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SIMULATION OF PHYSICAL PROPERTIES OF FUEL CONTAINING MATERIALS

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Numerical simulation of the dielectric permeability, electric fields and mechanical strains in the fuel containing masses (FCM) is carried out. Method of the determination of water content in the FCM by measuring the dielectric permeability is suggested. Analysis of 1) – processes leading to the appearance of electric fields; 2) – defect formation due to the irradiation by the incorporated radionuclides; 3) – mechanical strains emerging due to the different thermal expansion coefficient of the constituent components of the FCM is fulfilled. Estimates show that the third process may significantly influence degradation of the FCM.

1. Introduction

Fuel containing masses (FCM) on the premises of the 4 th unit of the Chornobyl Nuclear Plant are a unique object, that is a mixture of concrete, nuclear fuel and metal constructions in the fields of intense radiation. Study of the FCM is important for determination of the nuclear and radiation safety of the object, reproduction of the accident scenario, forecast of the FCM behaviour in future, understanding the properties of this new object. While the study of the element content and the structure of the FCM was the subject of numerous publications, there are practically no data on the physical parameters of the FCM (heat capacity, thermal conductivity, electroconductivity and mechanical properties) important for their description. This paper deals with the numerical simulation of some physical characteristics of FCM and suggests the methods for determination of their macroscopic properties. Special attention is given to the determination of the mechanisms leading to the degradation of the FCM, namely, electric fields, defect formation and mechanical strains emerging due to the gradual cooling of the masses.

2. Dielectric permeability of the FCM. Method of measuring the water content from data on dielectric permeability

One of important characteristics of condensed matter is dielectric permeability (ε). Calculation of permeability for many-component mixtures, which are FCM, is a very complicated task. For the calculation of effective dielectric permeability ε_e in the two-component system we can use the effective-medium theory (EMT) [1]. Extending the EMT to many component systems we obtain an equation for the calculation of ε_e in the form

$$\varepsilon_e = \frac{z_1}{z_2}, \tag{2.1}$$

where

$$z_1 = \sum_{i=1}^{N} \frac{p_i \varepsilon_i}{\varepsilon_i + 2\varepsilon_e}, \tag{2.2}$$

$$z_2 = \sum_{i=1}^{N} \frac{p_i}{\varepsilon_i + 2\varepsilon_e}. \tag{2.3}$$

Here N is the number of the components in the system, p_i is the volume part of the components with dielectric permeability ε_i. We can obtain the solution of equation (2.1) using the iteration method. Practice shows that the iteration series for ε_e converge very rapidly for any ratio of components in the system. Let us consider the FCM which consist of a matrix with a random system of penetrating pores. The matrix consists of SiO_2 and 9 random components: K_2O, CaO, MgO, Al_2O_3, ZrO_2, U, FeO, UO_2 and Fe_2O_3. Percentage of the components is taken from [2]. With the decrease of temperature below the boiling temperature water can penetrate into the pores. Estimation of the water quantity which may occur in the FCM as a moderator of neutrons is very important for the control of nuclear safety when the topology of the pores is random. The rest of the pores volume is unattainable for water and empty (filled with gas). It is very important to estimate the maximum degree of filling with water for the given sample of the FCM.

We constructed a computer program for calculating the maximum system volume degree of filling with water by measuring the effective dielectric permeability and comparing it with the theoretical results.

We used the following input data for the program: the measured volume density and dielectric permeability of a FCM sample in the absence of water in pores (a dried sample) and in the presence of water in pores, a set of data for mass densities and weight parts of matrix components.

At the output of the program we have the following data: the ratio of the pore volume filled with water to the entire volume of the sample, the effective dielectric permeability of the FCM in the presence of water in pores. By measuring the effective dielectric permeability of the system in case of filling up with water and comparing one with the theoretical results we find the maximum degree of pores filled with water.

The proposed method is more precise comparing with the measured weight method due to the large value of dielectric permeability (figure 1).

3. Analysis of factors affecting mechanical destruction of FCM

3.1. Electric fields in fuel containing materials

One of the possible mechanisms of the reduction of mechanical durability of FCM is the occurrence of significant electrical fields as a result of accumulation of charges generated by ionizing particles. The possibility of electrical breakdowns, occurrence of mechanical stresses (electrostriction), movement of charges and dislocations can result in the growth of cracks, voids and facilitate the material destruction. Therefore, it is necessary to study the electrical properties of FCM and to estimate the influence of radioactivity on the accumulation of charges [3] and on electrical fields in FCM.

The electrical properties of media with the nuclear fuel and radioactive nuclides differ from the properties of those without the activity. First, electrical conductivity depends on the activity $A(t)$, that is connected both with the direct introduction of carriers at the interaction of α-, β-particles and

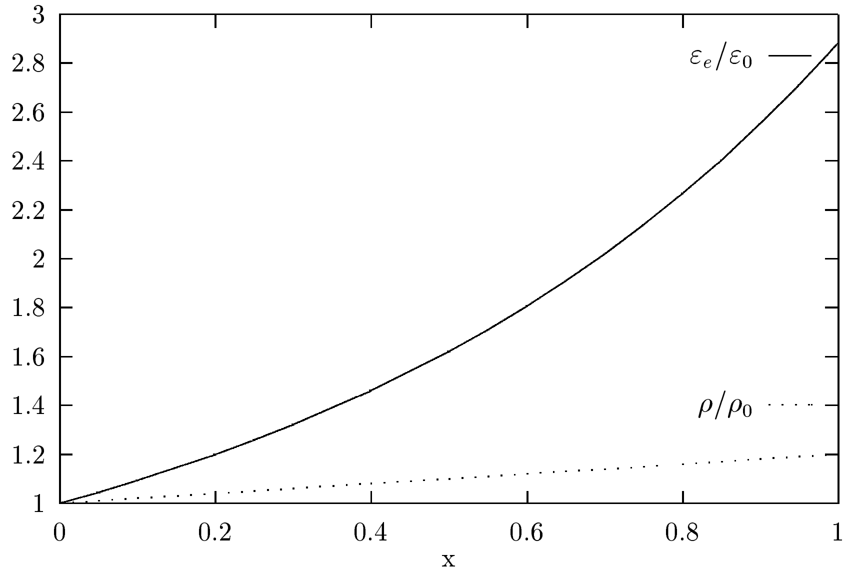

Figure 1. Theoretical dependence of effective dielectric permeability ε_e and mass density ρ of the sample (curves 1 and 2) on the ratio of the pores volume filled with water to the entire pores volume X in FCM.

γ-quants with the substance and with the introduction of radiation damages. Second, FCM are a strongly non-uniform medium and there are space charge regions (SCR) and electrical fields in them. A general model of FCM is a SiO_2 matrix with impurities, structural defects, inclusions of the various nature, radionuclides, nuclear fuel [2]. The microstructure of FCM is very complex, the electro-physical parameters and characteristics of the defect structure, which determine the distribution of carriers and their transport, are unknown. Therefore, only estimations are possible and at the present stage of the research it is expedient to use simple models with the minimal number of parameters.

Below we consider and compare different mechanisms of the formation of electrical fields in FCM.

3.1.1. Nonuniform distribution of defects (traps, recombination centres, radionuclides)

To estimate the electric fields let us consider the Poisson equation

$$\frac{\mathrm{d}^2\varphi}{\mathrm{d}^2 x} = \frac{4\pi e^2}{\varepsilon}(N_t^-(x) + n(x) - p(x)),$$

where concentrations of electrons, holes and charged traps are the following:

$$n(x) = n_0 \exp\left(\frac{e\varphi}{k_\mathrm{B}\,T}\right), \quad p(x) = p_0 \exp\left(-\frac{e\varphi}{k_\mathrm{B}\,T}\right),$$

$$N_t^-(x) = N_t(x) f\left(\frac{(E_t + e\varphi - \mu)}{k_\mathrm{B} T}\right),$$

f is the Fermi-Dirac function, μ is the Fermi energy, k_{B} is the Boltzmann constant. Setting the spatial distribution of the centres as $N(x) = N_0 + N_t \sin(kx)$, where k characterizes the spatial scale of nonuniformity, it is possible to obtain the solution to the Poisson equation as follows:

$$\varphi(x;k) = 4\pi e N_t \frac{\sin(kx)}{k^2 + k_d^2},$$

where $k_d = \frac{8\pi e^2 n_0}{\varepsilon k_{\mathrm{B}} T}$.

In case $k \gg k_d$, which is realized in high-resistivity materials, for the amplitude of the electric field we obtain

$$E = \frac{4\pi e N_t}{\varepsilon k}.$$

At $k = 10^2$ cm^{-1}, $N_t = 10^8$ cm^{-3}, we have $E = 0,46$ V/cm.

3.1.2. FCM as a strong doped disorder system at a large degree of compensation

In this case it is possible to use the results of [4]. The value of the fluctuation potential is

$$\varphi = \frac{e}{\varepsilon} N^{\frac{2}{3}} n^{-\frac{1}{3}},$$

where $N = N_a + N_d$ is the total concentration of donors and acceptors.

For the value of the fluctuation electric field one can obtain

$$E = \frac{e}{\varepsilon} N^{\frac{1}{3}} n^{\frac{1}{3}}.$$

At $N = 10^{18}$ cm^{-3}, $n = 10^9$ cm^{-3}, $\varepsilon = 3,9$ we obtain $E = 18,5$ V/cm.

3.1.3. Fields connected to inclusions of the other phase

In FCM various inclusions (UO_2, metal spherical inclusions, voids and others) are observed [2]. At interfaces SiO_2 /inclusion SCR can arise, caused by a contact-potential difference. For the interface SiO_2/UO_2 we have $\varphi \simeq 2$ V. Near the flat interface one obtains $E \approx \varphi/L_d \approx 40$ V/cm at $L_d = 0,05$ cm. For a spherical inclusion, if the condition $L \gg R$ holds, and R is an inclusion radius, it is possible to obtain $E(r) \approx 3\varphi/R$. For $R = 10^{-3}$ cm we obtain $E = 6 \cdot 10^3$ V/cm.

3.1.4. Fields connected to regions of radiation damages

Macroscopic regions of radiation damages are formed under the action of charged high-energy particles. We consider the contribution from α-particles, assuming that they dominate in the activity of a sample. It is known that for α-particles the ionization losses dominate, and the displacement cascades will be formed with a small density of defects. The average number of defects in a cascade can be estimated by using the formula [5]:

$$\nu = \frac{E_{\max}}{2(E_{\max} - E)} \left(1 + ln\left(\frac{E_{\max}}{2E_{\mathrm{d}}}\right)\right),$$

where $E_{\max}$ is the maximum energy transmitted to an atom by a particle with energy E at scattering. For instance, at $E = 3$ MeV, $E_{\max} = 797$ MeV, $E_d = 20$ eV, one obtains $\nu \approx 6$. The probability of the cascade creation with a large number of defects is small. So for the ratio of the cross-section of the formation of the cascade with the number of defects $\nu = sE_{\max}/E_{\mathrm{d}}$ to the total cross-section of the defect creation one can obtain

$$p = \frac{\sigma(\nu)}{\sigma_{tot}} = \frac{1-s}{s} \cdot \left(\frac{E_{\mathrm{pmax}}}{E_d} - 1\right)^{-1}.$$

For $s = 0.1$, $E = 30$ eV, $E_{\mathrm{pmax}} = 0.3$ MeV we obtain $p = 0.9 \cdot 10^{-3}$. Thus, a small number of damages in comparison with the damages existing in the material is introduced. Their role in the formation of electric fields is small in comparison with other mechanisms.

3.1.5. Accumulation of charge in "hot" particles

The distribution of the electric field around a "hot" particle is determined by the equality of the current of charges emitted from the 'hot' particle and reverse current induced by the electric field in the surrounding matrix. One can obtain the expression

$$E = Aze\rho_0(4\pi R^2)^{-1}$$

for the electric field at the 'hot' particle/SiO_2 matrix interface, where A is the total activity of the 'hot' particle, ze is the charge of the emitted particle, ρ_0 is the matrix resistivity, R is the 'hot' particle radius. For example, taking $\rho_0 = 10^{12}$ Ohm·cm, $R = 1$ μm, $A = 1$ s^{-1}, $z = 2$ one obtains $E = 2.55$ V/cm.

Thus, the most essential mechanisms for the appearance of the electric fields are

- charge fluctuations in condition of the strong compensation,
- contact potential at the interface of the inclusions of different phases and,
- accumulation of charges in and nearby the 'hot' particles.

The magnitudes of the fields can reach the values of about $10^2 \div 10^3$ V/cm.

4. Radiation induced defect formation in FCM as a result of internal irradiation due to radioactive elements

In fact, high dozes of internal irradiation may result in the change of mechanical properties of FCM. Intensity of the radiation damage is generally evaluated by the number of displacements per atom per second K. Let us estimate this value for FCM. The main contribution to this parameter comes from α-particles, while oxygen and silicon atoms, which have the highest percentage in the FCM composition, are displaced most easily. Therefore, it is sufficient to calculate the value of K in the vicinity of UO_2 fuel inclusion which is the source of α-particles.

Let $\boldsymbol{r}$ be the coordinate of a target atom, $\boldsymbol{r}'$ – the coordinate of a radioactive nucleus (α-activity will be considered from now on), $\mathcal{E}(\boldsymbol{r}, \boldsymbol{r}')$ – the energy of the α-particle emitted by the nucleus at point $\boldsymbol{r}'$ at reaching point

$\boldsymbol{r}$ of the target. The number of α-particles falling per unit of time onto the unit of surface at point $\boldsymbol{r}$ is equal to

$$\frac{\mathrm{d}N}{\mathrm{d}\mathcal{E}_1} = \int \frac{A\rho(\boldsymbol{r}')}{4\pi|\boldsymbol{r}-\boldsymbol{r}'|}\delta\left(\mathcal{E}_1 - \mathcal{E}(\boldsymbol{r},\boldsymbol{r}')\right)\mathrm{d}\boldsymbol{r}', \tag{4.1}$$

where A is the activity and ρ is the density of fuel inclusions.

Since the free path of the first generation of recoiled atoms is much smaller than the path of α-particle, one can suggest that the defects generated by the cascade also pertain to the region of the primarily displaced atoms.

Then the displacement rate at point $\boldsymbol{r}$ is determined by the formula

$$K(\boldsymbol{r}) = \int_0^{\mathcal{E}_0} \mathrm{d}\mathcal{E}_1 \int_{\mathcal{E}_d}^{\Lambda\mathcal{E}_1} \mathrm{d}\mathcal{E}_2 \nu(\mathcal{E}_2)\sigma(\mathcal{E}_1,\mathcal{E}_2)\frac{\mathrm{d}N}{\mathrm{d}\mathcal{E}_1}, \tag{4.2}$$

where $\nu(\mathcal{E}_1)$ is a cascade function and $\sigma(\mathcal{E}_1,\mathcal{E}_2)$ is an energy transfer cross section. Index 1 refers to the incident particle and index 2 designates the target atom.

For the region of $\boldsymbol{r}$ close to the inclusion surface where the effect is maximal equation (4.2) may be reduced to the form

$$K(0) = -\int_0^{\pi/2} \sin\theta' \mathrm{d}\theta' \int_{\mathcal{E}_1(\theta')}^{\mathcal{E}_0} \mathrm{d}\mathcal{E}_1 \frac{A\rho}{2(-\frac{\partial \mathcal{E}_1}{\partial r})} \int_{\mathcal{E}_d}^{\Lambda\mathcal{E}_1} \nu(\mathcal{E}_2)\sigma(\mathcal{E}_1,\mathcal{E}_2)\mathrm{d}\mathcal{E}_2, \tag{4.3}$$

where $\mathcal{E}_1(\theta)$ is determined by the condition

$$2R\cos\theta = \int_{\mathcal{E}_1(\theta)}^{\mathcal{E}_0} \frac{\mathrm{d}\mathcal{E}_1}{-(\frac{\partial \mathcal{E}_1}{\partial r})}, \tag{4.4}$$

$$\Lambda = \frac{4M_1M_2}{(M_1+M_2)^2}, \tag{4.5}$$

R is the radius of inclusion, $-\frac{\partial \mathcal{E}_1}{\partial r}$ is the braking ability depending on the energy of the particle (and for this problem on the coordinate as well).

Equation (4.3) may be simplified in two limiting cases.

a). The size of the inclusion is much smaller than the particle free path. Then integrating over $\mathcal{E}_1$ in (4.3) and (4.4) is performed in the narrow energy interval $\sim \mathcal{E}_0$, where $\mathcal{E}_0$ is the energy of α-particle at emission. After quite a simple reduction one may obtain

$$K(0) = \frac{A\rho R}{2} \int_{\mathcal{E}_d}^{\Lambda\mathcal{E}_0} \nu(\mathcal{E}_2)\sigma(\mathcal{E}_1,\mathcal{E}_2)\mathrm{d}\mathcal{E}_2. \tag{4.6}$$

In this case the number of displacements is proportional to the radius of the fuel particle.

b). The size of the inclusions is much greater than the particle free path. Then one can obtain from (4.3) and (4.4):

$$K(0) = \frac{A\rho}{2} \int_0^{\mathcal{E}_0} \frac{d\mathcal{E}_1}{-(\frac{\partial \mathcal{E}_1}{\partial r})_{\mathcal{E}_d}} \int_{\mathcal{E}_d}^{\Lambda\mathcal{E}_1} \nu(\mathcal{E}_2)\sigma(\mathcal{E}_1,\mathcal{E}_2)\mathrm{d}\mathcal{E}_2. \tag{4.7}$$

In this case the number of displacements does not depend on the radius and attains the same value as one at the surface of the semi-infinite inclusion. The effect is maximal here and that is why this case will be studied below in detail.

Let us choose the cascade function in the form suggested by Kinchin and Pease $\nu(\mathcal{E}) = \frac{\mathcal{E}}{2\mathcal{E}_\mathrm{d}}$. In the case of a screened Coulomb interaction between the target nucleus and α-particle the energy transfer cross section is equal to

$$\sigma(\mathcal{E}_1, \mathcal{E}_2) = \frac{\pi M_1}{M_2} \frac{(Z_1 Z_2 e^2)^2}{(\mathcal{E}_2 + \mathcal{E}_\lambda)^2 \mathcal{E}_1} \ , \tag{4.8}$$

where M_1 and Z_1 are the mass and the charge of the incident particle, M_2 and Z_2 are the corresponding values for the target atom, $\mathcal{E}_\lambda = \frac{\hbar^2 \lambda^2}{2M_2}$, $1/\lambda$ is the radius of screening the Coulomb interaction between the nuclei.

The main contribution to the integral of equation (4.3) gives the energy regions where the braking ability is small. They are: the region of high energies where the losses are determined by the electron braking and $-\frac{\partial \mathcal{E}_1}{\partial r} \sim v_1^2 \ln V_1^2$, and the region of low energies where $-\frac{\partial \mathcal{E}_1}{\partial r}$ with $\mathcal{E}_1 \to 0$ and the main losses are due to the energy transfer to the motion of target atoms (for the displacement of atoms in particular).

$$K(0) = K_\infty + K_0, \tag{4.9}$$

where

$$K_\infty = \frac{A \rho Z_2^2 m_e \mathcal{E}_0}{8 Z_2 n_2 M_2 \mathcal{E}_\mathrm{d}} \ln \left(\frac{2 M_1 M_2}{(M_1 + M_2)^2} \frac{M_1}{m_e} \frac{I}{\mathcal{E}_\mathrm{d}} \right) \tag{4.10}$$

$$K_0 = \frac{A \rho \tilde{M}_2}{4 \mathcal{E}_\mathrm{d} n_2 M_2} \left(\frac{Z_2}{\tilde{Z}_2} \right) \mathcal{E}_{\lambda \mathrm{q}}, \tag{4.11}$$

n_2 is the density of target nuclei in the braking region (in the region of active penetration), $\mathcal{E}_{\lambda\mathrm{q}}$ is the value of energy up to which the nucleus braking dominates, $\tilde{Z}_2$ is the nucleus charge in the region of braking, m_e is the electron mass.

Let us make the estimates. According to [6] $\mathcal{E}_\mathrm{d} = 16.5$ eV. By assuming $A = 2.7 \cdot 10^7$ s^{-1}, $\rho = 10.5$ g/cm^3, $\mathcal{E}_0 = 5 \cdot 10^6$ eV one obtains for the creation of the displacement vacancy of the oxygen atom

$$K(0) = 5 \cdot 10^{-12} \ \mathrm{dpa/s}. \tag{4.12}$$

The similar value is obtained for the creation of the defect involving silicon atom displacement. The value of K is small. Therefore, one may conclude that the radiation damage is not significant enough to affect the changes of the mechanical properties of FCM. Meanwhile, other properties of FCM may be affected (structure of electron levels, conductivity, optical properties). These problems require additional studies.

5. Strains in fuel containing masses (FCM) emerging at cooling

Let us consider an inclusion in FCM that was formed in the melted lava at high temperature T_0. It is natural to assume that during the formation of the inclusion at high temperature mechanical strains are absent. Due to the different thermal expansion coefficients of the FCM matrix and the material of the inclusion, mechanical strains appear to be leading to microcracks and finally to the destruction of FCM.

Let us consider mechanical strains that appear in the vicinity of the spherical inclusion. Owing to the spherical symmetry of the problem the equilibrium equation reads

$$\nabla \mathrm{div} \boldsymbol{U} = 0, \tag{5.1}$$

and the general solution outside the inclusion is

$$U_r = a_g r + b_g / r^2. \tag{5.2}$$

Here $\boldsymbol{U}$ is the displacement field vector. Index g denotes FCM here. Inside the inclusion

$$U_r = a_m r. \tag{5.3}$$

and index m denotes the material filling the inclusion.

Displacement field should satisfy boundary conditions at the inclusion-matrix interface and at infinity. We will allow the FCM expand or contract at infinity, so the strains should decrease to zero with increasing. At the inclusion-matrix interface we assume continuity of the displacement field an equality of the pressure at both sides of the boundary.

Field of mechanical strains is calculated by the formula

$$\sigma_{ik} = -K\alpha(T - T_0)\delta_{ik} + KU_{ll}\delta_{ik} + 2\mu(U_{ik} - 1/3\delta_{ik}U_{ll}) \ . \tag{5.4}$$

Thus we obtain outside the inclusion

$$\sigma_{rr} = -K_g a_g (T - T_0) + 3a_g K_g - \frac{8b_g}{3r^3}\mu, \tag{5.5}$$

and inside it

$$\sigma_{rr} = -K_m \alpha_m (T - T_0) + 3K_m a_m. \tag{5.6}$$

Satisfying the boundary conditions the mechanical strains around the inclusion are

$$\sigma_{rr} = -\frac{8}{3}\mu_g (T - T_0)\frac{R^3}{r^3} K_m \frac{\alpha_m - \alpha_g}{3K_m + 8\mu_g/3}. \tag{5.7}$$

One can clearly see from the obtained formula that the mechanical strains are proportional to the difference of the temperatures, to the difference of the expansion coefficients of the inclusion and the material of the matrix. It is interesting to note that the strains at the boundary do not depend on the radius of the inclusion.

Let us make some estimates that will show magnitudes of the appearing strains. It is widely known that FCM were formed from the lava at the temperature of order 1500 ^{o}C. At present the temperature of FCM is about 50 ^{o}C. For the estimates we will take silicon dioxide (which is the main constituent part of FCM) as the matrix material and will consider an iron inclusion in it for the SiO_2 matrix to be about $6 \cdot 10^{-6}$ K^{-1} and for the Fe inclusion – $12 \cdot 10^{-6}$ K^{-1}.

Thus we obtain that the emerging strains may reach the values up to $10^{-2}\mu g$ at the boundary of the inclusion. Is this enough for cracking FCM? Unfortunately, this question cannot be answered unambiguously. The reason for this is, in fact, that we have to deal with the glass-like substance. Destruction of glass is a very complicated problem. The fragility of glass in the everyday life is well known. Yet theoretical estimates for the fragility threshold of SiO_2 give the value by 2 orders of magnitude greater. This discrepancy is usually explained by the theory of microcracks network inside the glass bulk. That theory allows one to reduce the fragility threshold by 2 orders of magnitude. And taking into account those results, as well as the experimental values of the fragility threshold for the glass not specially treated, we obtain that the strains emerging due to the difference in thermal coefficients are of the order of magnitude of the fragility threshold.

Therefore, one may conclude that the mechanical strains appearing in FCM during their cooling may not be strong enough to promote cracking of the material. This mechanism is one of the most probable reasons for the gradual destruction of FCM.

6. Conclusions

Therefore, of all the considered physical processes the most significant for the degradation of FCM is the emergence of the mechanical strains that appear at the FCM cooling from the high post-accident temperatures to the present ones due to the difference of the thermal expansion coefficients of the FCM components.

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ЧИСЕЛЬНЕ МОДЕЛЮВАННЯ ФІЗИЧНИХ ВЛАСТИВОСТЕЙ ПАЛИВОМІСНИХ МАТЕРІАЛІВ

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Виконано чисельне моделювання діелектричної проникності, електричних полів та механічних напруг в паливомісних матеріалах (ПММ). Запропоновано метод визначення вмісту води в ПММ з вимірів діелектричної проникності. Проаналізовано

1) – механізми, що ведуть до появи електричних полів, 2) – дефектоутворення під дією опромінення інкорпорованими радіонуклідами, 3) – механічні напруги, що виникають внаслідок різних коефіцієнтів теплового розширення компонент ПММ. Оцінки показують, що механічні напруги можуть істотно впливати на деградацію ПММ.