Dissolution kinetics of particles of irradiated Chernobyl nuclear fuel: influence of pH and oxidation state on the release of radionuclides in the contaminated soil of Chernobyl

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Abstract

The radioactive contamination in the exclusion Chernobyl zone is mainly due to fuel particles present on the ground. Dissolution of these particles causes leaching of radionuclides to increase with time. The kinetics of dissolution of Chernobyl fuel particles were determined in solutions of different acidities, using material obtained by crushing actual irradiated Chernobyl fuel and by its oxidation for 1–21 h in air at a temperature of 670 K. Oxidation results in superficial cracking of the particles and an increase in their surface area and, therefore, higher dissolution rates for such particles than for non-oxidised ones. Dissolution rate of fuel particles increases in acid and alkaline mediums. The dissolution rates obtained for non-oxidised and oxidised fuel particles (UO_{2+x}) in solutions of different acidities can be used as a basis for narrowing the scope of assessments for prognosis of changes in the radiological situation in the Chernobyl zone. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Radiologically significant radionuclides such as ^{90}Sr, $^{238-241}Pu$ and ^{241}Am were released from the Chernobyl reactor during the accident, mainly as fuel particles of sizes ranging from a few microns to hundreds of microns. Fuel particles were detected in the radioactive fallout both close to the reactor [1–5] and at great distances in Western Europe [6–11].

The particles of low-oxidised uranium oxide were formed due to the destruction of nuclear fuel during the explosion on 26 April 1986, which has created a narrow western trace of release. In the subsequent reactor fire, which lasted from 26 April 1986 to 5 May 1986, the nuclear fuel was oxidised and deposited mainly in the north and the south of the plant. While the release of the volatile products of nuclear fission (Kr, Xe, I, Te, Cs, etc.) increased with the temperature of the nuclear fuel, the formation of fuel particles due to the oxidation of fuel in air prevailed at temperatures of 600–1200 K (with a maximum oxidation rate at a temperature of about 900 K) [12]. Thus, even at relatively low temperatures, when the release of volatile fission products was not significant, the oxidation of nuclear fuel and further release of radionuclides associated with the matrix of fuel particles is possible in some accidental situations.

Due to the dissolution of the fuel particles, the radionuclides passed from the fuel matrix into soil solution. Being associated with the fuel particle matrix, radionuclides in initial fallout had very low migration ability compared to the same radionuclides in condensed fallout and in global fallout. This is why, in the initial period after the Chernobyl accident, the attempts to use the regularities obtained earlier for global fallout resulted in substantial overestimation of radionuclide

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migration in the near field (especially for ^{90}Sr). The quantification of the radionuclides source term is necessary to determine the evolution of the transfer of the radionuclides in the soil and thus the level of plant contamination for radiological situation prognostics and for the planning of countermeasures.

Therefore, the definition of dissolution rates of the fuel particles in different conditions is one of the most important key tasks for long-term prediction of radionuclide behaviour in the vicinity of the Chernobyl nuclear power plant and in the study of any other abnormal hypothetical situations resulting from the release of particles of irradiated nuclear fuel.

It was shown in studies of the kinetics of Chernobyl fuel particle dissolution in soil that the fuel particle dissolution rate under natural conditions is determined both by the properties of the fuel particles (oxidised or not) and by the acidity of the medium [13]. For a better understanding of the processes and to determine fuel particle dissolution rates in a wider range of parameters, experiments were carried out under strictly controlled conditions. In the experiments, described below, fuel particle oxidation and solution acidity were studied as the main factors influencing and determining the fuel particle dissolution rate.

2. Materials and methods

2.1. Preparation of samples

Two small pieces of UO_2 of the real irradiated undestroyed Chernobyl nuclear fuel were mechanically cracked from the same tablet and were used as the initial samples in the current research. The uranium dioxide had a burnup 6.5 ± 0.9 MW day/kg, a ^{235}U/^{238}U ratio of 0.0143 ± 0.0017, a density of 10.5 ± 0.9 g/cm^3 and a specific activity on 26 April 1986 of 650 ± 90 MBq/g for ^{90}Sr and 770 ± 50 MBq/g for ^{137}Cs [14].

To determine the dissolution rate of the non-oxidised fuel particles (analogous to the particles of the first Chernobyl release) the first piece of UO_2 (about 1 mg), was crushed in a quartz crucible. In order to simulate the oxidation of fuel particles during the accident, the second piece of fuel (about 5 mg), was annealed in the 50 cm^3 quartz crucible for 1 h in a muffle furnace in air at a temperature of 670 ± 20 K. As a result of annealing, the piece was crushed into small particles. The resulting powder was divided into six aliquots. Five of them were placed in separate quartz crucibles and then annealed at the same temperature as before for different periods of time (2, 4, 6, 12 and 20 h). As previously shown [14], such annealing causes negligible release of radionuclides from hot particles (<1%) even for radiocaesium (temperature of volatilisation close to 700°C). Then, the particles from each quartz crucible were washed with

100 ml of 0.001 M hydrochloric acid, stirred and filtered through a filter with a diameter of 47 mm and a pore size of 0.22 μm (Millipore MF). The washing solutions were then analysed by γ-spectrometry and beta counting. ^{154}Eu contents in these solutions were lower than the detection limit, which means that there were no submicron fuel particles in the solutions that could pass through the filter. On each filter, the particles sizes were evaluated.

2.2. Dissolution of the samples

Each filter with the fuel particles was cut into five approximately equal parts. Each part of the filter was analysed using the γ-spectrometer. ^{90}Sr activity in each sample was calculated using the results of ^{137}Cs measurements and the known ^{90}Sr/^{137}Cs activity ratio in the initial piece of fuel [14], taking into account the proportion of activity that passed into the washing solution. Each sample was placed in a plastic beaker containing 100 ml of solution of different acidity:

- pH 3: 0.001 M HCl;
- pH 4: 0.0001 M HCl;
- pH 5: 0.00001 M HCl;
- pH 6.86: 0.025 M Na_2HPO_4 + 0.025 M KH_2PO_4;
- pH 9.18: 0.01 M $Na_2B_4O_7 \cdot 10H_2O$.

The samples were shaken every two days. Once the solutions had stood for a given time (total reaction times 1, 8, 24, 42, 70, 100, 133, 182, 281 and 342 days) at room temperature (20–25°C) the solid samples were transferred to new beakers with the fresh corresponding solutions. ^{134,137}Cs, ^{154}Eu, ^{90}Sr activities had been determined in 'old' solutions as well as the activity of γ-emitting radionuclides in the solid in order to check the radioactivity balance. At the end of the experiment (342 days), the residual ^{90}Sr activity in fuel particles was measured in some of the samples. The correct radioactivity balance was found for all radionuclides. No significant difference was found in the transfer of the different radionuclides (^{134,137}Cs, ^{154}Eu, ^{90}Sr) from the particles to the solution, which shows that radionuclides had been leached because of the dissolution of the fuel particle matrix. All the calculations below are therefore shown for ^{137}Cs as its activity measurements had the smallest uncertainty (the uncertainty did not exceed 5% in any of the measurements). Solution acidity was checked using a pH-meter during all phases of the experiment, and its variation in hydrochloric acid solutions did not exceed 0.2.

2.3. Measurements

Particles size was determined by an image analysis system using BioScan Optimate software in a mode involving automatic step-by-step scanning of the filter surface.

Measurements of the activities of γ-emitting radionuclides in all samples were carried out using a γ-spectrometer with GEM-30185 semiconductor detector of high-purity germanium ADCAM-300 and a multichannel analyzer (Ortec, USA). Standard radiochemical methods and a Canberra-2400 low-background radiometer (USA) were used for measurement of ^{90}Sr activity in the samples.

Electron micrographs of the fuel particles were taken with a Hitachi S-3500N scanning electron microscope using a secondary electron detector (resolution 3.5 nm).

3. Results and discussion

3.1. Fuel particles size distribution

The median diameter of the particles obtained by crushing and annealing varied between 12 and 6 μm. It decreases to 6 μm (i.e., the size of the initial uranium dioxide crystallite) at longer annealing periods (Table 1). Size distribution of fuel particles is well described by a lognormal law [14]. These results are in good agreement with the data obtained earlier, when the size distribution of fuel particles was measured by means involving a method of sedimentation in water [14]. There is also a good agreement with the fuel particle sizes observed in the Chernobyl radioactive fallout [6,13].

^{134,137}Cs transfer into washing solutions was higher than ^{90}Sr (Table 1), which can be explained by the higher migration ability of radiocaesium in uranium dioxide matrix compared to that of ^{90}Sr during normal reactor operation. The portion of easily washable activity (Table 1) increases with the duration of annealing of the fuel particles in air. This cannot be explained by the diffusion of radionuclides on the surface of the particles at the relatively low annealing temperature (670 K), because at this temperature the diffusion of radiocaesium is very low and the diffusion of radiostrontium negligible.

Fuel particles observed from SEM analyses show that mechanical crushing causes the fuel to break into separate grains and conglomerates of grains (crystallites) with a smooth surface (Fig. 1(a)). As a result of annealing in air, the oxygen penetrates (for a period of less than 1 h) the entire grain matrix and causes it to crack (Fig. 1(b)–(f)). It significantly increases the free porosity of the fuel particles and their effective surface available to the washing solutions. Thus the measured differences are explainable in terms of different surface areas. Unfortunately a direct measurement of the surface area of fuel particles by BET is difficult because of their high activity level (mass required 2 g).

The results for easy washing of radionuclides (2–7% for ^{90}Sr and 3–10% for ^{137}Cs) from the fuel particle surface are in good agreement with analogous data for initial Chernobyl fuel fallout in the first few days of the accident [15].

3.2. Particles dissolution kinetics

The process of fuel particles dissolution has been described as a first-order kinetic equation [16–18].

$$\mathrm{d}A/\mathrm{d}t = -kA, \quad \Delta FP = A_t/A_0 = \exp(-kt), \qquad (1)$$

where ΔFP is portion of undissolved particles, and A_t and A_0 are the activity of particles at time t after deposition and at the time of initial deposition, respectively.

Results obtained in studies of the dynamics of radionuclide leaching from the matrix of fuel particles oxidised to different extents into solutions of different acidities are shown in Table 2.

Non-oxidised particles of crushed nuclear fuel have the lowest dissolution rate. In 342 days, less than 2% of activity passed from these particles to solutions of pH 5–7, and only about 6% into solutions of pH 4 and 9. It was only in acid solution of pH 3 (which is untypical in the environment) that the transfer of radionuclides reached about 30% (Fig. 2).

Fuel particles annealed in air at a temperature of 670 K dissolved much more readily than non-annealed

Table 1
Parameters of lognormal distribution of fuel particle size distribution and proportions of activity passing into the washing solution

Annealing period (h)	m^a	s^b	Median diameter of particles (μm)	Portion of activity in solution (%)		
				^{137}Cs	^{134}Cs	^{90}Sr
0	2.05	0.60	9.3	3.3	3	2.3
1	2.33	0.63	12.5	5.1	6	2.9
3	2.13	0.52	9.7	6.4	5	3.9
5	1.92	0.44	7.5	5.3	6	4.0
7	1.85	0.41	6.9	7.7	9	4.6
13	1.91	0.42	7.4	8.5	10	6.2
21	1.78	0.36	6.3	10.3	12	6.6

[a] m = mathematical expectations of particles diameter logarithm.
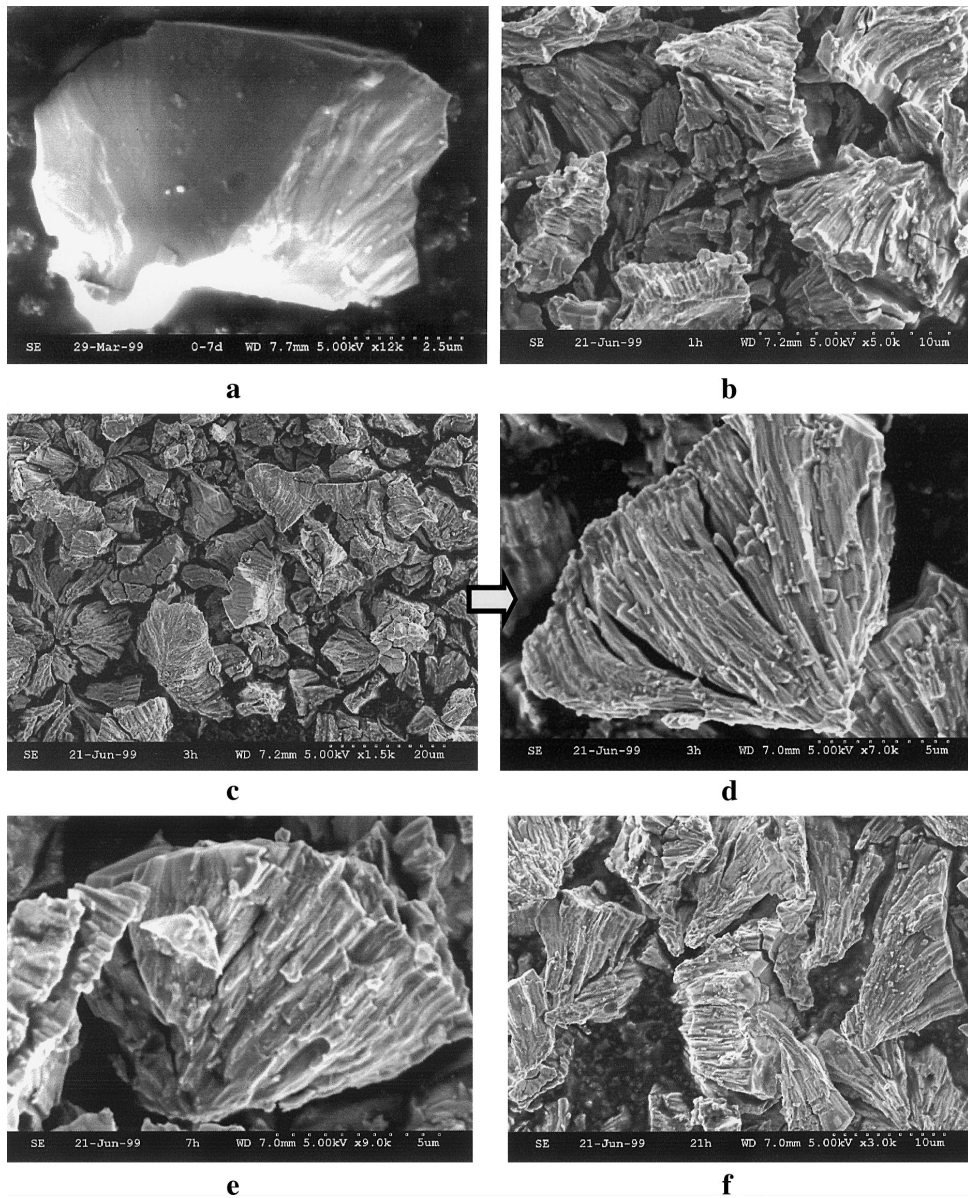[b] s = mean quadrant deviations of particles diameter logarithm.

Fig. 1. Scanning electron micrograph (SE-mode) of the particles obtained by crushing nuclear fuel (a) and by its annealing in air at a temperature of 670 K for 1 h (b), 3 h ((c) and (d)), 7 h (e) and 21 h (f).

ones whatever the pH. For instance, even in a neutral medium at pH 7 more than 10% of their activity was leached into solution in 342 days. At pH 3, most of these particles dissolved and only about 20% of activity remained in them. Varying the annealing period between 1 and 21 h induces a slight increase of dissolution of fuel particles (Fig. 2). However, it is difficult to determine a relationship between the annealing period and the oxidation degree of the fuel particles.

For both non-annealing and annealing fuel particles, the dissolution rate during the first month of the experiment was higher as compared to the following period (Table 2). This can possibly be explained by the washing out of activity that remained in the free porosity of particles after the previous washing. Besides, dissolution of fuel particles is initially more effective at the various defects of matrix structure as compared with the dissolution of deeper (more stable) layers. For this reason, with the most acid solutions (pH 3) it is better to apply a two-exponential function for description of the kinetics of fuel particle dissolution:

Table 2
Dynamics of ^{137}Cs leaching from matrix of particles of irradiated Chernobyl nuclear fuel of different oxidation into solutions of different acidity, and derived fuel particle transformation constants (k, year^{-1}) and reliability of approximation (R^2)

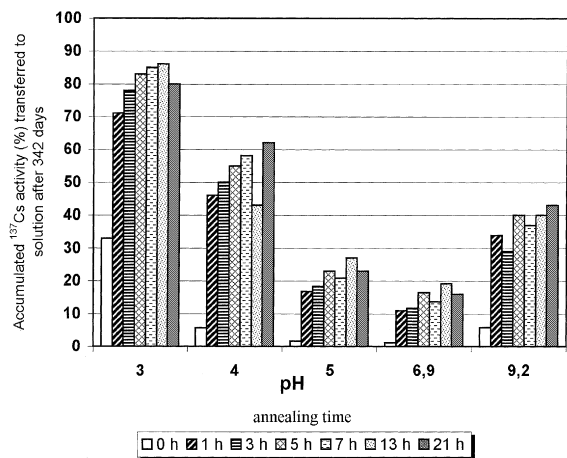| Roasting period	Acidity of solution (pH)	Portion of accumulated ^{137}Cs activity (%), transferred from FP matrix into solution after:										k (year^{-1})		R^2
		1 day	8 days	24 days	42 days	70 days	100 days	133 days	182 days	281 days	342 days	k	σk	
0	3	1.25	6.3	15.7	22	25	27	29	31	32	33	0.6	0.1	0.32
	4	0.16	0.19	0.53	0.73	1.04	1.53	1.93	2.8	4.6	5.7	0.062	0.001	0.99
	5	0.12	0.19	0.25	0.29	0.34	0.41	0.61	0.75	1.40	1.65	0.018	0.001	0.97
	6.9	0.10	0.12	0.15	0.17	0.19	0.20	0.30	0.47	0.89	1.20	0.012	0.001	0.94
	9.2	0.11	0.29	0.96	1.25	1.66	1.95	2.6	3.7	4.8	5.8	0.066	0.001	0.97
1	3	2.4	9.7	16.8	24	34	43	53	60	67	71	1.5	0.1	0.92
	4	0.58	1.37	2.4	4.6	9.4	14.1	19.8	29	40	46	0.66	0.02	0.99
	5	0.53	1.25	2.3	3.2	4.4	6.7	9.4	11.7	15.4	16.8	0.22	0.01	0.97
	6.9	0.66	1.25	2.1	2.9	3.9	4.9	6.4	7.7	9.3	11.0	0.14	0.01	0.91
	9.2	0.37	0.92	2.7	4.9	10.4	14.0	18.6	24	30	34	0.47	0.02	0.98
3	3	3.2	19.4	29	34	42	51	59	69	76	78	1.9	0.3	0.87
	4	0.44	1.22	3.7	8.1	9.8	18.8	26	32	42	50	0.73	0.02	0.99
	5	0.46	1.16	2.5	3.5	4.7	8.1	12.0	14.6	17.4	18.4	0.22	0.01	0.94
	6.9	0.64	1.22	2.0	2.8	4.3	5.4	7.1	9.0	10.6	11.7	0.15	0.01	0.91
	9.2	0.39	0.84	2.7	4.6	8.2	11.0	14.0	19.6	24	29	0.37	0.01	0.99
5	3	2.1	10.2	19.4	30	43	53	64	72	80	83	2.2	0.2	0.95
	4	0.49	1.74	3.9	7.9	10.2	19.3	28	35	48	55	0.84	0.02	0.99
	5	0.41	1.12	2.6	3.9	6.1	8.7	10.7	15.7	21	23	0.30	0.01	0.99
	6.9	0.36	1.12	2.3	3.5	5.7	7.4	9.8	11.7	14.2	16.4	0.21	0.01	0.94
	9.2	0.34	1.01	3.1	5.8	10.9	15.1	20.4	27.0	33.9	40	0.55	0.01	0.99
7	3	2.2	10.2	19.9	30	43	54	65	73	81	85	2.3	0.1	0.95
	4	0.35	1.68	3.5	6.2	8.1	14.8	23	35	50	58	0.88	0.05	0.98
	5	0.46	1.35	3.0	4.6	6.3	9.3	12.7	16.0	18.4	20.8	0.28	0.02	0.94
	6.9	0.51	1.29	2.3	3.5	5.2	6.6	8.7	10.2	12.0	13.7	0.18	0.01	0.9
	9.2	0.31	0.94	2.9	5.2	10.1	13.0	17.9	24	31	37	0.51	0.01	0.99
13	3	2.6	10.7	21	32	46	56	67	75	82	86	2.3	0.2	0.95
	4	0.78	3.1	6.0	10.9	13.5	22	24	26	40	43	0.66	0.03	0.97
	5	0.69	1.44	2.8	4.2	6.3	9.3	12.7	16.7	23	27	0.34	0.01	0.99
	6.9	0.64	1.28	2.2	3.3	5.4	7.2	9.6	12.2	16.1	19.2	0.24	0.01	0.98
	9.2	0.46	0.95	3.1	6.3	12.4	16.1	22	28	34	40	0.58	0.02	0.98
21	3	3.6	14.7	24	33	46	56	64	71	77	80	2.0	0.3	0.88
	4	0.69	3.6	5.5	10.6	13.4	22	31	40	53	62	0.99	0.02	0.99
	5	0.79	1.93	3.4	4.8	6.9	9.6	12.5	16.6	21	23	0.31	0.01	0.96
	6.9	0.80	1.67	2.5	3.6	5.4	6.9	9.1	11.7	14.9	16.0	0.21	0.01	0.94
	9.2	0.54	1.43	3.9	7.0	13.3	17.5	23	30	37	43	0.62	0.02	0.99

Fig. 2. ^{137}Cs leaching from fuel particles matrix of different oxidation state into solution of different acidity after 342 days.

$$\Delta FP = c\exp(-k_1 t) + (100 - c)\exp(-k_2 t), \qquad (2)$$

where for particles of crushed non-oxidised nuclear fuel the fast and slow components of the transformation constant at pH 3 are equal to $k_1 = 19 \pm 1$ year^{-1} and $k_2 = 0.11 \pm 0.01$ year^{-1}, and $c = (26 \pm 1)\%$ – the corresponding part of activity ($R^2 = 0.997$). For annealed particles the corresponding values are $k_1 = 5 \pm 2$ year^{-1} and $k_2 = 0.7 \pm 0.2$ year^{-1}, and $c = (70 \pm 20)\%$.

The results obtained show that the minimum fuel particle dissolution rate is found (for all degrees of particles oxidation) in neutral solutions with a pH close to 7. Fuel particles dissolution rate increases both in acid and alkaline mediums, which agrees well with the literature data [19]. Dependence of the fuel particle transformation constant on the solution acidity can be described by the function

$$k = a10^{(-\alpha pH)} + b10^{(\beta pH)}$$
$$= a\left(10^{-(\alpha pH)} + \frac{\alpha}{\beta}10^{-7(\alpha+\beta)}10^{(\beta pH)}\right) \qquad (3)$$
$$\text{for } \partial k/\partial(pH)|_{pH=7} = 0.$$

For the particles of non-annealed irradiated nuclear fuel, $a = 9 \pm 14$ year^{-1}, $\alpha = 0.5 \pm 0.1$ and $\beta = 0.6 \pm 0.2$. For oxidised fuel annealed in air for 1–21 h at a temperature of 670 K, $a = 23 \pm 5$ year^{-1}, $\alpha = 0.35 \pm 0.05$, $\beta = 0.3 \pm 0.1$.

The appearance of particles after dissolution in solutions of different acidities is shown in Fig. 3. Non-oxidised fuel particles remaining in neutral solution for 342 days do not differ from particles before dissolution (Fig. 3(a)). It seems that this transfer to solutions of less than 2% of their activity during the period of observation can be related to the washing of radionuclides from the surfaces of the particles. Dissolution of analogous

particles in an acid medium (pH 3) resulted only in surface corrosion (Fig. 3(b)). Annealed fuel particles crumbled into small pieces even in neutral solutions (Fig. 3(c) and (d)). Dissolution of these particles is observed at structure defects throughout the particles. This creates specific 'filamentary' structures (Fig. 3(f) and (g)). The effective surface of such particles is much higher than their observed external geometrical surface.

This seems to be the reason why the first-order kinetic equation adequately describes the process of dissolution of oxidised fuel particles both under natural conditions [13] and in laboratory mediums even without consideration of their real initial particle size distribution.

The dissolution rates obtained both by laboratory experiments and field data [13] are gathered on Fig. 4. For annealed fuel particles, the dissolution rates are higher than those of oxidised fuel particles deposited on soils (north and south direction). Conversely, for non-annealed fuel particles, the dissolution rates obtained in laboratory are lower (Fig. 4). Several explanations may be suggested:

- First, it is possible that the fallout of particles in the north, south and west directions are not constituted of pure forms, respectively, oxidised (north and south) or non-oxidised (west). We can assume that different kind of particles with different oxidation degrees have been deposited on the soils. A study devoted to the isolation of hot particles from the peaty soil Chistogalovka (located 4.4 km WSW of the Chernobyl nuclear power plant) [20] has shown that some of these particles present traces of fusion on their surfaces due to a high temperature during the explosion which has induced an oxidation of the fuel.
- During the accident, the fuel particles were in contact with different substances at high temperature, which could result in the formation of complex chemical species with dissolution kinetics different from those of uranium oxides (for example particles with surfaces covered with zirconium).
- The chemical compositions of soil solutions differ under natural conditions from the chemical compositions of solutions used in the experiments.

Nevertheless, these results allow to estimate the maximum and minimum dissolution rates of fuel particles for different pH whatever their oxidation states.

4. Conclusions

Radionuclide leaching rates from fuel particles deposited after the Chernobyl accident are dependent on both the physico-chemical characteristics of particles and environmental conditions. The results presented in this paper show the influence of fuel particles oxidation and solution acidity on the fuel particle dissolution rate.
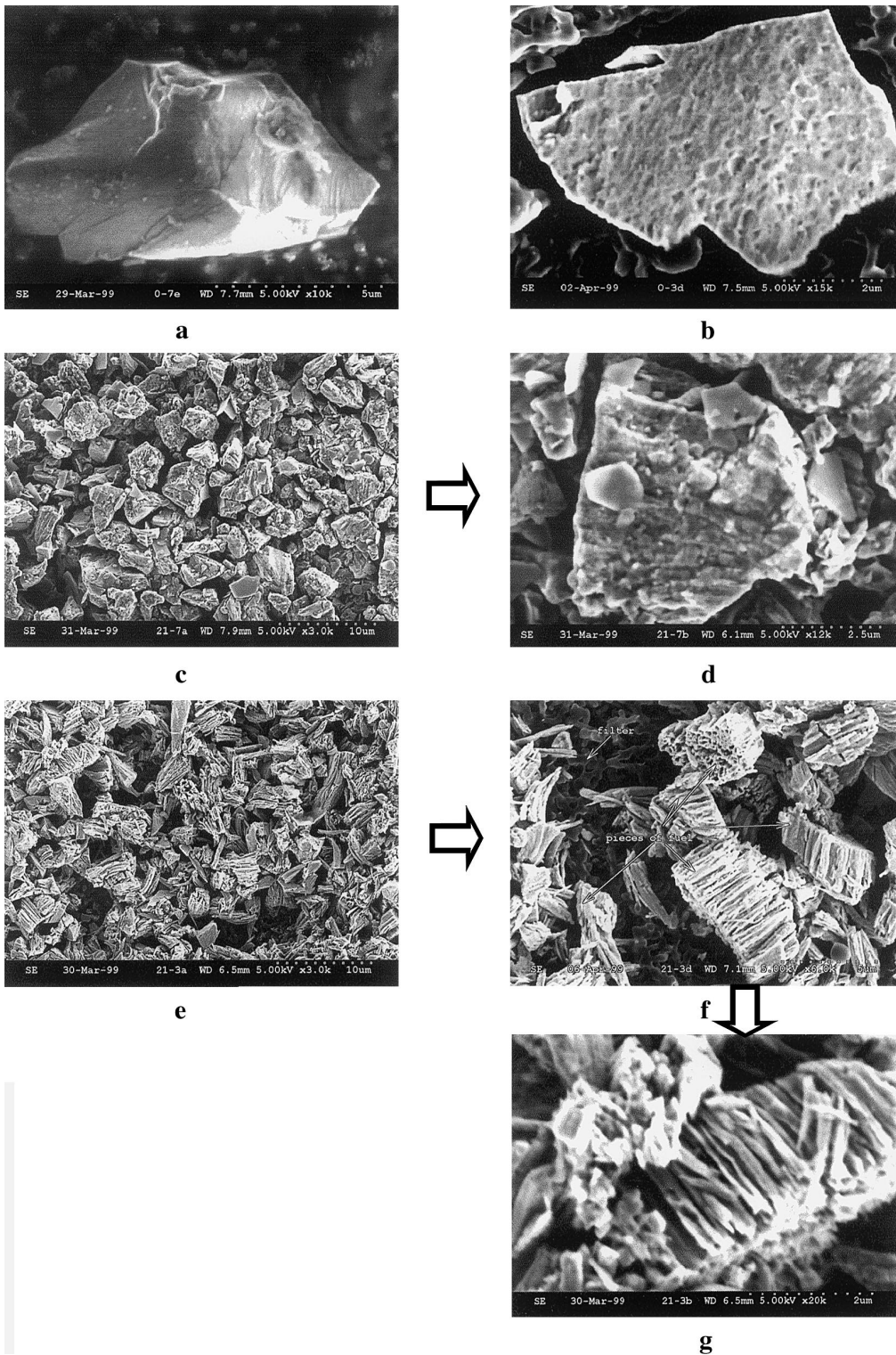
Fig. 3. Scanning electron micrograph (SE-mode) of the particles obtained by crushing nuclear fuel ((a) and (b)) and by its annealing in air for 21 h fuel ((c)–(g)) after 342 days dissolution in solutions of pH 7 ((a), (c) and (d)) and pH 3 ((b), (e)–(g)).
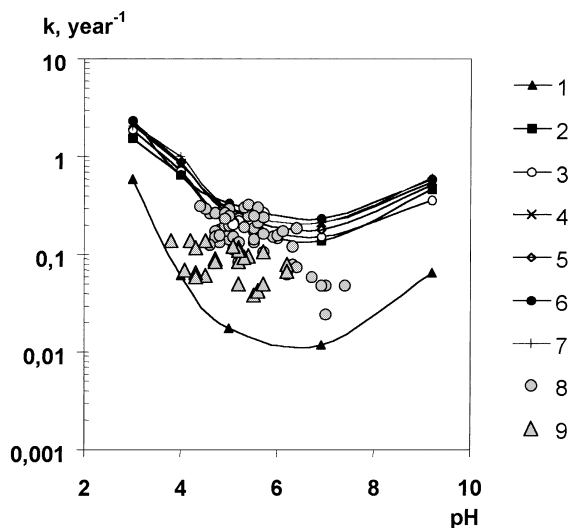
Fig. 4. Dependence of the transformation constants on the acidity of solutions (soil, 8.9) for particles of irradiated nuclear fuel: 1 – non-annealed fuel particles; 2 – annealed for 1 h; 3 – 3 h; 4 – 5 h; 5 – 7 h; 6 – 13 h; 7 – 21 h; 8 – dependence for the real Chernobyl fuel particles deposited at the northern and southern and 9 – western trace of release.

The most significant factor influencing the fuel particle dissolution rate is oxidation of the particles. Oxidation causes superficial cracking of the particles which greatly increases their effective surface and, therefore, their dissolution rate. The process of dissolution of fuel particles, both under natural conditions and in laboratory mediums, is adequately described by a first-order kinetic equation [13]. Fuel particles oxidised for 1–21 h in air at a temperature of 670 K have transformation constant values in solutions of pH 4–9 that are greater by one order of magnitude than analogous values for non-oxidised fuel particles obtained by crushing the fuel. The dissolution rate for both oxidised and non-oxidised fuel particles is minimal in a neutral medium, while in acid and alkaline mediums it increases. Dissolution of oxidised fuel particles results in their crumbling into small pieces which exhibit 'filamentary' structures and greater areas. This can explain the good fit of the first-order kinetic equation to the process of fuel particle dissolution in various solutions.

The parameters obtained for dissolution of non-oxidised and oxidised fuel particles (UO_{2+x}) in the solutions of different acidities can be used to estimate changes in the radiological situation in the Chernobyl zone and in cases of radiation accidents resulting in the release of particles of irradiated nuclear fuel. For a quantitative prediction of radionuclides release by dissolution of fuel particles, a precise determination of their oxidation degree and surface area is necessary. The acquisition of these parameters requires complementary studies.

On the basis of the results obtained, the different admixtures can, if necessary, be applied to modify the acidity of a medium, as an effective countermeasure for decreasing fuel particle dissolution in the environment and thus, inhibition of the leaching of radionuclides into mobile forms and their involvement in processes of biomigration and migration in ground and surface water.

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