface of Chernobyl “Lava”**. *Mat. Res. Soc. Symp. Proceedings Scientific Basis for Nuclear Waste Management XX, Vol.465, 1997, 1309-1311.*
10. Burakov B.E., Shabalev S.I., Anderson E.B. **Principal Features of Chernobyl Hot Particles: Phase, Chemical and Radionuclide Compositions**. In S. Barany, Ed. Role of Interfaces in Environmental Protection, Kluwer Academic Publishers, 145-151, NATO Science Series, Earth and Environmental Sciences, Vol. 24. 2003.
11. Боровой А.А., Велихов Е.П. **Опыт Чернобыля**, Часть 1, Москва, 2012 – *Borovoy A.A., Velihov E.P.* ***Experience of Chernobyl****, Part 1, Moscow, 2012 (in Russian)***.**
12. Nasirow R., Poeml P. **Gamma-ray spectrometry of Chernobyl ceramic samples**. Internal Report of JRC Institute of Transuranium Elements. Karlsruhe, 2013.

After explosion – first days [11]





Cross-section of Chernobyl "Shelter" [11]



Background

general information

- About 3.5 wt.% spent fuel was ejected from the core [1,2]
- About 50 kg Pu was spread in European part of USSR [4]
  (it means 6 wt.% of total Pu of Chernobyl NPP's 4th Unit)
- More than 90 wt.% fuel is inside "Shelter" or "Sarcophagus" [2], *but this information is controversial*
- At least 11-15 wt.% fuel (inside "Shelter") is related to Chernobyl "lava" [5], *but this information is controversial*

Background

basic glossary

- **Chernobyl "lava"** – it is a result of high-temperature interaction between destroyed fuel, Zr-cladding and silicate materials (concrete, sand, serpentinite) – can be called *silicate-rich corium*
- **Chernobyl corium** – melted fuel-containing material, which does not have silicate matrix
- **Chernobyl "hot" particles** – are highly radioactive solid particles from less than 1 µm to hundreds µm in size

Note: Chernobyl hot particles usually contain U but not always!

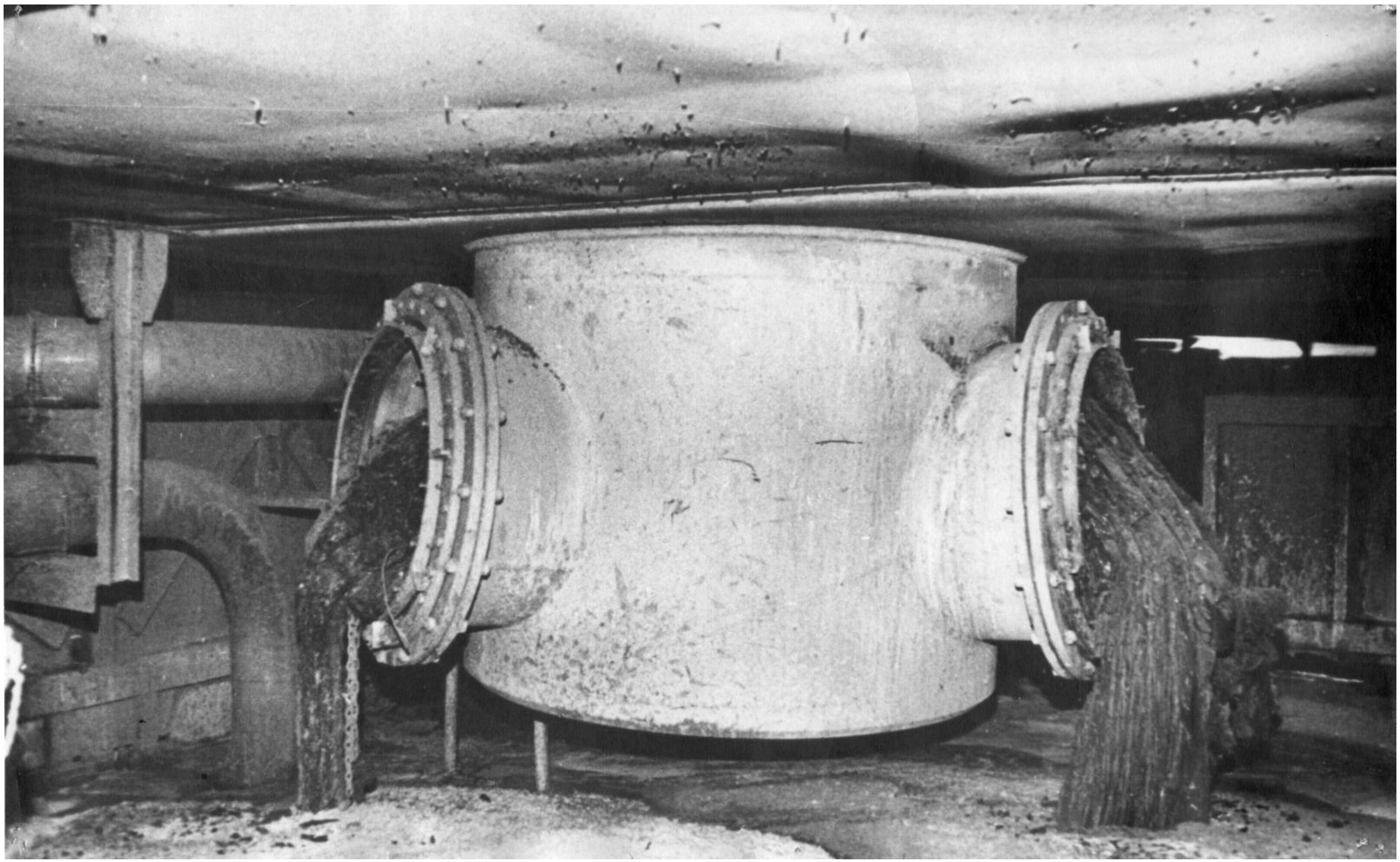
Natural volcanic lava




Chernobyl “lava”-stream called “Elephant foot”, 1990 [2]

gamma-doze on the surface of “Elephant foot” in 1990 exceeded 10 Sv/h, and gamma-radiation field in the room was 6-7 Sv/h

Chernobyl "lava" in steam discharge corridor, 1990 [2]



Background

general information

- Initial mechanical durability of Chernobyl “lava” was very high. Shooting by machine-gun AK-47 was applied to break “Elephant foot” matrix and collect first samples in 1987 [2]
- Essential decrease of mechanical durability and even self-destruction of “lava” matrices was observed in 1990 [2]
- Chemical alteration of “lava” matrices was observed in 1990 – formation of “yellow stains” consisted of secondary uranium minerals (uranyl-phases) [9]

New-formed yellow minerals at the surface of Chernobyl "lava", 1991

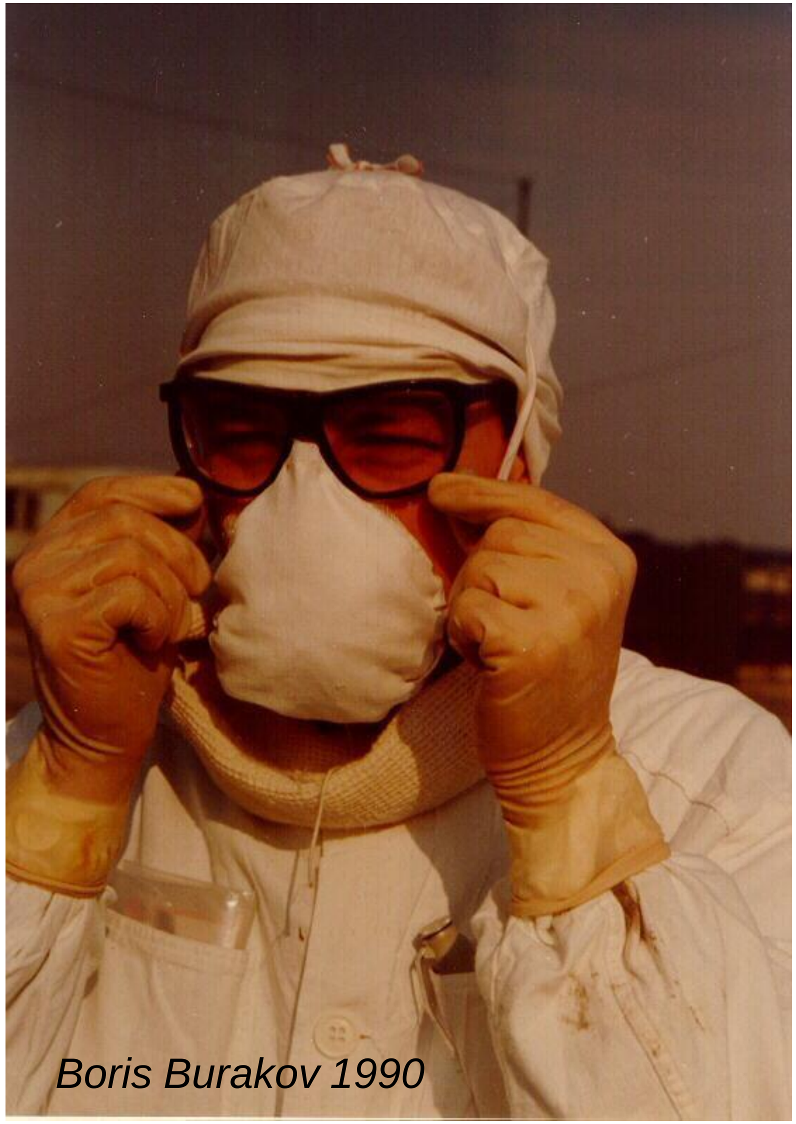
Samples of Chernobyl “lava”

collection of V.G. Khlopin Radium Institute

- Most samples of “lava” were collected at different locations in 1990 *using hands and hammer only. All people involved into sampling were overirradiated*

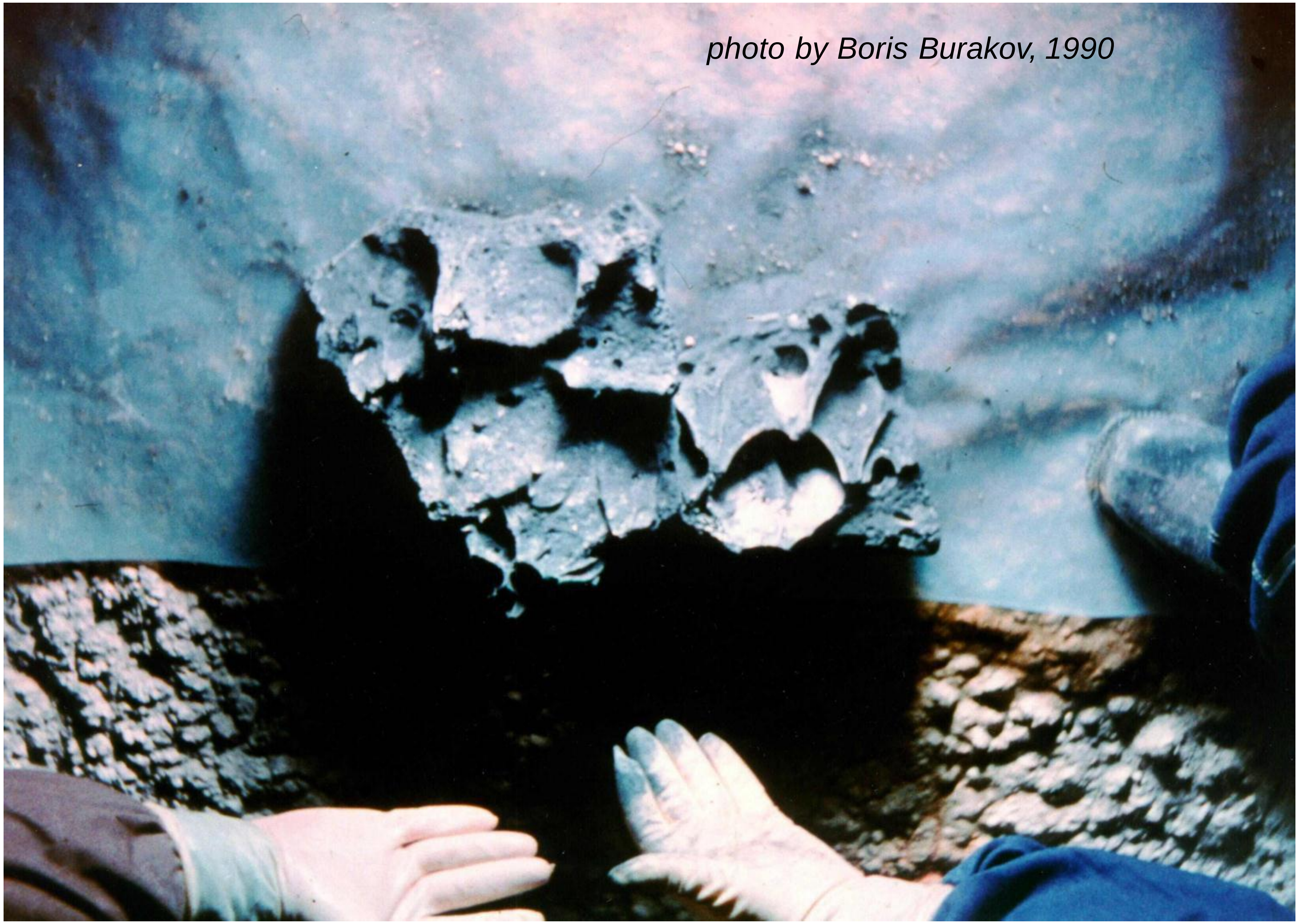
- Some pieces of “lava” (dozens cubic cm each) were partially dissolved in HF in order to extract inclusions of different uranium-bearing phases

Before going inside “Shelter”, 1990

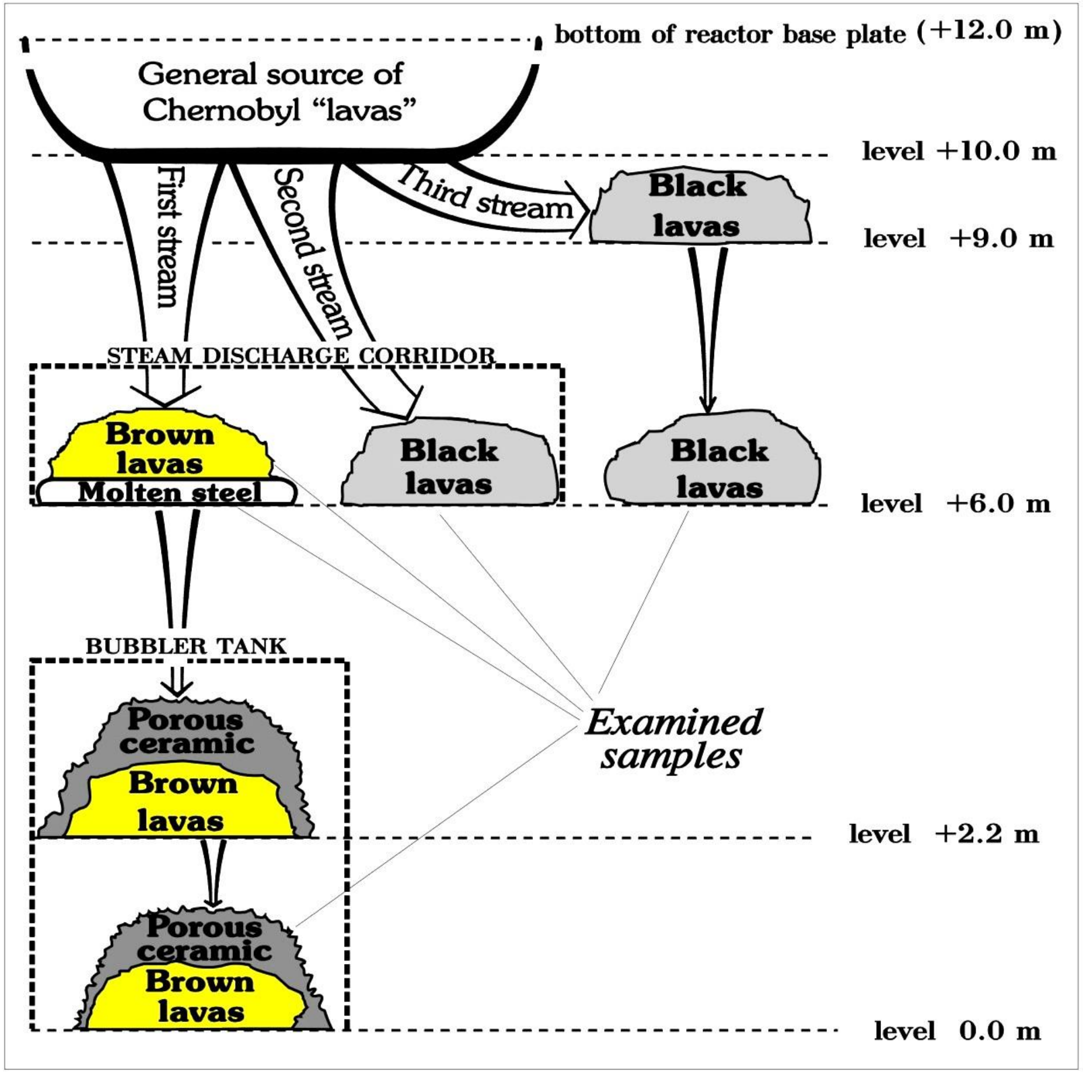


Boris Burakov 1990

Inside “Shelter”: packing “lava” sample for shipment to Leningrad, 1990



Map of KRI sampling inside “Shelter” [8]



Current study of highly radioactive Chernobyl samples – what for?

- *Transfer of experience to young scientists*
- Tutorials on material science of highly radioactive solids
- Modeling of severe nuclear accidents and corium behavior
- *Modeling of properties of Fukushima's corium*
- Chernobyl "lava" as analogue of HLW glass
- New-formed artificial unstable radioactive phases as a result of corium and "lava" chemical alteration
- New-formed very stable crystalline radioactive phases (inclusions in the "lava" matrices) as perspective durable host-phases of radionuclides

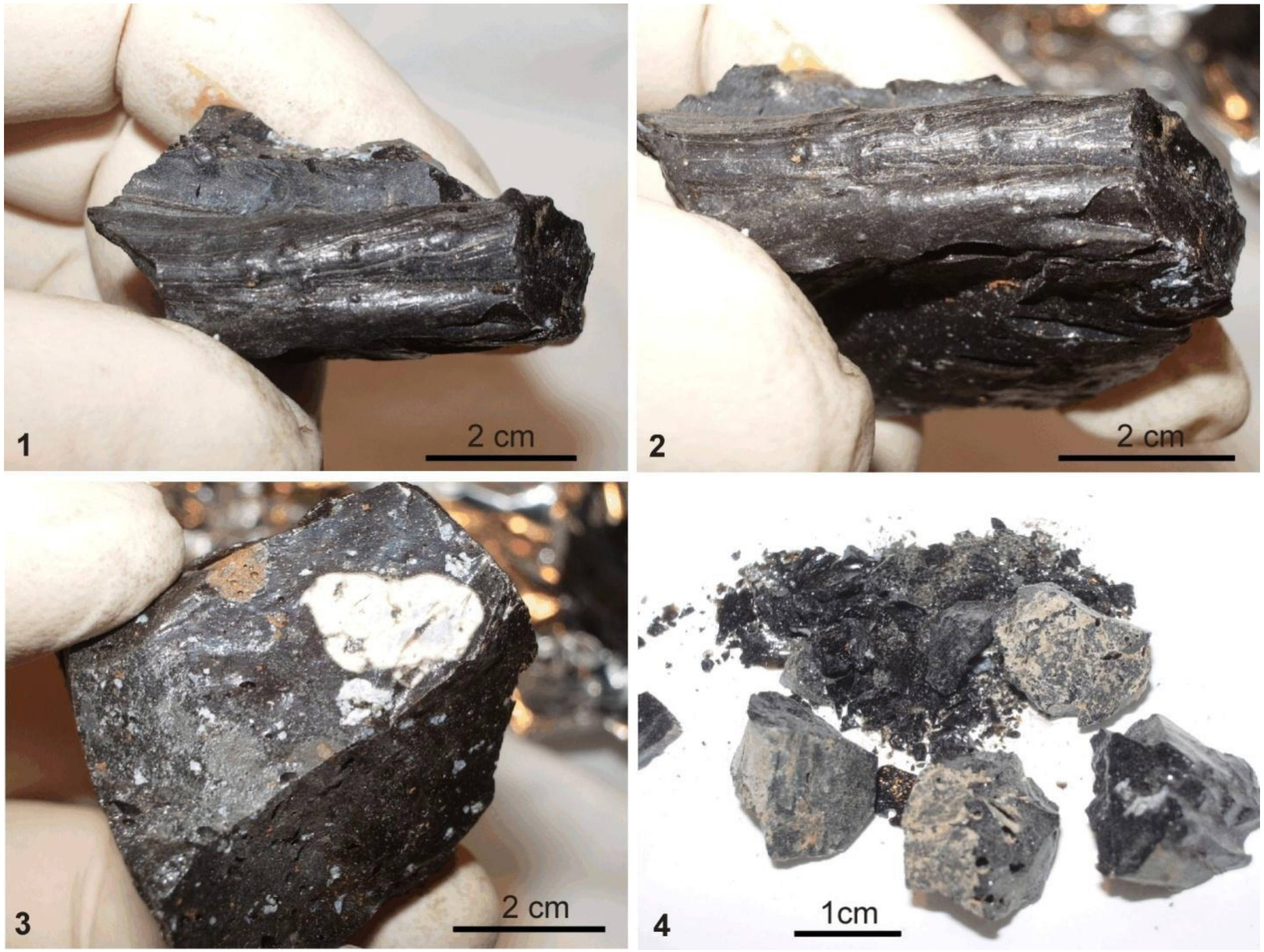
Samples of black "lava" – "Elephant foot"

collection of V.G. Khlopin Radium Institute

samples were collected in 1990 and stored at KRI under laboratory conditions

partial self-destruction was observed for some pieces in 2011 (picture 4).

photo by V. Zirlin and B. Burakov

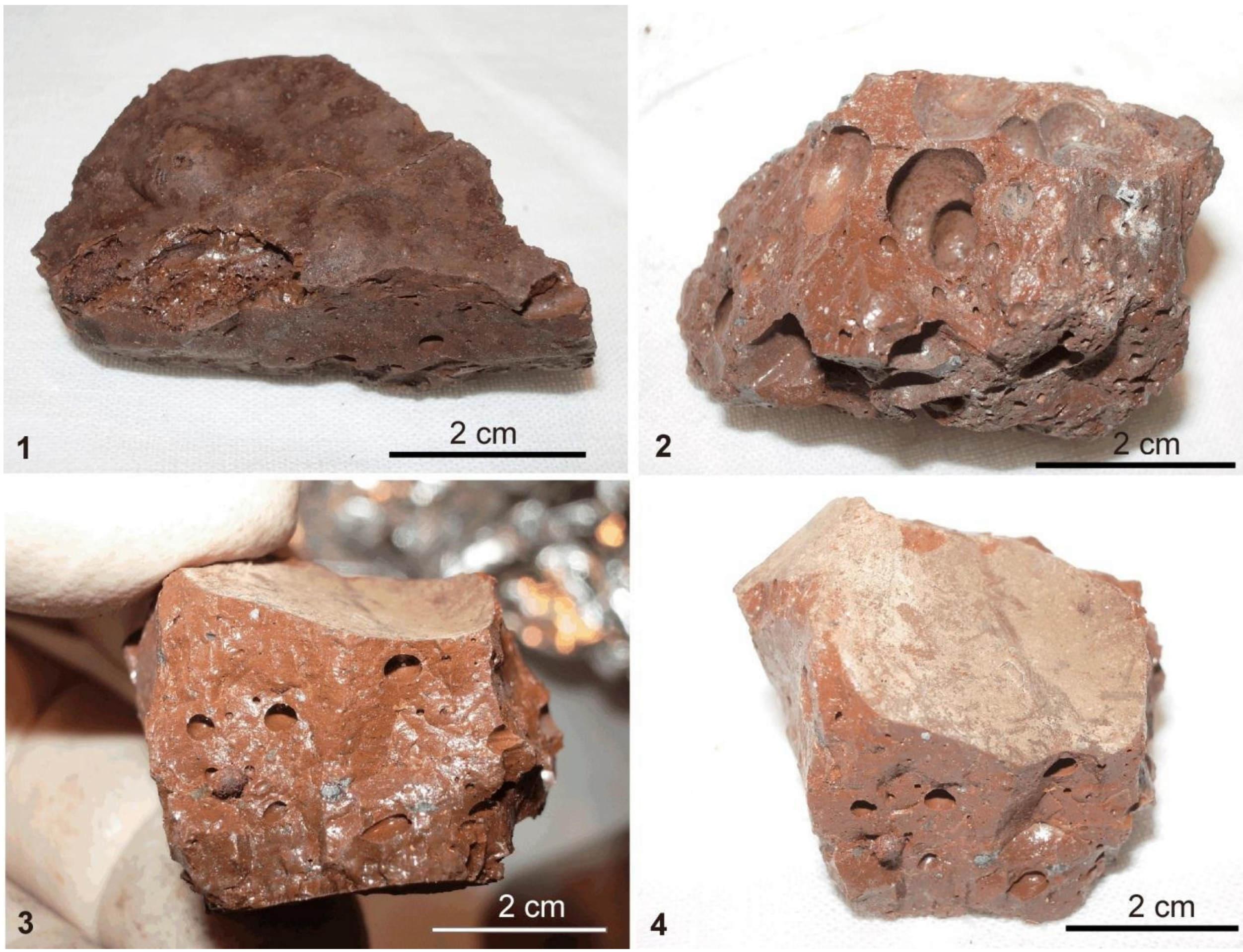


Samples of brown "lava" – from steam discharge corridor

collection of V.G. Khlopin Radium Institute

samples were collected in 1990 and stored at KRI under laboratory conditions

pictures were taken in 2011 *by V. Zirlin and B. Burakov*



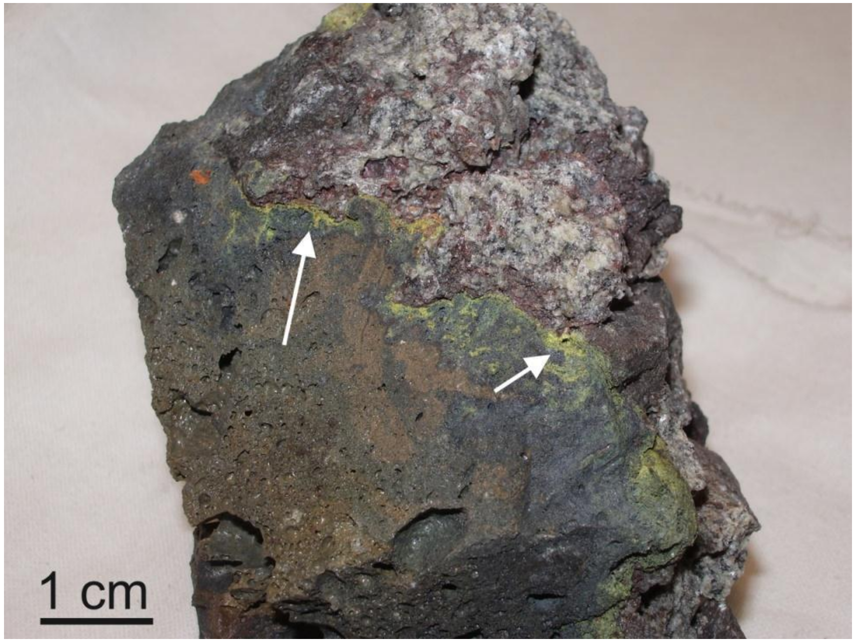
Samples of brown "lava" – from room #305

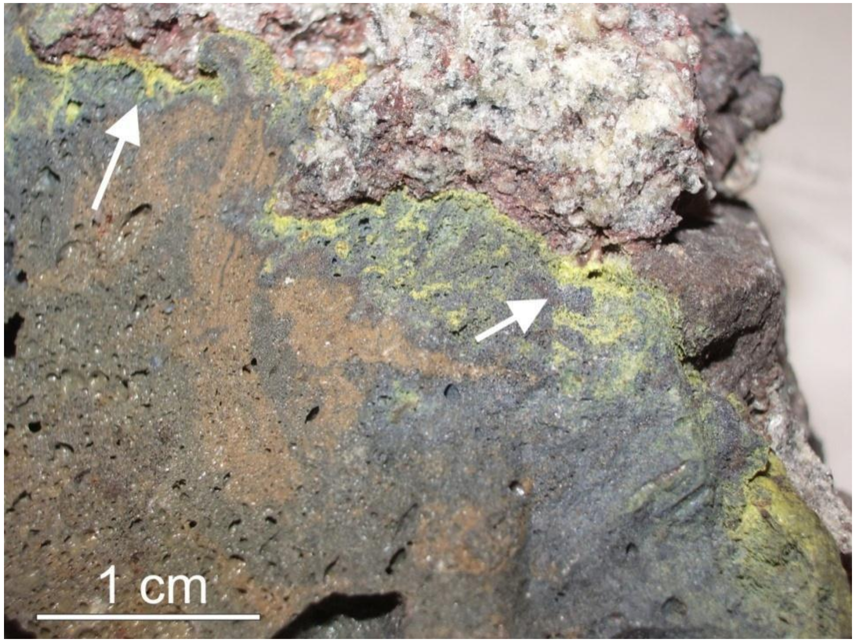
collection of V.G. Khlopin Radium Institute

sample was collected in 1990 and stored at KRI under laboratory conditions

pictures were taken in 2011 *by V. Zirlin and B. Burakov*

formation of secondary uranium minerals under laboratory conditions ?





Some Chernobyl samples from KRI collection have not been studied yet

Sample of corium (fuel-steel melt) – from room #305

collection of V.G. Khlopin Radium Institute

sample was collected in 1990 and stored at KRI under laboratoryconditions

picture was taken in 2011 *by V. Zirlin and B. Burakov*

Sample of corium (fuel melt on the surface of steel pipe) room #305

collection of V.G. Khlopin Radium Institute

sample was collected in 1990 and stored at KRI under laboratoryconditions

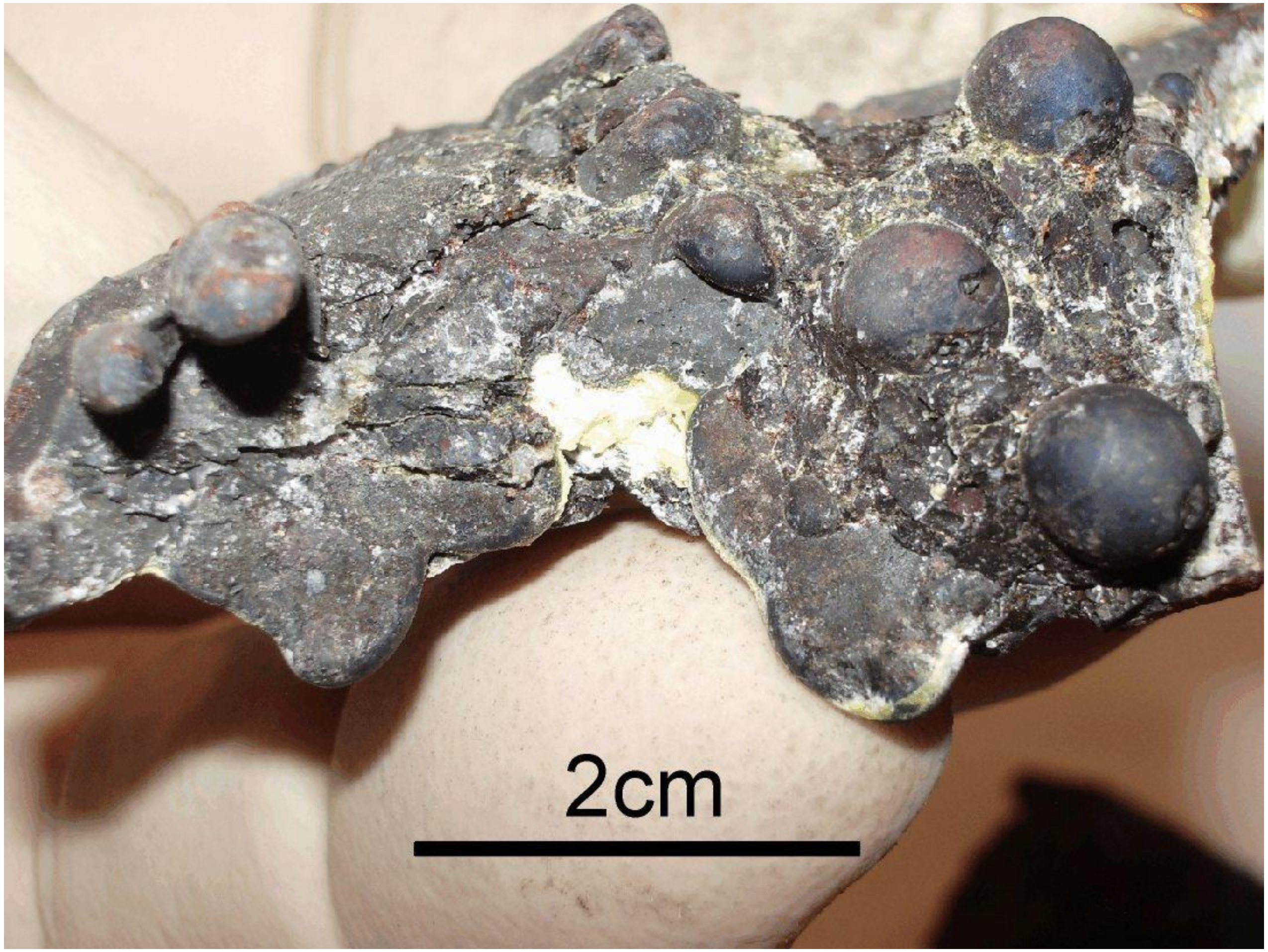
picture was taken in 2011 *by V. Zirlin and B. Burakov*

Sample of corium (fuel-steel melt) – near “Elephant foot”

collection of V.G. Khlopin Radium Institute

sample was collected in 1990 and stored at KRI under laboratoryconditions

picture was taken in 2011 *by V. Zirlin and B. Burakov*



Samples of Chernobyl "hot" particles

collection of V.G. Khlopin Radium Institute

- Some particles were separated from soil samples collected near 4th Unit in 1986
- Most particles were separated from soil samples collected at Western Plume (0.5-12 km from 4th Unit) in 1990-1991
- Some fuel fragments and particles were collected inside "Sarcophagus" in 1990

“Red forest” – pine-tree forest died as a result of Chernobyl fallout, June 1986

photo – courtesy of Dr. R.V. Arutyunyan (IBRAE, Russia)

Collecting hot particles, 1990

Separation of hot particles from soil sample [7]

at V.G. Khlopin Radium Institute – using collimated beta-gamma-detector

Methods of analyses

at V.G. Khlopin Radium Institute

- Optical microscopy
- SEM (BSE imaging)
- Quantitative and qualitative EMPA
- Bulk powder XRD (secondary uranium minerals, mineral inclusions separated from “lava” matrices)
- Precise XRD of single hot particles and mineral inclusions separated from “lava” matrices
- Gamma-spectrometry of bulk samples and single hot particles

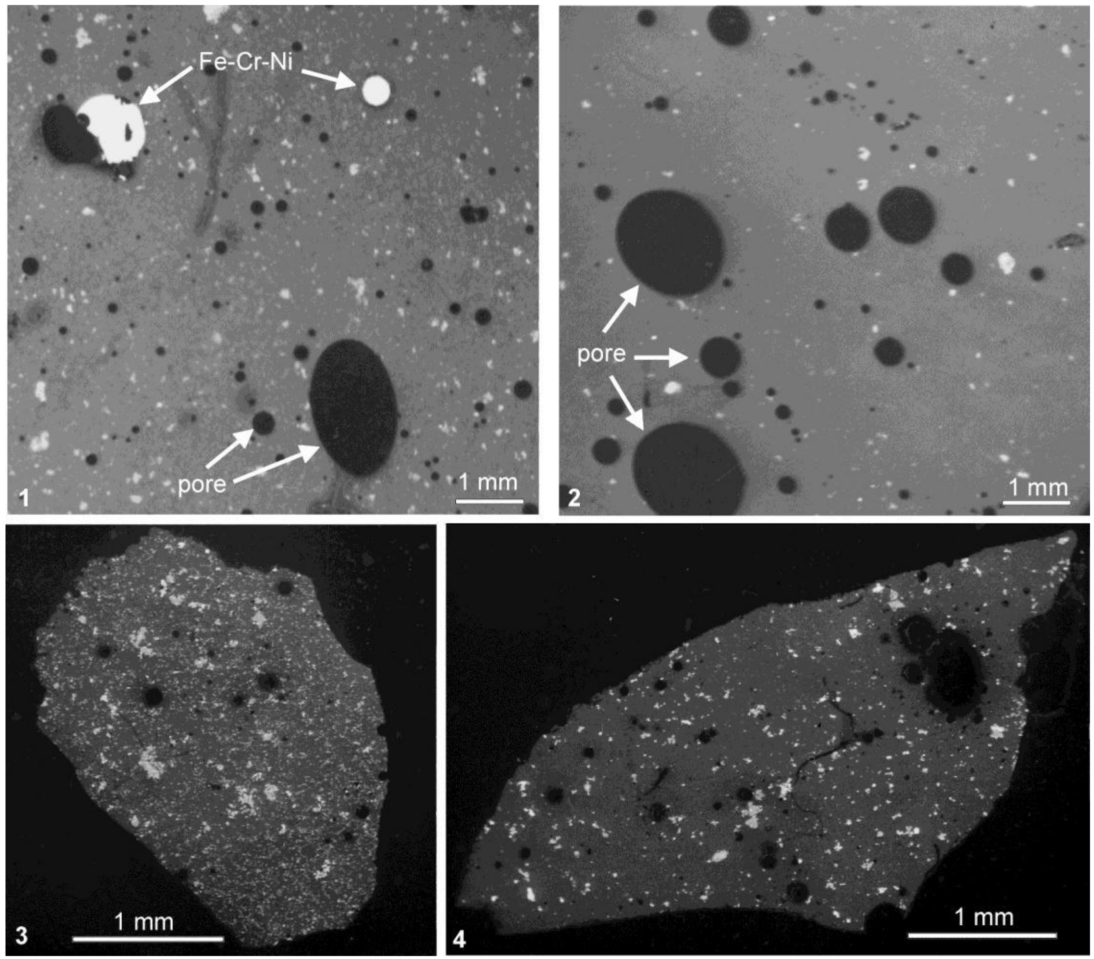
Chernobyl “lava”

brief summary or the results obtained
at V.G. Khlopin Radium Institute

Images of polished "lava" samples

1,2 – in reflected light in optical microscope; 3,4 – SEM-BSE

1,3 – brown "lava" from steam discharge corridor; 2 – black "lava" from "Elephant foot; 4 – black"lava" from steam discharge corridor



Chernobyl "lava" consist of silicate glass matrix + inclusions

Radionuclide composition of Chernobyl “lava” on June 2013 [12]

(recalculated for 26.04.1986) [3,7]

Type of “lava”	Radionuclides, Bq/g						
	^{137}Cs	^{144}Ce	^{154}Eu	^{244}Cm	^{241}Am	^{239,240}Pu	^{238}Pu
Black	$2 \cdot 10^7$ *($2.3 \cdot 10^7$)*	*($2 \cdot 10^9$)*	$5 \cdot 10^5$ *($1.3 \cdot 10^6$)*	$5 \cdot 10^4$ *($1.2 \cdot 10^7$)*	$1.2 \cdot 10^6$ *($3.5 \cdot 10^7$)*	$8.2 \cdot 10^5$ *($7.3 \cdot 10^7$)*	$4.3 \cdot 10^7$ *($3.8 \cdot 10^7$)*
Brown	$4.1 \cdot 10^7$	*($2.1 \cdot 10^9$)*	$1.2 \cdot 10^6$	$1.1 \cdot 10^5$	$2.8 \cdot 10^6$	$1.8 \cdot 10^6$	$9.2 \cdot 10^5$

Simplified bulk chemical composition (*matrix* + *inclusions*) of Chernobyl "lava" [6,7]

Type of "lava"	Element content, wt. %							
	U	**Zr**	**Na**	**Fe**	**Mg**	**Ca**	**Si**	**Al**
Black	4 - 5	2 - 6	2 - 10	0.3 - 6	1 - 5	3 - 13	19 - 36	3 - 8
Brown	8 - 7	5 - 6	4	1 - 2	4	5	31 - 33	4

Results of electron-probe microanalyses of glass-like silicate matrix of Chernobyl "lava" avoiding inclusions of crystalline phases [6,8]

Type of "lava"	Element content, wt. %								
	U	**Zr**	**K**	**Na**	**Fe**	**Mg**	**Ca**	**Si**	**Al**
Black	2.7-4.0	3.1-3.7	1.4-2.7	0.4	0.3-6.7	1.2-3.2	5.1-7.2	28-37	2.7-4.4
Brown	2.0-2.4	2.4-2.9	1.2-2.3	0.6	0.2-0.4	3.5-4.4	4.5-8.2	37	2.8-4.0
Porous	2.9	4.0	2.3	0.5	0.2	4.5	7.5	35	3.8

Inclusions in matrices of Chernobyl “lava” are of

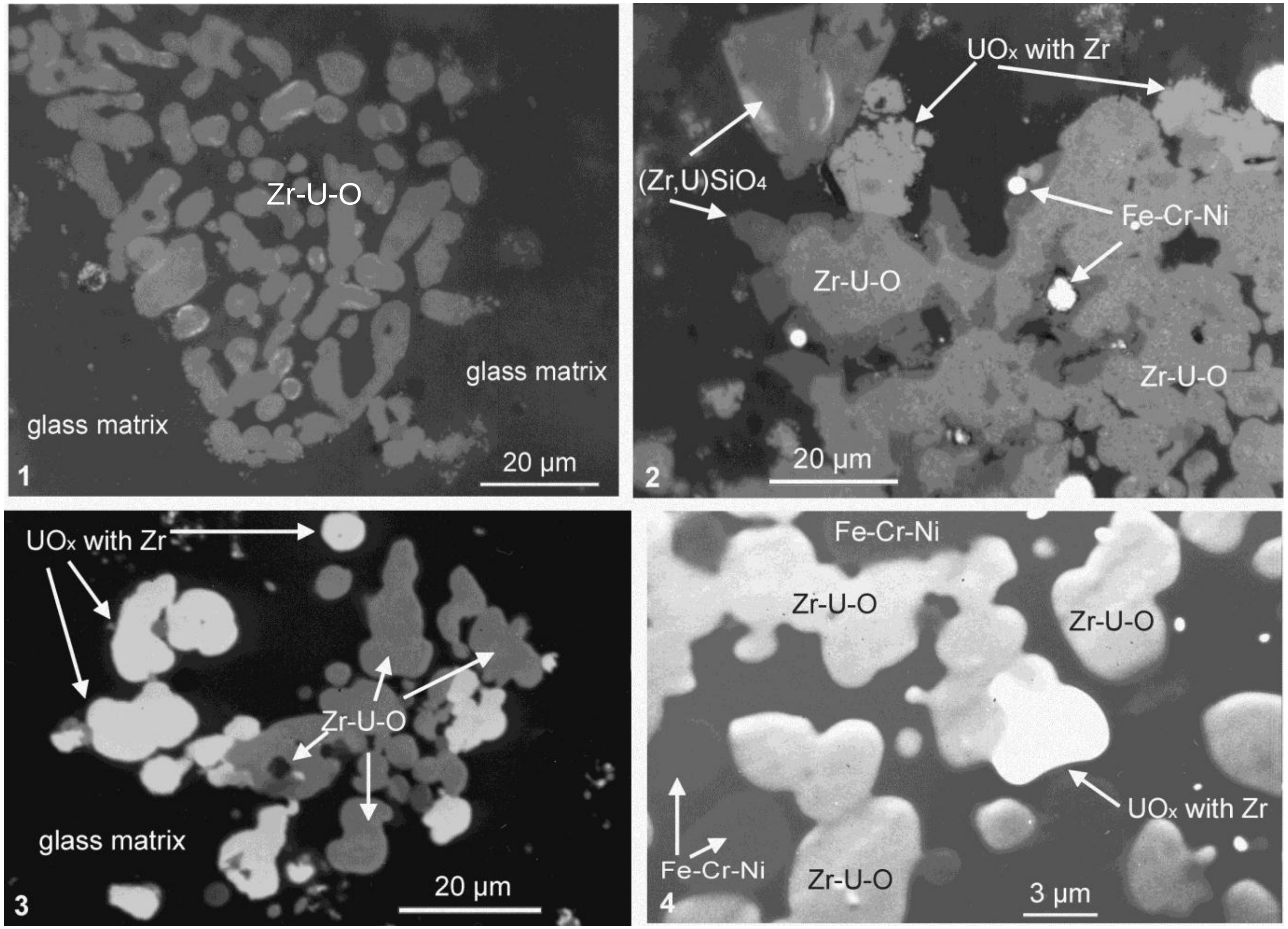
very different phase and chemical compositions!

Inclusions in brown "lava" matrix

(from steam discharge corridor)

1,2 – in reflected light in optical microscope; 3,4 – SEM-BSE

photo by Boris Burakov, 1990-1991

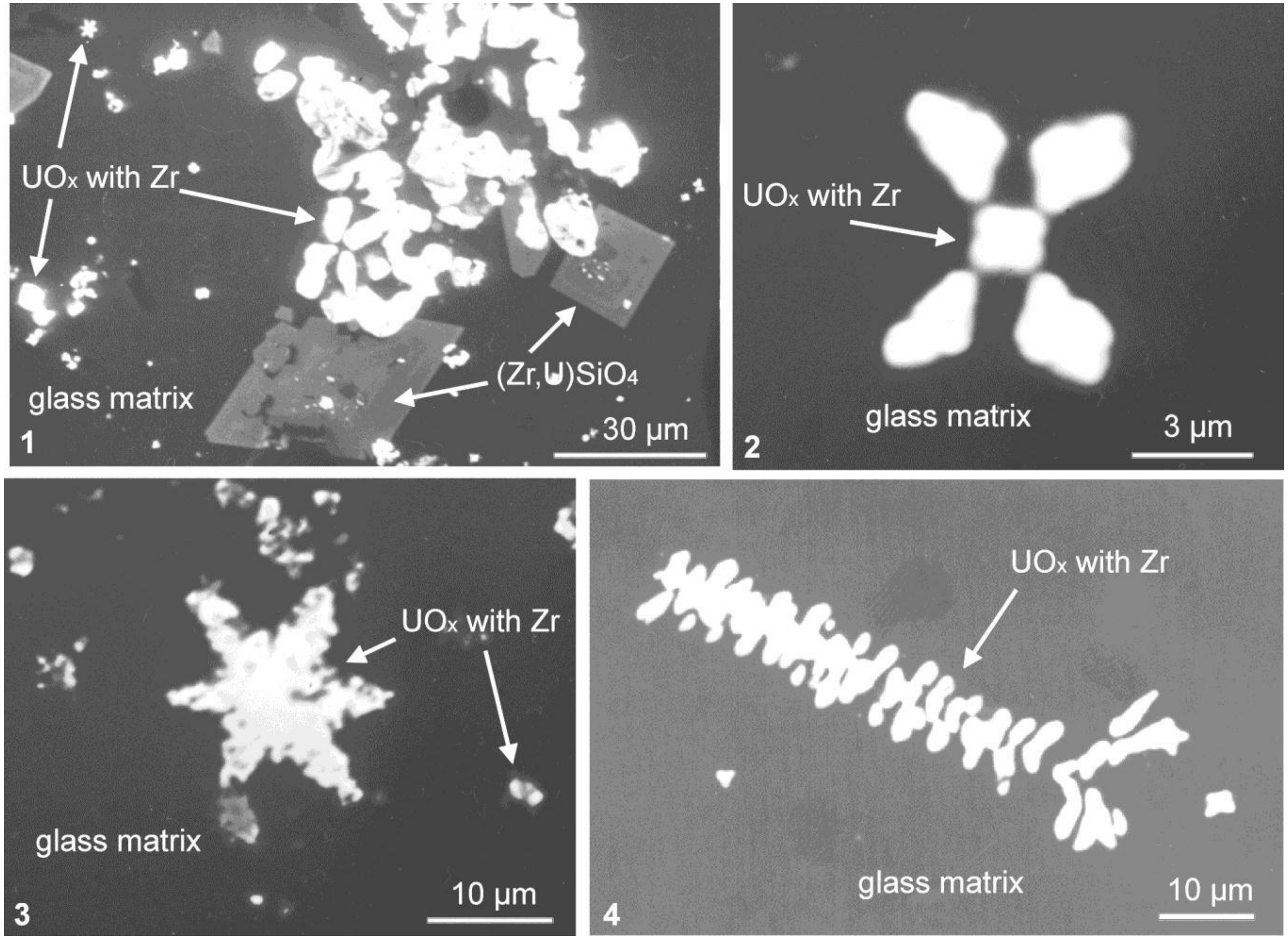


Inclusions in black and brown "lava" matrices

SEM-BSE

1,2 – black "lava" from steam discharge corridor; 3 – brown "lava" from steam discharge corridor; 4 – black "lava" from "Elephant foot

photo by Boris Burakov, 1990



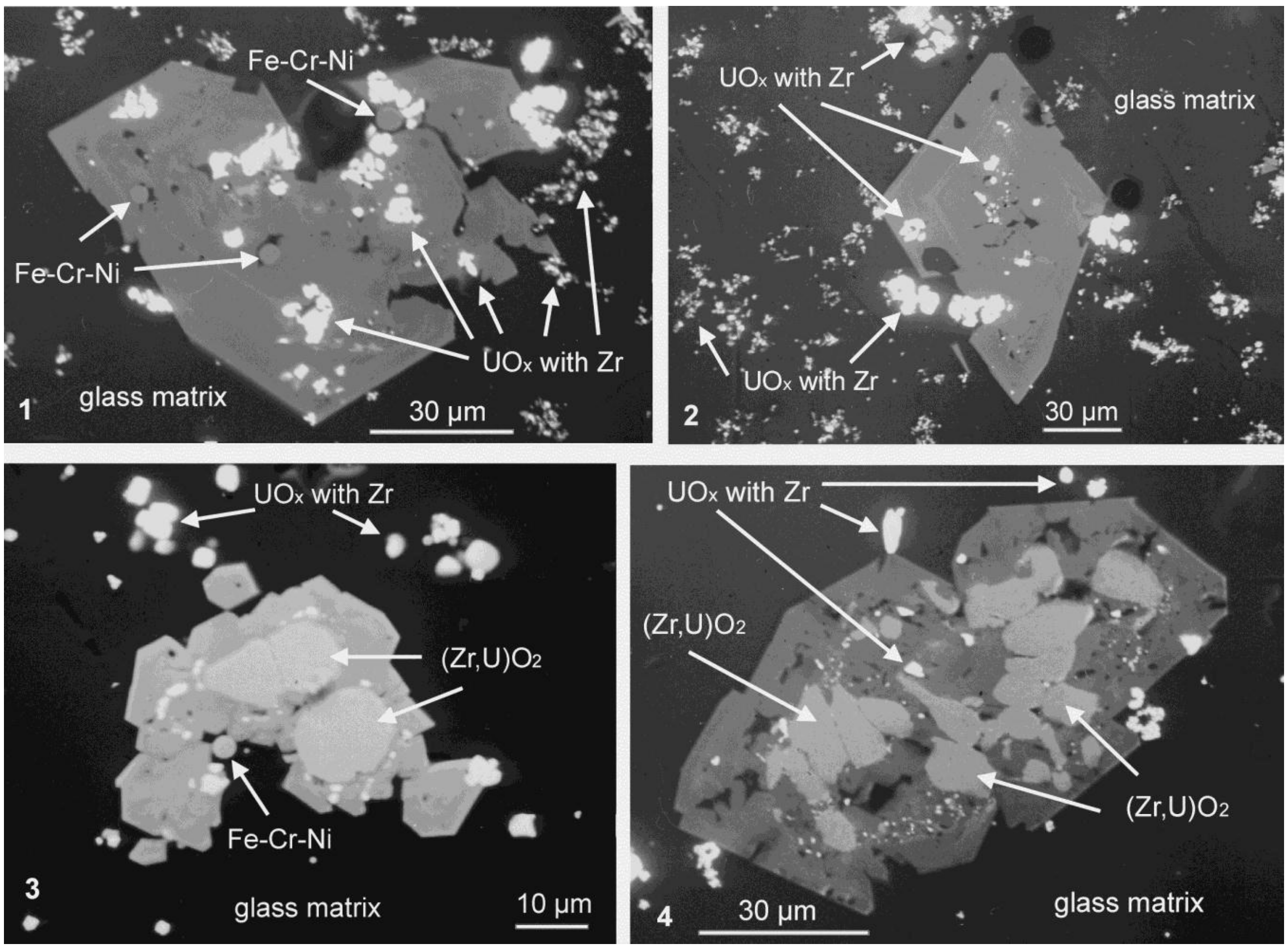
Inclusions in black and brown “lava” matrices

(from steam discharge corridor)

SEM-BSE

1,2 – brown “lava”; 3,4 – black “lava”

photo by Boris Burakov, 1990



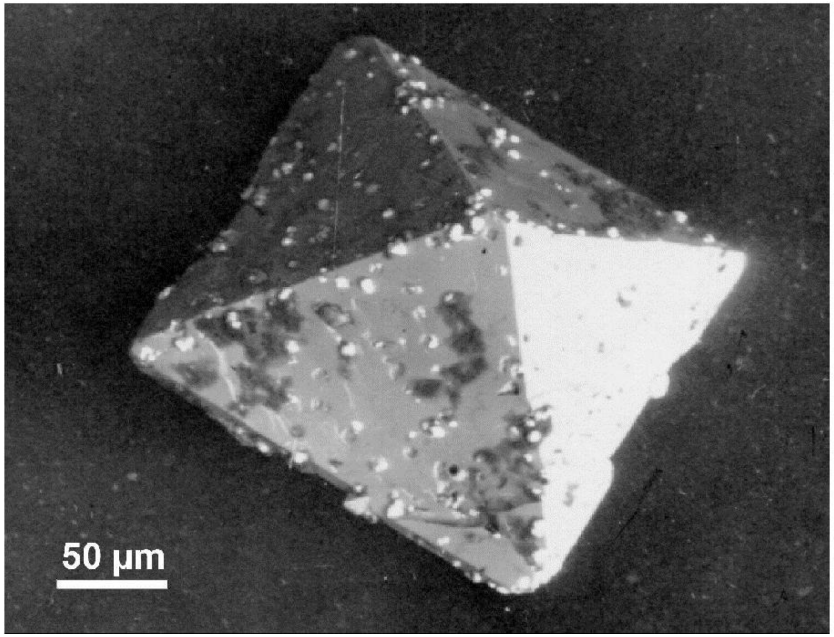
Crystals of high-uranium zircon, $(Zr,U)SiO_4$, are typical for all types of Chernobyl "lava"

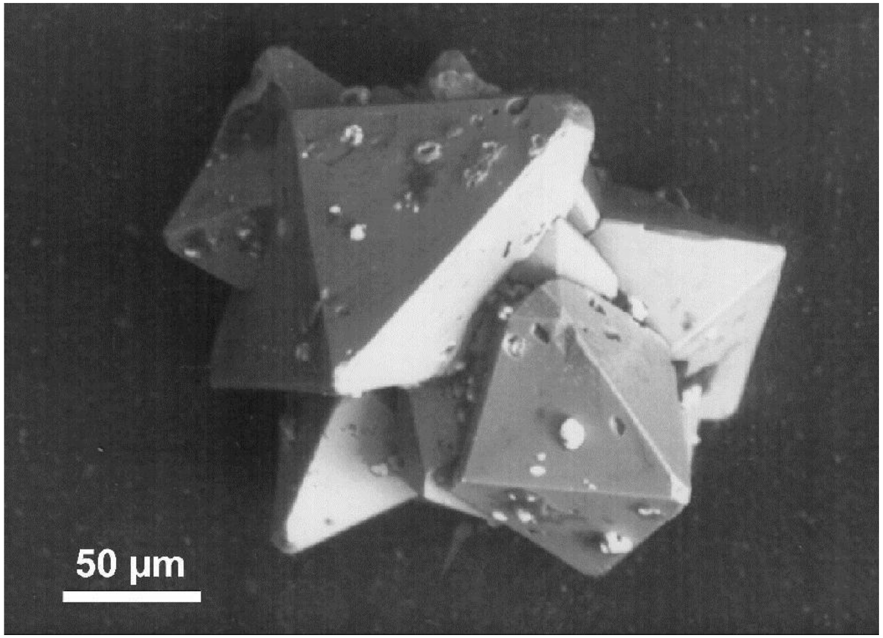
Up to 10 wt. % uranium was incorporated into the crystalline structure of zircon in the form of solid solution !

High-uranium zircon, $(Zr,U)SiO_4$, from Chernobyl “lava”

crystals were extracted after partial dissolution of “lava” matrix in HF

photo by Boris Burakov, 1990

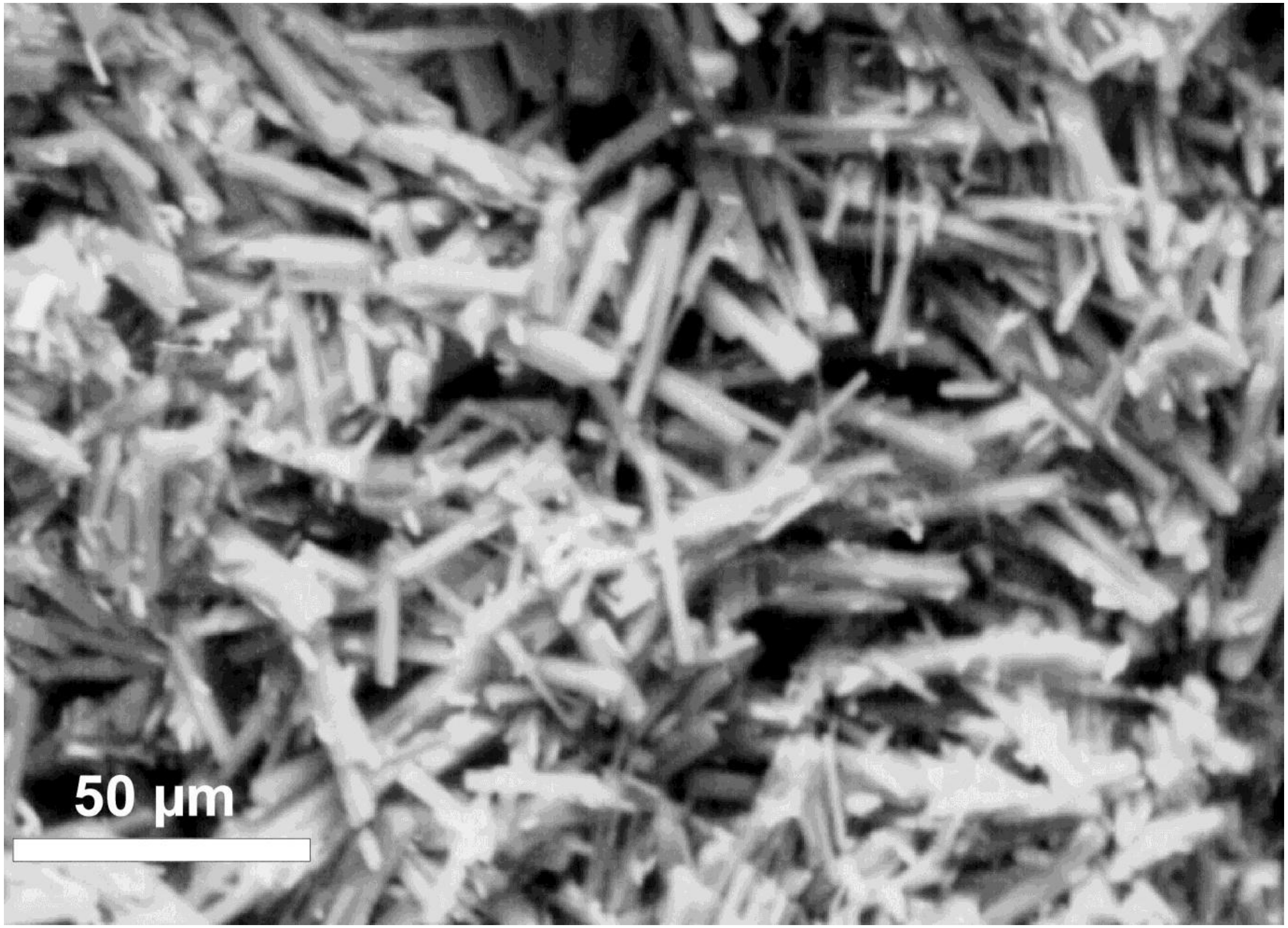




New-formed yellow minerals at the surface of Chernobyl "lava", 1991

SEM-BSE image of new-formed minerals at the surface of Chernobyl “lava” [9]

photo by Boris Burakov, 1990



Phase composition of new-formed minerals at the surface of Chernobyl "lava" (from powder XRD analysis [6])

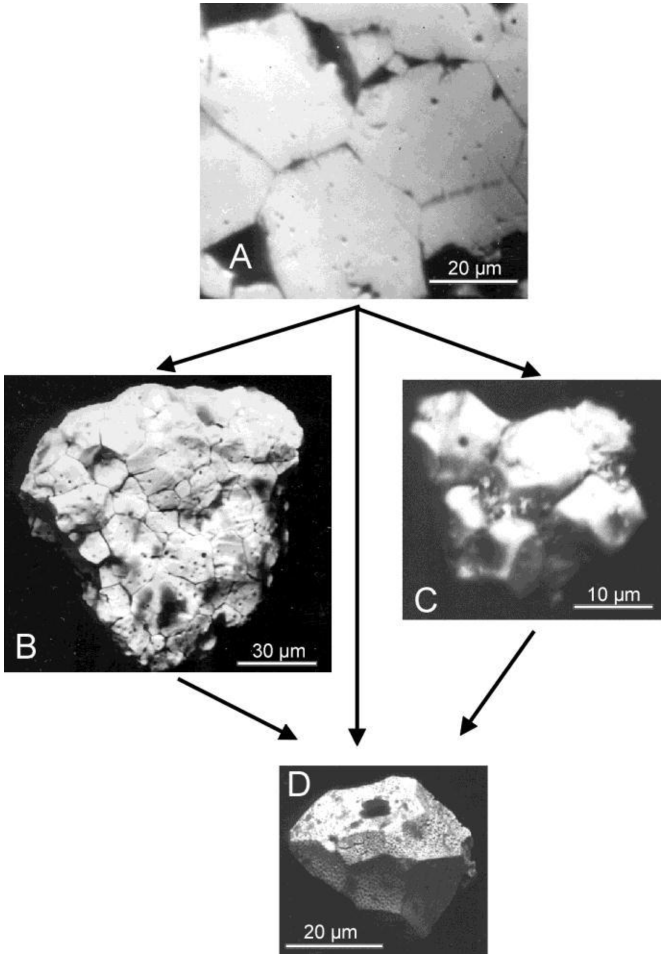
- $Na_3H(CO_3)_2 \times 2H_2O$
- $UO_3 \times 2H_2O$
- $Na_4(UO_2)(CO_3)_3$
- $Na_2CO_3 \times 2H_2O$
- $UO_4 \times 4H_2O$
- UO_2CO_3

Chernobyl “hot” particles

brief summary or the results obtained
at V.G. Khlopin Radium Institute

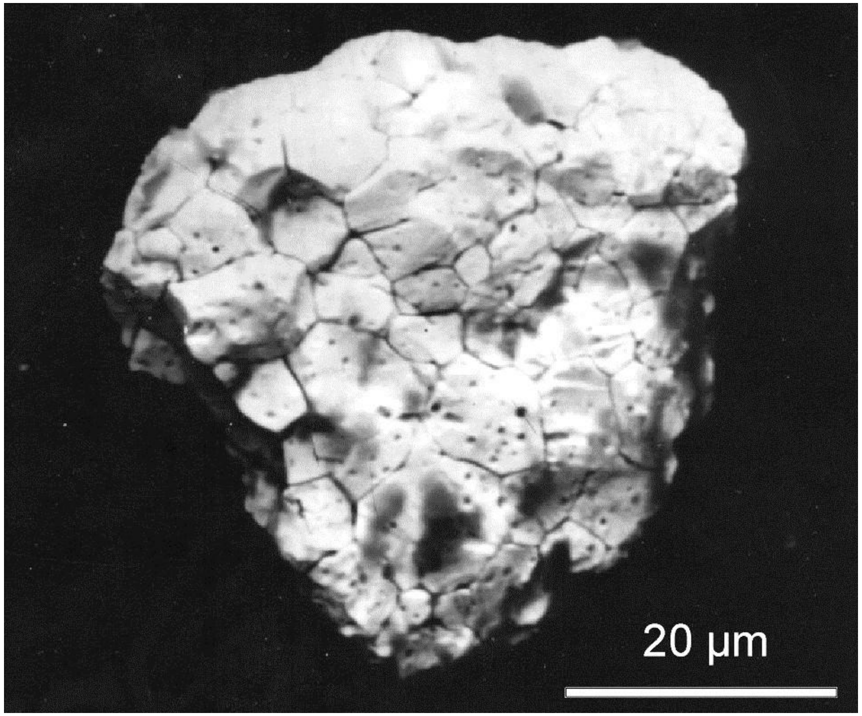
SEM-BSE images of fuel fragment (A) and hot particles (B,C,D) of fuel composition (UO_x) [10]

possible mechanical self-destruction along grain boundaries

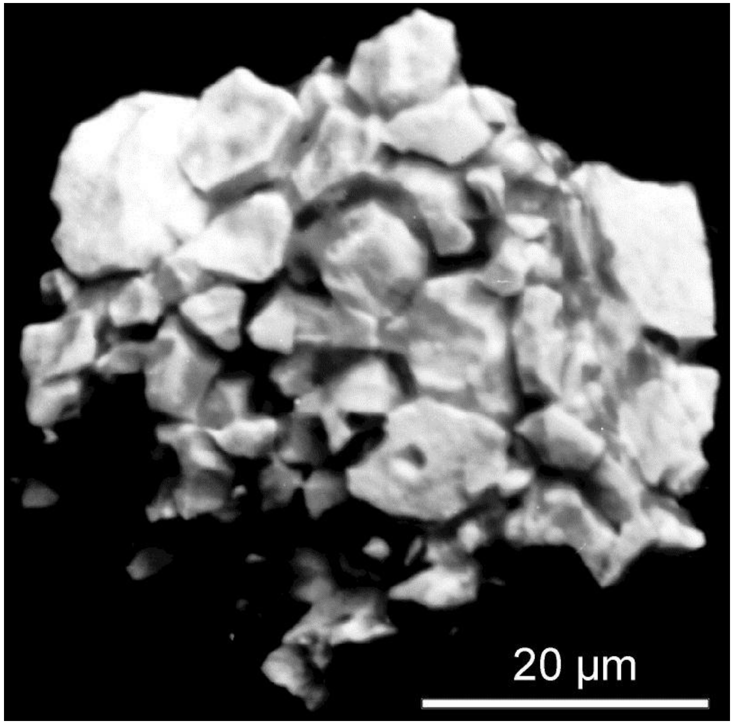


Multi-grain fuel (UO_x) hot particles (collected in 1990)

photo by Boris Burakov, 1990



non-altered

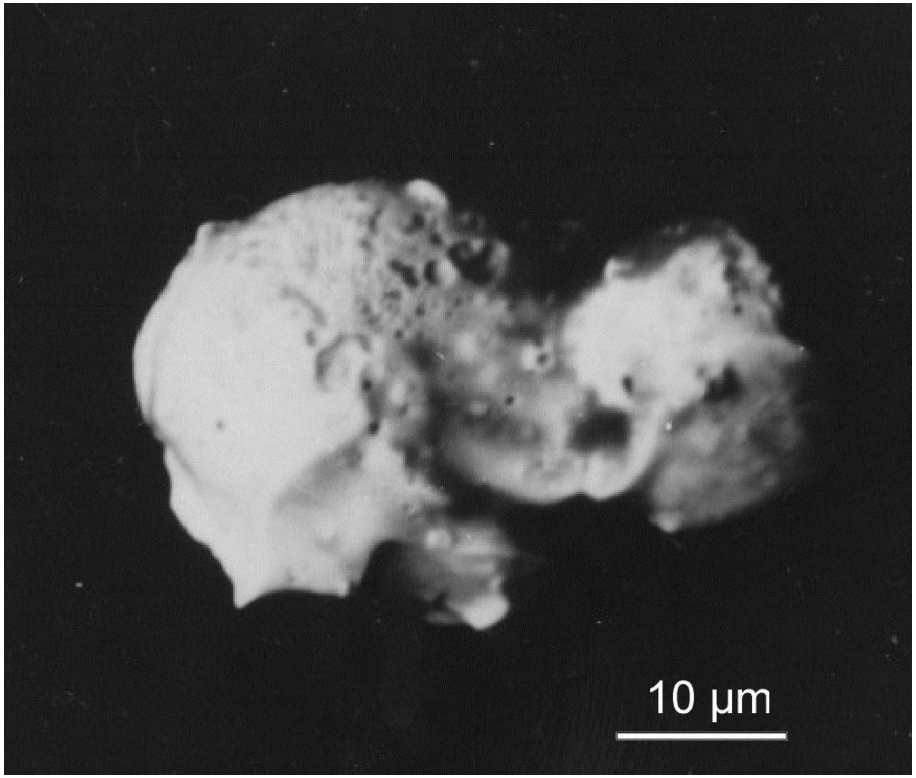


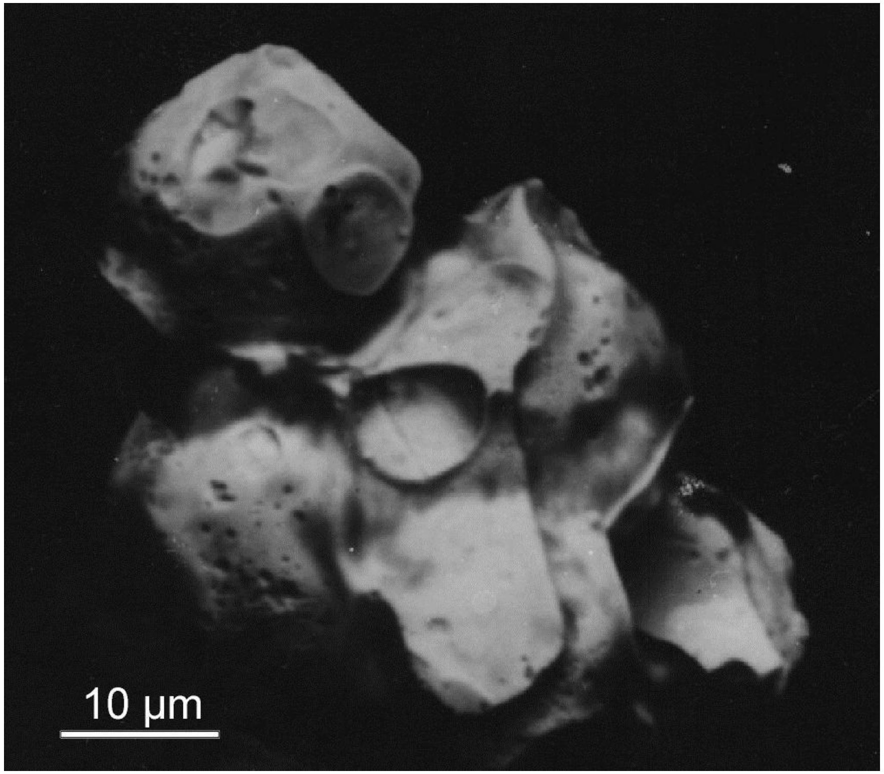
altered ???

dissolution along grain boundaries ?

Fuel hot particles (UO_x) with molten morphology (?)

photo by Boris Burakov, 1990



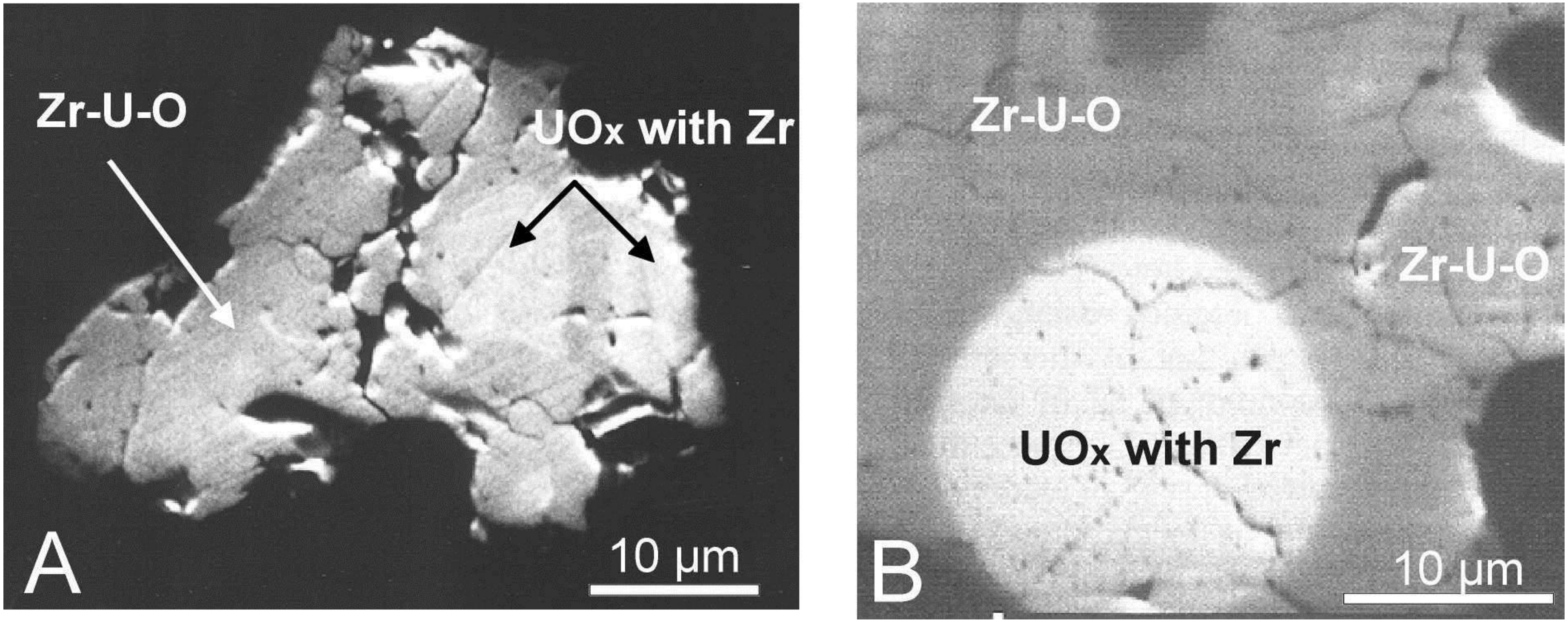


We also found Zr-bearing hot particles with phases:
Zr-U-O and UO_x with Zr etc.

up to 40 % of all particles in some places of Western Plume !

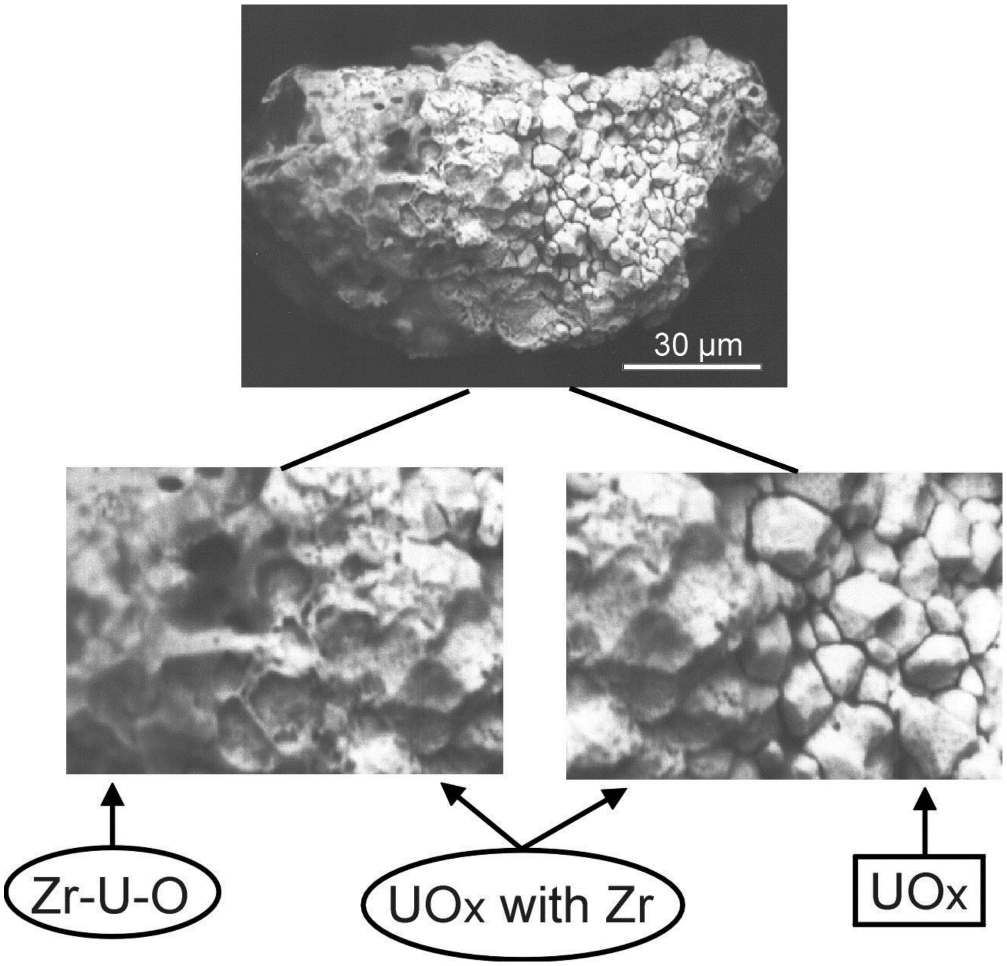
Multi-phase hot particles [10]

polished cross-sections, SEM-BSE



Multi-phase hot particle [10]

SEM-BSE image



No interaction between fuel (UO_x) and zircaloy (almost pure metallic Zr) is possible in air. Fast oxidation of metallic Zr blocks this process

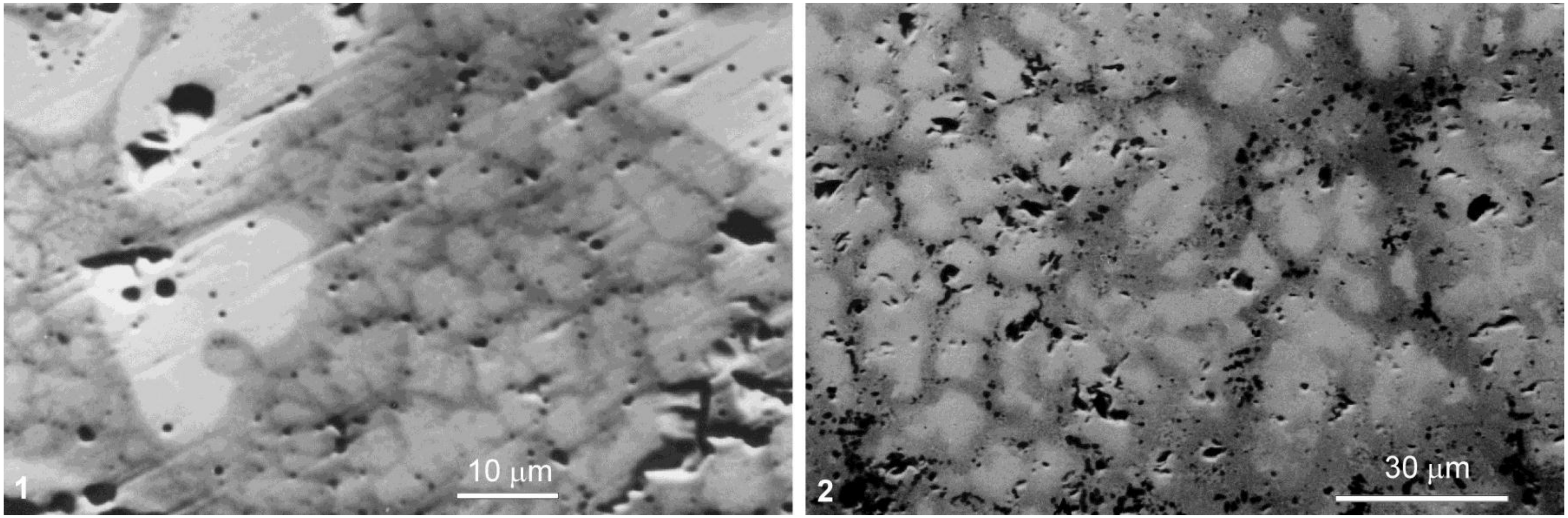
No conditions for Zr-U-O melt formation after explosion of reactor core !

Formation of Zr-U-O melt

1) In corium of Chernobyl reactor (before explosion) – *it is matrix of real hot particle*

2) Laboratory experiment (melting at 2600ºC in vacuum) of Zr + UO_2 – *it is synthetic sample*

photo by Boris Burakov, 1990



Crystalline U-bearing phases identified in Chernobyl "lava" and hot particles

- Cubic UO_x – similar to stoichiometric UO_2 (***a* = 5.462-5.473 Å**)
- Cubic UO_x with Zr (0,5 to 20 wt.% Zr) (***a*** decreases from **5.468 to 5.318 Å**). Chemical composition – $(U_{0.985}Zr_{0.015})O_2$-$(U_{0.895}Zr_{0.105})O_2$. In some hot particles Zr content is higher – up to $(U_{0.56}Zr_{0.44})O_2$
- Tetragonal phase Zr-U-O with varied chemical composition from $(Zr_{0.86}U_{0.14})O_2$ to $(Zr_{0.89}U_{0.11})O_2$
- Monoclinic zirconia with U (up to 6 wt.% U) with varied chemical composition from $(Zr_{0.995}U_{0.005})O_2$ to $(Zr_{0.967}U_{0.033})O_2$
- Solid solutions with non-identified structures: $(Zr_{0.56}U_{0.44})O_2$; $(Zr_{0.68-0.71}U_{0.32-0.29})O_2$; $(Zr_{0.75-0.77}U_{0.25-0.23})O_2$ – only in hot particles
- High-uranium zircon, $(Zr_{0.95}U_{0.05})SiO_4$-$(Zr_{0.90}U_{0.10})SiO_4$ (for bulk concentrate: ***a* = 6.617; *c* = 5.990 Å**).

Conclusions

- High-temperature (at least 2600°C) interaction between nuclear fuel and zircaloy cladding took place in local part of Chernobyl reactor core before the explosion

- Solid highly radioactive materials were formed and partially dispersed as a result of Chernobyl accident. **They have different phase and chemical composition.** It means their different behavior in environment

- Active chemical alteration of Chernobyl corium is going on

- Results of Chernobyl material study can be used for modeling severe nuclear accident at different types of nuclear reactors (not only RBMK)

- *Results of Chernobyl material study can be used for development of ceramic waste forms and other durable advanced materials*

KRI collaboration on study of Chernobyl “lava” and “hot” particles

KRI collaboration with French colleagues on study of Chernobyl “lava”, 1991-1993

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Radium Institute"

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KRI collaboration with German colleagues on study of Chernobyl “lava” and hot particles since 2004 till now

- *Geisler T., Burakov B. E., Zirlin V., Nikolaeva L., and Poml P.* **A Raman spectroscopic study of high-uranium zircon from the Chernobyl “lava”.** Eur. J. Mineral., (2005),17, pp. 883-894.
- *P. Pöml, B. Burakov, T. Geisler, C.T. Walker, M.L. Grange, A.A. Nemchin, J. Berndt, R.O.C. Fonseca, P.D.W. Bottomley, R. Hasnaoui*, **Micro-analytical uranium isotope and chemical investigations of zircon crystals from the Chernobyl „„„lava“““ and their nuclear fuel inclusions.** J. Nucl. Mater., (2013) Vol. 439, pp. 51-56.
- *P. Pöml. P., Burakov B.,* **Study of a “hot” particle with a matrix of U-bearing metallic Zr: Clue to supercriticality during the Chernobyl nuclear accident.** J. Nucl. Mater. (2017), Vol. 488, pp. 314-318.
- *Two fragments of black and brown Chernobyl “lava” are under investigation at ITU now in the framework of ITU-KRI Agreement.*

KRI collaboration with Russian colleagues (from Moscow) on study of Chernobyl "lava" and hot particles since 2012 till now

- *Batuk O.N., Conradson S.D., Aleksandrova O.N., Boukhalfa H., Burakov B.E., Clark D.L., Czerwinski K.R, Felmy A.R., Lezama-Pacheco J.S., Stepan N. Kalmykov S.N., Moore D.A., Myasoedov B.F., Reed D.T., Reilly D.D., Roback R.C., Vlasova I.E., Webb S.M.,Wilkerson M.P.* **Multiscale Speciation of U and Pu at Chernobyl, Hanford, Los Alamos, McGuire AFB, Mayak, and Rocky Flats.** J. Environmental Science and Technology, (2015) pp. 6474-6484.
- *Vlasova I., Shiryaev A., Ogorodnikov B., Burakov B., Dolgopolova E., Senin R., Averin A., Zubavichus Y., Kalmykov S.* **Radioactivity distribution in fuel-containing materials (Chernobyl "lava") and aerosols from the Chernobyl "Shelter".** Radiation Measurements, 83 (2015) 20-25.
- *Shiryaev A.A., Vlasova I.E., Burakov B.E., Ogorodnikov B.I., Yapaskurt V.O., Averin A.A., Pakhnevich A.V., Zubavichus Y.V.,* **Physico-chemical properties of Chernobyl lava and their destruction products.** Progress in Nuclear Energy, 92 (2016) 104-118.
- *Some polished micro-samples of Chernobyl "lava" and hot particles are under joint analysis at Radiochemical Department of Moscow State University now in the framework of collaboration between KRI, MSU and A.N. Frumkin Institute of Physical Chemistry and Electrochemistry.*

Testing of neutron detector (developed by “PDC UGR” JSC) using samples of Chernobyl “lava”, February 2017

Collaboration between KRI and Russian Pilot and Demonstration Center for Decommissioning of Uranium-Graphite Nuclear Reactors” JSC (“PDC UGR” JSC)

KRI proposals on cooperation with Japanese colleagues

- Consulting on fuel-debris and corium investigation, removal, treatment and final disposal
- KRI participation in material study of Fukushima's corium in Japan
- Training of Japanese colleagues at KRI using real highly radioactive Chernobyl samples ("lava", corium, hot particles)
- *Joint search, extraction and study of Fukushima's hot particles*

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