

# The Chernobyl pilot site project: Isolation and microscopic characterisation of fuel particles

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**Abstract.** A method of fuel particles separation from contaminated soil samples in heavy liquid was developed. Numerous particles have been isolated and characterised by Scanning Electron Microscopy. 14 years after Chernobyl accident 52% of Cs, 70% of Eu and Sr and 65% of Am still are associated with fuel particles. Hence the source term is mainly represented by fuel particles with a mean diameter of 4.17  $\mu\text{m}$ . The source term consist of 2 types of particles : UO and ZrUO. Zr-U-O fuel particles account for 74% of the studied particles and showed much higher stability. Therefore the description of Chernobyl source term weathering requires 2 dissolution laws: a first one concerning UO particles with a higher dissolution constant and a second one with a low dissolution constant describing Zr-U-O dissolution.

## 1. INTRODUCTION

In the year 1987, following the Chernobyl accident, Site of Temporary Localisation of Radioactive Wastes (STLRW) was created in the "Red Forest" zone. Trenches in the STLRW contain Red Forest materials (upper soil, litter and vegetable debris) contaminated by fuel particles released during the accident. A particular trench (trench 22), namely Chernobyl Pilot Site located at 2.5 km south west of Chernobyl Nuclear Power Plant, was selected for carrying out experiments to validate radionuclides transport models in soil and aquifer. For that, it is necessary to define the source term. The infiltration of water through the trench induces a weathering of fuel particles and associated radionuclides are mobilised with time. Different laws describing the dissolution process of spent fuel are available in literature [1]. A specific study conducted on Chernobyl fuel particles led to analogous conclusions [2,3]. The dissolution process depend on solution composition (pH, redox) and physico-chemical characteristics of fuel particles: chemical composition, oxidation state and surface. This work was therefore focused on determination of physico-chemical characteristics of fuel particles occurring in the trench. In this study we describe a new method allowing an isolation of fuel particles from soil components. After isolation, the particles are observed by Scanning Electron Microscopy (SEM) in order to determine the size distribution pattern, matrix composition, morphology and structure of fuel particles.

## 2. METHODS AND MATERIALS

### 2.1 Soil sampling

Waste sample from the trench was collected to a depth of 210-220 cm. In this study 15.6 g of waste were used for isolation of fuel particles by sedimentation in bromoform. Activity levels of gamma emitting radionuclides associated with this soil sample are given in table 1.

Table 1 : Gamma emitting radionuclides activities in trench soil (2000).

	$^{134}\text{Cs}$	$^{137}\text{Cs}$	$^{154}\text{Eu}$	$^{155}\text{Eu}$	$^{241}\text{Am}$
Activity (Bq/g)	$11.7 \pm 0.7$	$2051 \pm 139$	$12 \pm 1.7$	$7.5 \pm 1$	$19.4 \pm 2.3$

### 2.2 Methods

#### 2.2.1 Methodologie and Scanning Electron Microscopy

In order to separate fuel particles from soil minerals a methodology was developed. This methodology includes three steps [4]:

step I : 2-3 g of soil were dispersed in 30 ml of bromoform (density = 2.88) in plastic tubes (Nalgen, FEP Oak Ridge tube). After centrifugation the majority of soil components (density < 2.88) remains on the air/bromoform interface while heavy fraction (fuel particles and heavy minerals of soil) sinks.

step II : After filtration of the heavy fraction, the presence of fuel particles among the numerous heavy minerals present on the filter is confirmed by an  $\alpha$ ,  $\beta$ ,  $\gamma$  probe (Eurisys) with a sensitive area of 18 mm<sup>2</sup> (surface area of the filter is 1590 mm<sup>2</sup>). In the zone where activity was detected, the heavy particles were collected onto sticky double-faced carbon tabs and were mounted onto aluminium stubs.

step III : Aluminium stubs were analysed with Scanning Electron Microscopy (SEM, Hitachi S-3500N) interfaced with an Energy-Dispersive Spectrometer (ISIS 300). SEM analytical conditions are : accelerating voltage-20 keV, working distance-15mm. Due to the number of particles expected to be analysed (each stub containing hundreds of particles), it was decided to apply a new automatic system named GunShot, to sort out the fuel particles. The GunShot program carries out automatic searching and chemical classification of particles on standard stubs. The system uses a threshold set-up using a standard to detect particles in a backscattered electron image. Detected particles are classified and can be relocated for detailed manual review and confirmation. For this study, a <sup>22</sup>Ti/<sup>83</sup>Bi standard was specially developed.

### 2.2.2 Gamma spectrometry

Gamma emitting radionuclides were determined using Eurisys N-type HPGe detectors with 40% relative efficiency. The resolution is respectively 0.76 keV and 1.95 keV for 6 keV and 1332 keV.

## 3. RESULTS AND DISCUSSION

All fuel particles have been separated from sample and 568 of them have been characterized by Scanning Electron Microscopy. After a residence time in the trench of 14 years a large amount of particles is still present.

### 3.1 Chemical composition and morphological description of fuel particles

Based on X-ray microanalysis results, the particles can be classified into two main types : 1) fuel particles with UO<sub>x</sub> matrix (figure 1a); 2) fuel particles with Zr-U-O matrix (figure 1b).

Among the 568 fuel particles analysed, 150 have an UO<sub>x</sub> matrix and 418 have a Zr-U-O matrix. Thus, fuel particles with Zr-U-O matrix account for 74% of fuel particles in the studied sample. These particles are the result of the interaction between nuclear fuel and zircaloy tubes during melting event at the moment of the accident.

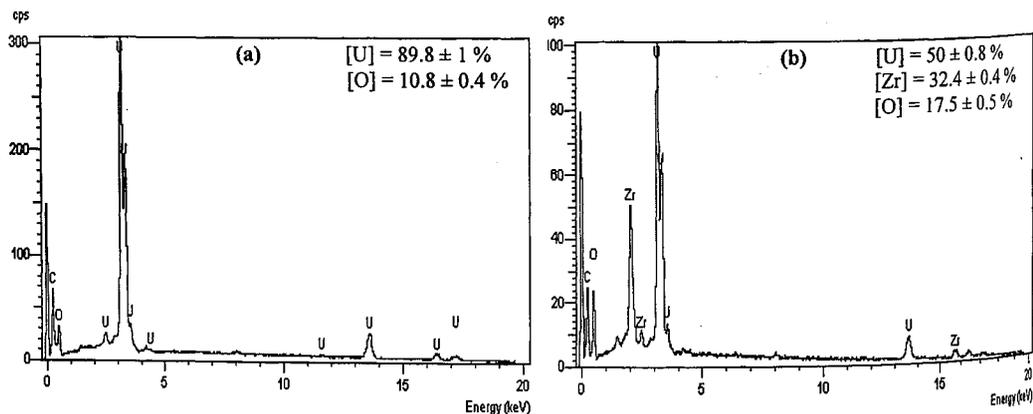


Figure 1: X-ray analysis spectrum and quantitative X-ray analysis of UO fuel particles (a) and Zr-U-O fuel particles (b).

The morphological peculiarities make it possible to single out the following particles :

### 3.1.1 $UO_x$ fuel particles with irregular morphology.

This is the basic type of particles in trench. This type of particles is characterized by the presence of numerous pores already merged together, forming large caverns on the surface (figure 2, photos 1 to 4), which may be caused by dissolution in the natural medium.

More rarely, fuel particles with individual microblock structure have been observed (figure 2, photos 5 and 6). These particles represent fragments of unmodified fuel dispersed by the initial explosion. Their shape has been preserved.

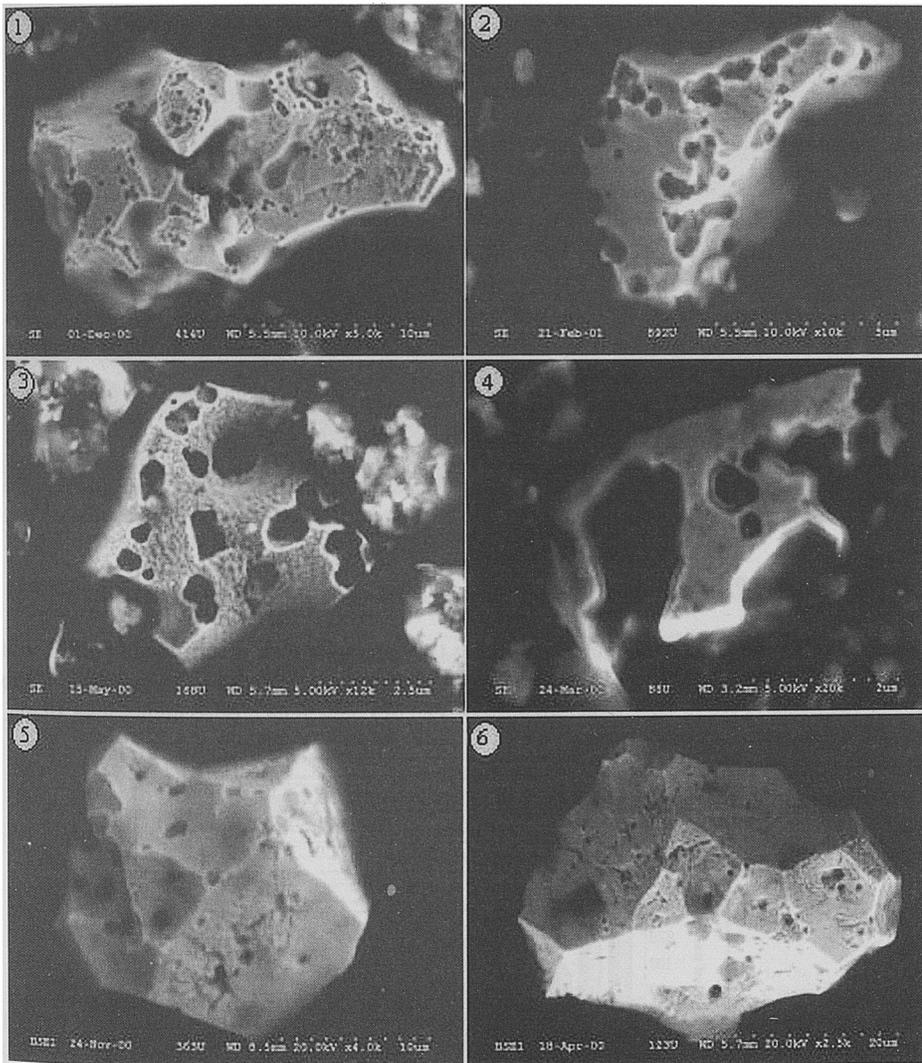


Figure 2 : Microphotograph of  $UO_x$  fuel particles with irregular morphology (photos 1to 4) and individual microblock of unmodified nuclear fuel (photos 5 and 6).

3.1.2 Zr-U-O fuel particles with irregular morphology.

The common characteristic feature of these particles is the presence of melting marks on their surface (figure 3). These particles have probably been in molten state. Some pores can be observed on some particles but their number is lower than in the case of  $UO_x$  particles (figure 3, photo 3).

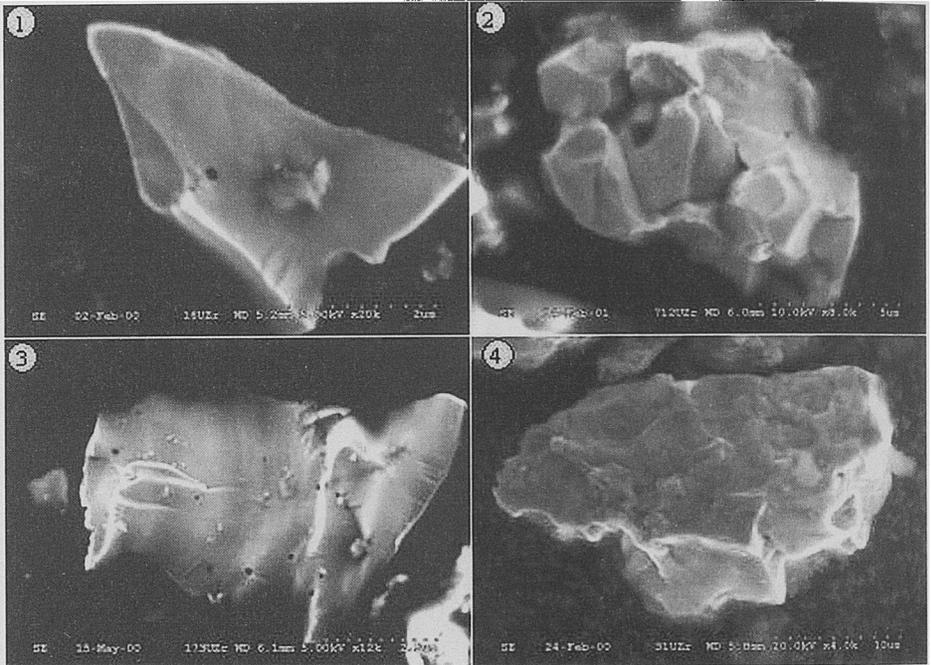


Figure 3: Microphotograph of typical forms of fuel particles with Zr-U-O matrix.

3.2 Size distribution of fuel particles

The distribution of fuel particles diameter, assuming they are spherical, determined by SEM is presented in figure 4. The size of  $UO_x$  and Zr-U-O fuel particles vary from 1.1 to 34  $\mu m$  and from 0.8 to 76  $\mu m$  respectively. However, 87% of  $UO_x$  type and 95% of Zr-U-O type have a size smaller than 10  $\mu m$ .

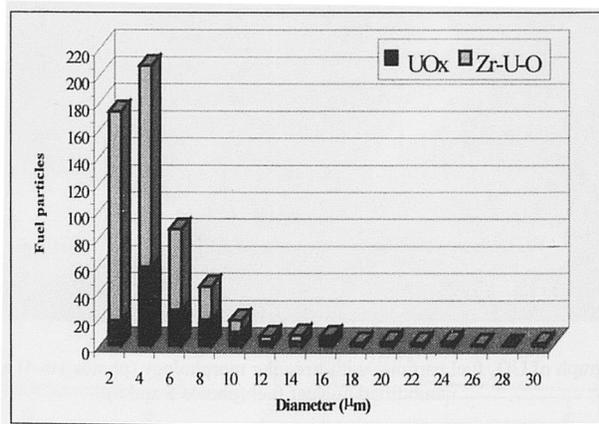


Figure 4 : Size distribution of fuel particles from trench

The size of fuel particles is assumed to be distributed following a lognormal law :

$$f(d) = \frac{1}{\sqrt{2\pi \cdot s \cdot d}} \exp \left\{ -0.5 \left[ \frac{\ln(d) - m}{s} \right]^2 \right\}$$

where

$d$  = diameter of particles ( $\mu\text{m}$ )

$m$  = 1.15 (mathematical expectations of particle diameter logarithm)

$s$  = 0.75 (mean quadrant deviation of particle diameter logarithm).

The mean diameter of fuel particles is  $4.17 \mu\text{m}$  and standard deviation is  $3.62$ .

Assuming that fuel particles are spherical, we can calculate a specific area of particles from diameter.

Thus, specific area :

$$S = \frac{6}{\rho \cdot d} = 1308 \text{ cm}^2/\text{g}$$

$d$  = mean diameter of particles

$\rho$  = density of fuel =  $11 \text{ g/cm}^3$

Specific area determined above is a minimum value. In the case of  $\text{UO}_x$  fuel particles, specific area is higher, because of presence of numerous large pores. The dissolution rate depends on the particles surface, therefore the dissolution is more effective for  $\text{UO}_x$  particles than for Zr-U-O.

### 3.3 Activity mass balance

In order to quantify the part of activity associated with fuel particles in trench, gamma activity of sample was determined before separation of fuel particles and after separation. Results show that 52 % of Cs, 70 % of Eu and 65 % of Am are still associated with fuel particles (table 2). It means that in the trench, source term is mainly represented by fuel particles. The fraction of the total Cs activity associated to fuel particles is lower than those of Eu and Am because of its high volatility during the accident. Temperature reached in a local section of the reactor was  $2500\text{-}2600 \text{ }^\circ\text{C}$  [5] .

Table 2 : Gamma emitting radionuclides activity mass balance in trench waste in 2000.

Radionuclides	Gamma activity in waste sample (Bq)		% of activity in fuel particles
	Activity before fuel particles separation	Activity after fuel particles separation	
$^{134}\text{Cs}$	183	90	51
$^{137}\text{Cs}$	32065	15452	52
$^{154}\text{Eu}$	188	58	69
$^{155}\text{Eu}$	117	35	70
$^{241}\text{Am}$	303	107	65

$^{90}\text{Sr}$  activity associated with fuel particles is calculated using measured activity of  $^{154}\text{Eu}$  and activity ratio  $^{90}\text{Sr}/^{154}\text{Eu}$  in Chernobyl nuclear fuel ( $^{90}\text{Sr}/^{154}\text{Eu} = 72$  in 2000). Results show that 69 % of  $^{90}\text{Sr}$  still are associated with fuel particles.

#### 4. CONCLUSIONS

This study has shown that the sedimentation in bromoform liquid is an efficient technique for separation and concentration of fuel particles from contaminated soils. Numerous particles have been isolated and their physico-chemical characteristics were studied by Scanning Electron Microscopy.

52 % of Cs activity, 70 % of Eu and Sr activities and 65 % of Am activity are still associated with fuel particles. Hence 14 years after Chernobyl accident, source term is mainly represented by fuel particles.

Assuming that particles are spherical their mean diameter is 4.17  $\mu\text{m}$  and therefore calculated specific area is 1308  $\text{cm}^2/\text{g}$ . The source term consists of two types of fuel particles: One type is composed of U and O while the other is composed of Zr, U, and O. Zr-U-O fuel particles account for 74% of source term. Scanning electron microscopy, showed that the  $\text{UO}_x$  particles have undergone weathering after their deposition while Zr-U-O particles showed much higher stability. This data is in agreement with high stability of natural zircon. Therefore an important part of activity will remain associated to Zr-U-O particles. Hence the transfer of mobilised associated radionuclides within the ecosystem will be delayed until weathering takes place.

Model development and modelling results of the spent fuel and Chernobyl fuel particles dissolution process considered that the source term is only made of UO. Therefore, dissolution constants from these models could not be applied to the Chernobyl source term because they would imply an overestimation of assessment of mobilisation, transfer and short term consequences of radionuclide releases. The description of Chernobyl source term weathering requires 2 dissolution laws: a first one concerning UO particles with a higher dissolution constant and a second one with a low dissolution constant describing Zr-U-O dissolution.

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