SOURCE TERM ASSESSMENT OF SPENT FUEL IN WET STORAGE

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Estimarea activităţii combustibilului nuclear uzat depozitat în bazinele cu apă, este importantă pentu analiza degradării tecii de combustibil datorită coroziunii. Această estimare permite personalului de operare să coreleze valoarea activităţii apei din bazine cu numărul de elemente care prezintă defecte de teacă. Prezentul articol prezintă rezultatele obţinute în estimarea activităţii combustibilului nuclear uzat EK-10 şi C-36 ce este depozitat la Măgurele. Pentru estimarea stării termenului sursă s-au utilizat de coduri de calcul ce valorifică istoria exploatării combustibilului. Calculele sunt importante pentru estimarea nivelului radionuclizilor (produşi de fisiune, gaze de fisiune, produşi de activare) din interiorul combustibilui uzat atsfel încât să poată fi luate măsurile necesare pentru protecţia împotriva radiaţiei şi prevenirea eliberării de material radioactiv din combustibil în mediu.

An estimate of spent fuel activity in storage ponds is important to any analysis of actual cladding failure due to corrosion. This allows the operator the possibility to correlate the estimated values of the activity to the actual values in water samples, thereby predicting the number of failing fuel elements. This paper presents the obtained results in estimation of activity of spent fuel activity, EK-10 and C-36, which is in wet storage at Magurele in Romania. Validated computer codes and history of fuel exploitation have been used to estimate the actual state fuel source term. Also the computation is important for estimation of radionuclides level (fission products, fission gasses, activation products) presents in spent fuel inside so for establishing of the necessary measures which could be taken for radiation protection and for prevention of radionuclides release into the environment.

Keywords: source term, radionuclides level, activity

1.Introduction

The integrity of fuel cladding is very important because the cladding represents the first barrier to retain the radioactive fission products. The first stage in the treatment of spent fuel is the so-called cooling phase. The spent fuel assemblies are transferred from the reactor to a tank in which the water serves to remove the radioactive decay heat and also provide radiation shielding. The

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presence of actinides and fission products in the storage pool would be a clear indication of fuel damage or significant degradation.

The physical state of the fission products, in conjunction with their chemical characteristics, determines the volume occupied by these species. If the volume of irradiated fuel is greater than that of the fresh fuel that was consumed, the resulting swelling can cause the fuel to exert a contact pressure on the cladding. The pressure inside the fuel element produced by the release of fission gasses contributes to the internal stress on the cladding.

The ratio of the volume of the plenum to that of the solid oxide inside a fuel element is chosen so that the pressure due to released fission gasses does not become large enough to rupture the cladding.

Detailed studies of the slow-neutron fission of U-235 have shown that the compound nucleus splits in more than 40 different ways, yielding over 80 primary fission products (or fission fragments) [1]. The purpose of this article is to present the estimation of the actual state of source term for the spent fuel (EK-10 and C-36) existing in spent fuel storage at Institute for Nuclear Physics "Horia Hulubei" (IFIN-HH) from Bucharest.

2. Review of existing computer tools for source term estimation

Several computer codes have been developed and validated for fuel burn-up estimation. Computer codes are necessary to determine the isotopic content of the fuel with acceptable precision. For example, the ORIGEN code [2] can determine the radioisotope inventory, for a given fuel composition, a power versus time history, and for a nominal neutron spectrum. ORIGEN will break down the photon (gamma) source by its origins: from actinides, from fission products, or from other structural or target materials. ORIGEN assumes a zero-dimensional model for a lump of fuel irradiated uniformly by a selected neutron spectrum, with the prescribed neutron flux level or power. The irradiation time history of the fuel lump is assumed to be represented by a histogram of constant-power (or flux) levels. The isotopic content for a given burn-up depends on the fuel history, but not strongly. Consequently it is relatively simple to bound the content for each scenario. ORIGEN will also provide information on the neutron source from the (gamma, n) reaction.

The MACCS2 code [3] can be used to estimate the movement, isotopic transmutation, and dissipation of a radioactive cloud formed by the release of a quantity of radioisotopes as a gaseous discharge. It provides estimates of radiation dose rates and integrated doses at various off-site distances from the source point, given certain assumptions about atmospheric conditions. Using the radioactive cloud content (radioisotope activity for emission of alpha, beta, gamma) provided

by MACCS2, hand calculations can also be used to estimate (and bound) the dose rates and committed doses from submersion, inhalation, and ingestion.

The MICROSHIELD code [4] can be used to estimate the gamma radiation dose rates to operations staff and research experiment staff on site. As a given scenario is analysed, certain geometrical arrangements will become clear that address the issue of direct and indirect irradiation to personnel in the reactor building. It is important to realize that a single calculation can be made by a code such as MICROSHIELD that includes all of the gamma radiation from a source containing many radioisotopes. (Refs. To point kernel numeric integration codes)

Generally, the (gamma, n) neutron source is small in any reactor fuel. Simple hand calculations can be used to estimate the dose rates from neutrons at key locations in the event that heavy actinides mixed with strong gamma sources are released from the core as solid lumps. Should the dose rates from neutrons become an issue, then fixed-source calculations will be necessary in order to determine the neutron flux and energy spectrum (and corresponding dose rate) at points of interest. For this type of calculation, radiation transport codes will be needed. Both multi-group (diffusion theory, Sn transport), and continuous-energy (Monte Carlo) methods can be used to determine the dose rate at specific locations. Radiation transport analysis by these methods is quite complex.

3. Input data: spent fuel type stored at IFIN-HH Romania

Fuel elements are often classified according to the structure of various types meaning the structure of elements composed of two or more material segments or regions. Thus one hears of aluminium clad, unbounded uranium elements, for example. The fissile phase is commonly called the “fuel meat.” The “cladding” or “canning” is the containment of material, or the sheath that prevents corrosion of the fuel by the coolant and also prevents contamination of the coolant by fission products from the fuel. The fuel meat may consist of a metallic phase (U metallic), alloy (U-Al alloy), ceramic (UO_2 dispersed in MgO), intermetallic compound, interstitial compound, or dispersed.

The spent fuel used in the VVR-S research reactor at the Magurele site consists of fuel element types EK-10 and C-36. The nominal power of this research reactor was 2 MW. Reactor operation was stopped at the end of 1997. The spent fuel is stored in wet conditions at the site in storage ponds. As discussed previously, the purpose of this article is to identify a mechanism for estimating the nuclide activity in storage ponds. This estimate can be used to determine the number of failed fuel elements, to study the conditions in which the spent fuel is stored, and to provide further advice to improve the storage conditions to keep the integrity of fuel cladding until the spent fuel is returned to the country of origin. The characteristics of the spent fuel [5] are presented in the Table1.

Table 1

Characteristics of EK-10 and C-36 fuel elements

Characteristics	EK-10	C-36
Assembly material	Al	$AlMg_3$
Assembly thickness	1-1.5mm	1-1.5mm
External diameter of fuel element	10 mm	10 mm
Inner diameter of fuel element	6 mm	6,9 mm
Cladding thickness	2 mm	1.55 mm
Cladding material	Al	Al
Active lengths	500 mm	500 mm
Fuel type	UO_2+MgO	U-Al alloy
Geometry of fuel element	Cylindrical	Cylindrical
Total high of fuel element	750 mm	750 mm
Enrichment in U-235	10%	36.63%
U-235 weight in one assembly	128g	143g
Burn-up degree	25-35%	25-45%

The reactor was operated at 2 MW maximum power from 1957 until it was shutdown in December 1997. The average burn up of the EK-10 fuel was 49%; average for the C-36 fuel was 54%. Further details of the operating regimes are given in Table 2 [5].

Table 2

The operating regime for EK-10 and C-36 fuel elements

	Operating regime		
	1	**2**	**3**
Start date of regime	Aug. 1957	May 1984	May 1993
End date of regime	May 1984	May 1993	Dec 1997
Max Power [MW]	2	2	2
Effective full power hours [h]	64920	36408	10512
No of fuel assemblies in equilibrium reactor core	36 – 51	36 – 51	36 – 51
Fuel type	EK-10	EK-10 and C-36	C-36
Average burnup [%]	35	35 – 45	45
No of assemblies discharged	104	EK-10 - 49 C-36 - 2	68

On discharge, the fuel was retained in the “at-reactor” (AR) storage pond for at least 1 year and then transferred to the “away from reactor” (AFR) pond, where it continues to be stored.

For both types of fuel elements, calculations were performed considering the minimum, median and maximum wet storage time as follows:

- EK-10: minimum – 14 years; median – 26 years; maximum – 40 years;
- C-36: minimum – 9 years; median – 12 years; maximum – 14 years.

4. Description of the computer tools and mathematic model

Starting with the initial composition of the nuclear fuel, the irradiation history (Table 2) and the specific neutron flux in VVR-S, $2x10^{13}$n/(cm^2 s), the ORIGEN-S computer code was used to solve a set of coupled differential equations which describe the generation and transformation of all radioisotopes and provide the final composition of the spent fuel.

To determine the radionuclide composition in time, the ORIGEN-S code provided the solution to the following unsteady state species balance equations:

$$\frac{dN_i}{dt} = \text{Formation rate} - \text{Destruction rate} - \text{Decay rate} \qquad (1),$$

The nuclide *i* (mass number *A*, atomic number *Z*) can be formed by fission, by neutron capture in the nuclide *j* (mass number *A-1*, atomic number *Z*), and by radioactive (usually beta) decay of the nuclide *k* (mass number *A*, atomic number *Z-1*). In neutron capture, the mass number is increased by unity, but the atomic number is unchanged; in beta decay the reverse is true. The species *i* consumption occurs by fission by capture and by radioactive disintegration. With these considerations the species differential balance equations become:

$$\frac{dN_i}{dt} = \sum_j \gamma_{ij}\sigma_{fj}N_j\Phi + \sigma_{cj-1}N_{i-1}\Phi + \lambda_{ji}N_j - \sigma_{fj}N_i\Phi - \sigma_{cj}N_i\Phi - \lambda_i N_i \qquad (2)$$

where i=1,2,...I and

$\sum_j \gamma_{ij}\sigma_{fj}N_j\Phi$ = formation rate of Ni due to all fission of N_j

$\sigma_{cj-1}N_{i-1}\Phi$ = transmutation rate in N_i due to radiative capture of N_{i-1}

$\lambda_{ji}N_j$ = formation rate of N_i due to radioactive disintegration of N_j

$\sigma_{fj}N_i\Phi$ = destruction rate due to fission of N_i;

$\sigma_{cj}N_i\Phi$ = destruction rate of N_i due to radioactive capture ((n,γ), (n,α), (n,p), (n,2n));

$\lambda_i N_i$ = decay rate of N_i

The computer code contains all data needed (nuclear reactions (i=1,2,..I), species fission and capture specific surface (σ_{fj}, σ_{cj}), time constants of radioactive disintegration reactions (λ_i, λ_{ji})) to simulate the fuel transformation inside of reactor and also in wet storage. In fuel wet storage the missing of a neutron flux (Φ) simplifies the above presented model.

5. Results

Fig. 1 presents the computed evolution of Activity (expressed in Curie units (Ci); 1 Ci = 3.7 10^{10} Bq ; 1Bequerel = 1 Bq = 1 desintegration/s) over time (years) for spent fuel type EK-10 during the wet storage period;

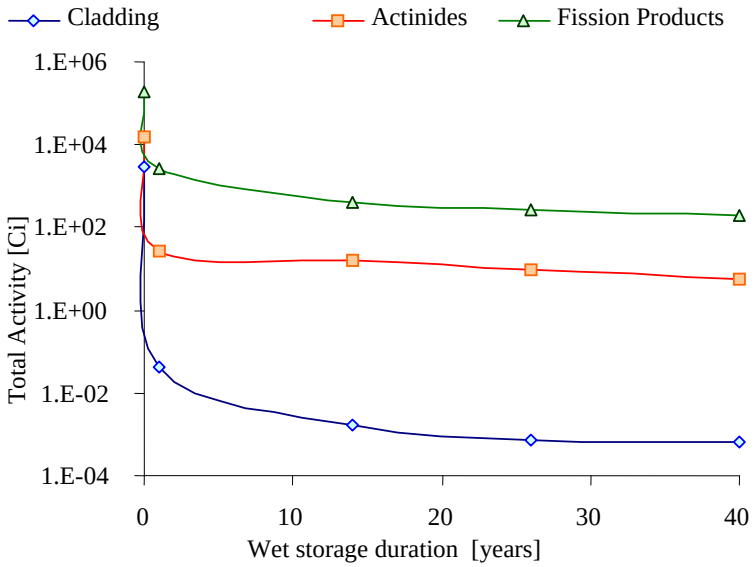

Fig.1 –Time evolution of activity for an assembly of spent fuel type EK-10 in wet storage .

The total activity of fission products in EK-10 spent fuel during different storage periods is presented in Table 3.

Table 3

The total activity (Ci units) of fission products in EK-10 spent fuel during different storage periods

Nuclide Inventory	At the removal from reactor	After 1 year of cooling	After 14 years of cooling	After 26 years of cooling	After 40 years of cooling
Cladding material	8.396E-02	4.300E-02	1.718E-03	7.197E-04	6.200E-04
Actinides	8.111E+03	2.737E+01	1.518E+01	9.260E+00	5.528E+00
Fission products	1.909E+04	2.615E+03	3.852E+02	2.804E+02	1.996E+02

The nuclides taken into account were: for structural material (cladding) C-14, Mn, Fe, Co; as actinide: Th, U, Pu, Am; and as fission products: H3, Kr, Sr, Y, Zr, Nb, Tc, Ru, Rh, Ag, Cd, Sn, Sb, Te, Cs, Ba, Ce, Pr, Pm, Sm, Eu.

Fig. 2 and Table 4 present the evolution of Activity over time (years) for C-36 fuel discharged from the reactor after 10 years of operation and then cooled down in wet storage for maximum 14 years.

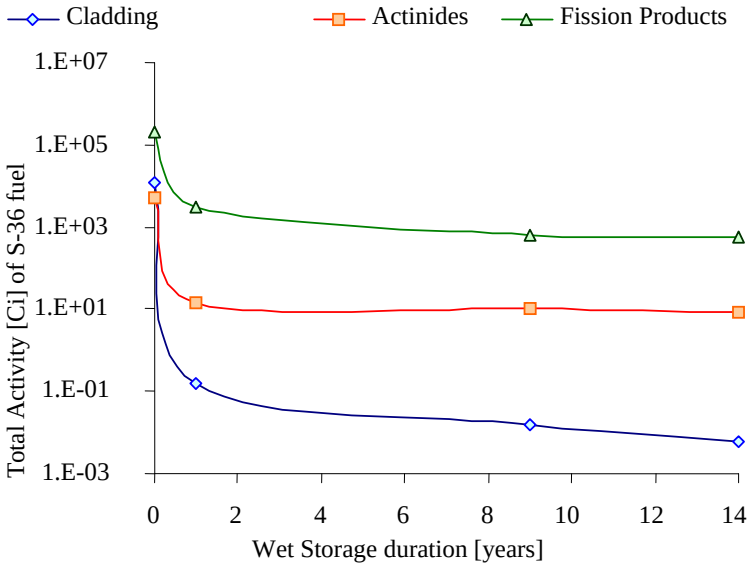

Fig. 2 Time evolution of activity for an assembly of spent fuel type C-36 in wet storage

Table 4

Total activity (Ci units) of C-36 spent fuel during different storage periods

Nuclide Inventory	At removal from the reactor	After 1 year of cooling	After 9 years of cooling	After 14 years of cooling
Cladding material	1.127E-01	5.778E-02	5.968E-03	2.383E-03
Actinides	2.412E+04	3.000E+03	6.552E+02	5.492E+02
Fission products	1.127E-01	5.778E-02	5.968E-03	2.383E-03

Further safety assessment of spent fuel storage conditions [6-8] could be done to estimate the possible release of radionuclides in the worst case accident scenarios.

The curves in Figs. 1 and 2 showed that the calculated activity of cladding, actinides and fission products are consistent with the expected concentration of the short lived nuclides for those three media. In this case the fission products have the highest concentration of the short-lived nuclides, thereby realizing the

greatest radiological decay in the shortest period of time. In contrast, the cladding has fairly consistent construction material, thereby demonstrating the least change due to short-lived nuclides in the same period of time and the most rapid achievement of a fairly flat decay state.

6. Conclusions

The three curves in both Figs. 1 and 2 are consistent with mathematical decay predictions (e.g., rapid short-lived nuclide decay followed by nearly flat medium and long-lived nuclide decay. Therefore, the absence of a significant increase in actinide and fission product levels in water samples would demonstrate that there is no significant increase in fuel degradation. Moreover an increase in the long-lived nuclide activity would clearly indicate an increase in the number of failed fuel elements. The rise in long-lived activity could be correlated to the number of failed fuel elements, which will assist in the study the conditions in which the spent fuel is stored and provide further advice to improve the storage conditions to keep the integrity of fuel cladding until the spent fuel is returned to the country of origin.

Therefore the ORIGEN-S code is one acceptable mechanism for estimating nuclide activity as a determinant of failed fuel. Validated computer codes have been used to estimate the existing radionuclides (fission products, fission gasses, activation products) present in spent fuel so that the necessary measures could be taken for radiation protection and for prevention of radionuclides released into the environment. The ORIGEN-S code was subsequently used to estimate the activity of EK-10 and C-36 spent fuel elements in storage pools at Magurele in Romania, and the authors of this article presented the results obtained as a practical example.

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