6th Conference on Nuclear and Particle Physics
17-21 Nov. 2007 Luxor, Egypt

RADIO NUCLIDES RELEASE IN RESEARCH REACTORS

El-Messiry A. M*., Khalil Y. , Abd Semea A.***

** National Center for Nuclear Safety and Radiation Control,*
P.O.Box 7551, Postal code 11762 Nasr city, Cairo , EGYPT
*** Nuclear Engineering Dept., Alexandria Univ., Egypt.*

Abstract

One of the major topic in nuclear safety is the quantitative evaluation of the radionuclides source term in nuclear reactors under routine and accidental conditions. The present study considers the release paths from fuel to coolant during normal and accidental situations of research reactors. Equivalent full power days approach, has been adopted for implementing reactor operating history in the calculations. ORIGEN II code , recoil and Knock out phenomena, experimental correlations, and mathematical models have been employed in determining source term in fuel, releases to fuel clad interface, release from clad to coolant, and concentration in coolant. Different volatile fission products have been manipulated as: Br-83, Kr-85, I-129, I-131, I-133, Xe-133, Xe-135, Cs-137, Te-127, Te-131m, Tc-99, Tc99m, Mo-99, Sr-90, Ru-106. Normal operation and accidental situation have been studied. The results have been verified against published data during normal operating conditions, it showed a good agreement.

Keywords: *Releases, Fission Products, Radionuclides, Source Term, Leakages*

INTRODUCTION

Going back to the main productive source of fission products in reactor core, i.e. the fuel element, the amount of fission products existing at a given time during normal operation can be accurately estimated from the operation history of the core, as will as, the accomponing releases. However, the fraction of fission products that will be released under a given set of circumestances (accidents) cannot be accurately estimated in an easy way. The release pathways go through the fuel matrix to fuel clad air interface, to primary coolant through clad crack, to secondary coolant through heat exchanger wall defects, to reactor containment, and finally to surounding environment. The source term is the radionuclides generated in the fuel matrix due to fission process (radionuclides inventories in the fuel), it composes of actinides, fission products gases, and other radionuclides. The concentration of these nuclides depend on different factors such as fuel materials, operating power level, operating history, and fuel burn up [1].

The main purpose of this study is to utilize models, methods, and experimental correlations for determining the possible fission products releases into reactor coolant of an

operating reactor. Upon determining the routine releases in coolant, one may assess the efficency of filtering systems, and estimates the normal doses to workers, besides examines if these limts fufilled the safety requirement imposed by regulatory body or not. The possible crack in fuel cladding material could be visualized. The possible releases from an virtual desing base accident have been determined. A case study research reactor has been choosen for the application. The study may serve for:

- Detecting/or assessing failure in clad material.
- Highlight the reactor core criticality variation with time.
- Calculating the radiation levels in reactor hall.
- Assess the design parameters of containment ventilation system during normal and emergency situations.

RESEARCH REACTOR CASE STUDY

The study considers a MTR research reactor with the following specifications

1- Swimming open pool,
2- Plate type fuel element,
3- Fuel material is U_3O_8 powder dispersed in an Al matrix,.
4- The cladding is Al.
5- The coolant is demineraliazed light water.
6- The core is shielded axially by 12 m of light demineraliazed water, and radial shielding of cylinder with inner diameter 4 m plus heavy concrete with thickness of 200 cm .

The main characteristics of the reactor are given Table (1). We assume the reactor operates and shutdown for refueling and maintenance according to the time schedule given in Table (2).

Table (1) Main characteristics of the reactor case study

Item	Value	Item	Value
Power density	0.759 MW/FE	Enrichment	18%
Core dimension	60x60x100	# of FE in the core	29
Core volume	0.36 m^3	Weight of U per FE	2244.44 gm
Pool height	12 m	Clad thickness	0.5 mm
Core full power	22 MW	Pool volume	151.09 m^3

Table (2) Assumed operation and shutdown period

Period (Days)	State	Average power (mw)	Equivalent full power operation	Accumulated full power	Accumulated operating days
809	operation	0.166	6.1	6.1	809
120	Shutdown	---	----	----	----
45	operation	9.72	19.88	25.98	854
250	Shutdown	---	----	----	----
283	operation	1.06	13.64	39.62	1137
710	Shutdown	---	-----	----	----
483	Operation	0.63	13.83	53.45	1620

COMPUTATIONAL PROCEDURES

I- Normal Operation

The computational procedure adopted for this study composed of the following steps:

- **First Step:** Computing fission products inventory in the fuel region (first barrier), this is done using ORIGEN2 code [2]
- **Second Step:** Computing fission products released to fuel clad interface, this is done utilizing results obtained from recoil and Knock out phenomena [3]
- **Third Step:** Computing fission products released/migrated/transported/leakage through clad material (second barrier) into primary coolant, this is done exploiting published data [4]
- **Forth Step:** Computing fission products concentrations in primary coolant (third barrier), this is done by solving the balance equations of the fission products concentration.

The above three steps (second, third, forth) are repeated for each of the following two situations, where the first step is common for the two situations:

- Normal Operation.
- Accidental Conditions.

Fig (1) illustrates the computational chart. The full power days approach is convenient, realistic, and conservative it is computed from the following definition:

$$\text{Full power days} = \sum P_i * D_i / P, \quad (1)$$

where P_i : The i^{th} Power level in MW, D_i : Period during the reactor has been operated at P_i in days, P : maximum reactor power MW.

ORIGEN II code is used for calculating the build up and decay of radio-nuclides materials in the fuel matrix, the required nuclear data bases are: decay constants, cross– section libraries, fission product yields, and photon data. It should be noted that the data library selected for the reactor is based on PWR U^{235} with enriched UO_2 fuel for burn-up 50,000MWd/Metric Ton, although the fuel type, configuration, and enrichment of the reactor are different from those based upon by the code library, the following justifications has been received from ORIGEN II developers :

- No libraries based on data for research reactors are available, since the major guide is the energy spectrum and not the reactor type.
- Energy spectrum that ORIGEN libraries are based upon (applicable to PWR –UO_2 fuel at 50,000MWD/TU) is roughly similar to the energy spectrum derived from MTR-U_3O_8 fuel at 106,000MWD/TU.

The following mathematical model [5] has been adopted to account for the concentration of radionuclides in the reactor coolant $C_w(t)$.

$$C_w(t) = \frac{R_{fw}}{V_w}[1-\exp(-\lambda t)] + C_{w0}\exp(-\lambda t) \quad (2)$$

where:

$C_w(t)$ = The activity concentrations in the water (Ci/ m^3)

R_{fw} = The release rates from the clad into the water

V_w = Volume of water above the core

λ = The radioactive decay constant (sec^{-1})

t = The operating time (sec)

C_{w0} = The initial concentration of radionuclides in the water at the start.

If the calculations are restricted to the period directly after initial loading $C_{w0} = 0$, so can be calculated equation (1) will take the form:

$$\frac{C_w(t)}{R_{fw}} = \frac{1}{V_w}[1 - e^{-\lambda t}] \tag{3}$$

Equation (3) could be utilized to validate experimental results for the coolant inventory of certain radionuclides. V_w is calculated to be 151.09 m^3. The isotopes Xe^{133}, Xe^{135} and Kr^{85} are selected for verifications. Table (3) lists the selected radioisotopes for computation.

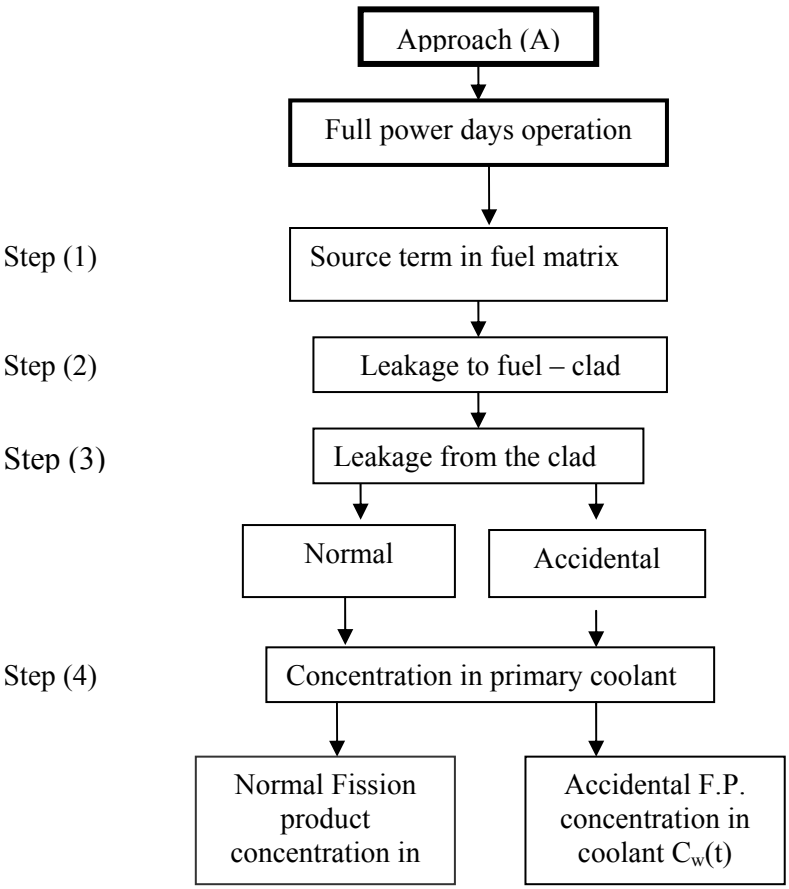


Fig (1) The computational chart

Table (3) Selected radioisotopes for computation

Radioisotope	Half life time ($T_{½}$)	Radioisotope	Half life time ($T_{½}$)
Kr-81 (Krypton)	2.1E5 y	Kr-81m (Krypton)	13 second
Kr-85m (Krypton)	4.48 h	Kr-83m (Krypton)	1.83 hours
Br-80m (Bromine)	4.4205 h	Kr-85(Krypton)	10.76 years
Br-82 (Bromine)	35.282 h	Br-83 (Bromine)	2.4 h
Sr-90 (Strontium)	28.1 years	Sr-89 (Strontium)	52 days
Mo-99 (Molybdenum)	65.94 h	Tc-99m (Technetium)	6.01 h
I-131 (Iodine)	8.02 d	Tc-99 (Technetium)	2.12E5 years
I-133 (Iodine)	20.8 h	I-129 (Iodine)	1.7E7 years
Te-127 (Tellurium)	9.35 h	Te-127m (Tellurium)	109 d
Te-127 (Tellurium)	9.35 h	Ru-103 (Ruthenium)	39.6 days
Xe-135 (Xenon)	9.2 h	Ru-106 (Ruthenium)	367 days
Xe-133 (Xenon)	5.245 days	Cs-137 (Cesium)	30.07 y
Te-131m (Tellurium)	30 h	Ba -133 (Barium)	7.2 years
Ba-140 (Barium)	12.8 days		

II- Accidental Situation

The fraction of activity released from the fuel into coolant will depend on the maximum temperature reached by the fuel under accident conditions. The accidental releases were based on the routine releases in fuel clad interface. The release fraction from clad to coolant R(t,T) depends on time and temperature of fuel during an accident. As the accident propagates, the fuel temperature increases and amount (%) of radioactive releases accordingly increases. For illustration the study considers the species: Cs, Te, I. The release factor R(t,T) is given by the following set of empirical equations [6]:

$$R(120,T) = A\exp(BT) \quad (4)$$

$$R(t,T) = R(120,T) * t/120 \quad \text{(For t<120 seconds)} \quad (5)$$

$$R(t,T) = R(120,T) + R(120,T) * [K'(T) - 1.0] * (t - 120)/3480 \quad \text{(for t>120 seconds)} \quad (6)$$

Where the assigned values for the constant A and B of the chosen species are extracted from experimental results( ) and listed in Table (4)

Table (4) Coefficients A and B for Cs, Te and I

species	A	B
Cs	0.0481	$3.45*10^{-3}$
Te	$5.12*10^{-5}$	$1.06*10^{-2}$
I	$1.26*10^{-2}$	$7.92*10^{-3}$ At T > 873 ^{0}K

R(t,T) is the amount (%) released into coolant, the exponential approximation of the percentage releases for the selected species I, Cs under different surface temperature are given in the following equations:

For I: $R(120,T) = 1.26*10^{-2}*\exp(7.92*10^{-3}T)$ (7)

For Cs: $R(120,T) = 0.0481 *\exp(3.45*10^{-3}T)$ (8)

RESULTS AND DISCUSSIONS

Table (5) shows the radionuclides that reaching the saturation levels. Where Table (6) lists the calculated values of the source term per FE (at the end of each operation days) in the fuel matrix for some selected isotopes. Figures from (2) to (11) illustrate the variation of the activities per fuel element of some of the selected radio-nuclides at different operation days.

Table (5) Radioisotopes reaching the saturation levels

Radio-nuclide	Saturation time (days)	Reference figure
I-131	49	Fig .(3)
Te-127	35	Fig .(4)
I-133	12	Fig .(5)
Te-131m	12	Fig .(6)
Kr-85m	8	----
Br-83	7	Fig .(7)
Xe-135	10	Fig .(10)

Table (6) Activity per FE (Ci) in the fuel matrix at the end of operation times

Nuclide	**Activity (Ci) for Different Operation times**			
	809 day	**854 day**	**1137 day**	**1620 day**
Fission products				
Nobel gases Xe 135	7.225E+03	6.781E+03	6.775E+03	6.550E+03
Xe^{133}	1.931E+04	4.073E+04	4.212E+04	4.231E+04
Kr 87	1.626E+04	1.606E+04	1.606E+04	1.593E+04
Kr^{85}	1.743	7.786	1.191E+01	1.604E+01
Halogens I 131	1.824E+04	1.779E+04	1.638E+04	7.147E+03
I133	4.179E+04	4.232E+04	4.235E+04	4.227E+04
Alkaline metal Tc99m	2.533E+04	3.429E+04	3.363E+04	3.361E+04
Mo99	2.968E+04	3.791E+04	3.802E+04	3.795E+04
Activation products				
Nobel gases Ar41	8.057E-18	1.666E-16	4.003E-16	7.702E-16
Alkaline metal Na24	2.905E+02	3.047E+02	3.063E+02	3.174E+02
Mg27	1.347E+03	1.437E+03	1.444E+03	1.496E+03
Mn56	3.426E-04	1.639E-03	2.552E-03	3.610E-03
Actinides				
Np239	4.084E+04	5.169E+04	5.234E+04	5.416E+04

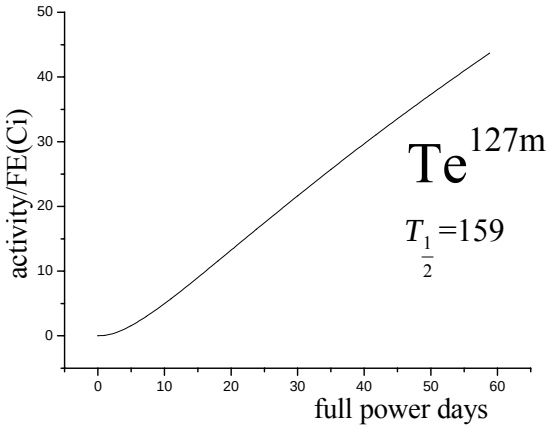


Fig.(2) Activity of Te^{127m} per FE

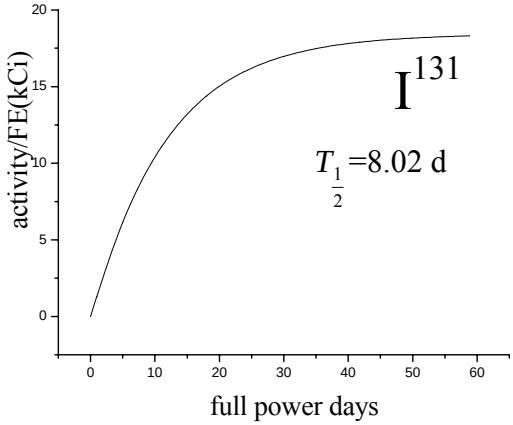


Fig.(3) Activity of I^{131} per FE

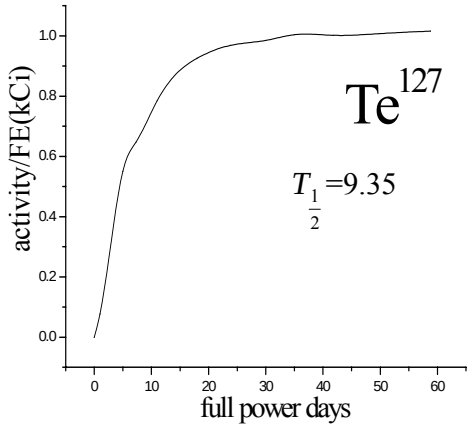


Fig.(4) Activity of Te^{127} per FE

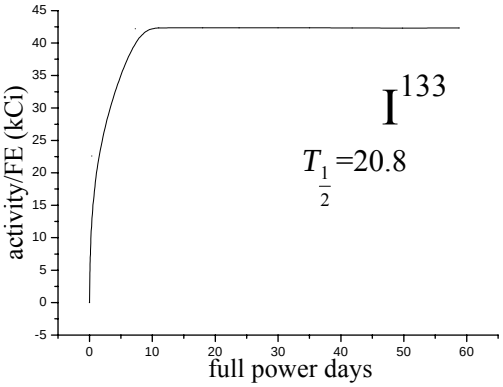


Fig.(5) Activity of I^{133} per FE

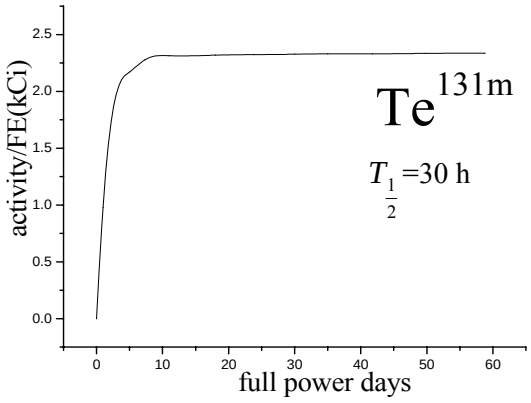


Fig.(6) Activity of Te^{131m} per FE

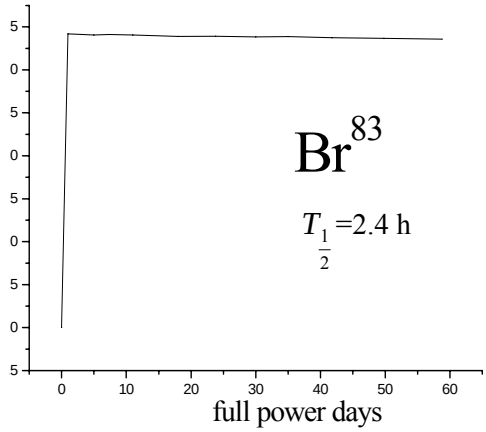


Fig.(7) Activity of Br^{83} per FE

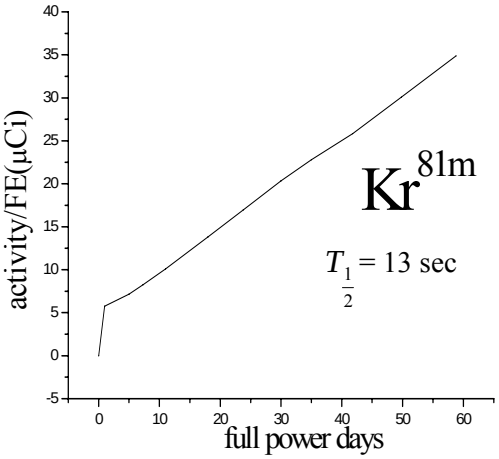


Fig.(8) Activity of Kr^{81m} per FE

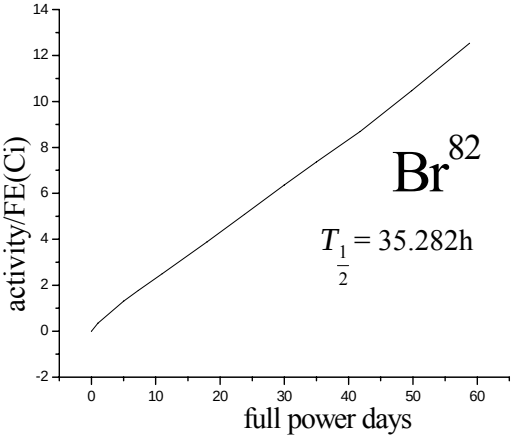
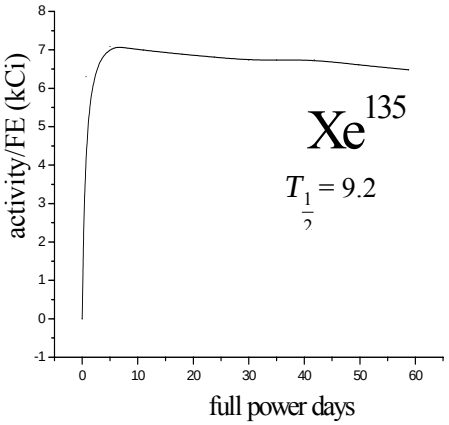


Fig.(9) Activity of Br^{82} per FE



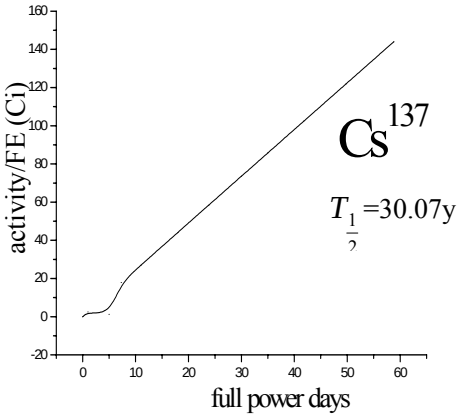


Fig. (10) Activity of Xe^{135}per FE

Fig.(11) Activity of Cs^{137}per FE

Table (7) gives the release fractions from the fuel matrix to fuel clad interface of the selected isotopes Kr^{85} , Xe^{133} and Xe^{135} due to recoil and knockout collisions[3]. The activities of the selected radionuclides in the core (all fuel) as a function of operation days are calculated and given in Table (8), while the fission products released in fuel – clad interface in the core is computed and listed in Table (9). The percentage leakage of fission product (high volatile) contained in fuel –clad interface due to minor cracks of the cladding material under normal operation is taken[4] of value 10^{-6}. The release rate into coolant from fuel – clad interface is calculated and listed in Table (10). The activity concentration of the selected isotopes in reactor coolant is calculated and tabulated in Table (11).

Table (7) Release fraction % for some selected nuclides

nuclide	Release fraction %		
	Recoil	knockout	Total
Kr^{85}	0.876	0.0013	0.8773
Xe^{133}	0.599	0.0021	0.6011
Xe^{135}	0.66	0.0016	0.6616

Table (8) Activities of the core fuel for selected radionuclides

Oper. time Isotope	**809 day**	**854 day**	**1137 day**	**1620 day**
Kr^{85}	5.051E+01	2.257E+02	3.454E+02	4.649E+02
Xe^{133}	5.96E+05	1.081E+06	1.221E+06	1.226E+06
Xe^{135}	2.095E+05	1.966E+05	1.965E+05	1.899E+05

Table (9) Fission products release amount (Ci) in fuel-clad interface (core)

Oper. time Isotope	**809 day**	**854 day**	**1137 day**	**1620 day**
Kr^{85}	44.312	198.006	303.019	407.857
Xe^{133}	33.64E+4	70.99E+4	73.39E+4	73.69E+4
Xe^{135}	13.86E+4	13.007E+4	13.00E+4	12.56E+4

Table (10) Release amount (Ci) from clad into coolant at normal operation R_{fw}

Oper. time Isotope	**809 day**	**854 day**	**1137 day**	**1620 day**
Kr^{85}	4.4312E-5	1.98 E-4	3.03 E-4	4.07857 E-4
Xe^{133}	0.3364	0.7099	0.7339	0.7369
Xe^{135}	0.1386	0.13007	0.13	0.1256

Table (11) Activity concentration in reactor coolant (Ci/m^3)

Oper. time Isotope	**809 day**	**854 day**	**1137 day**	**1620 day**
Kr^{85}	2.8445E-11	5.443E-9	1.8^{E}-8	4.08E-8
Xe^{133}	1.033^{E}-4	2.735E-3	2.919E-3	2.9317E-3
Xe^{135}	6.069^{E}-4	5.6956E-4	5.6925E-4	5.4998E-4

Accidental Situation

i- Clad release fraction R(t,T)

The design base accidents (DSBs) are considered in this situations. Quantification of accident severity is assumed to be on scale from 1 to 6 and is proportional to fuel temperature. The obtained results for determining release fraction R(t,T) for the selected isotopes Cs, I are given in Tables(12) and (13). These results are sketched in Figures (12) and (13) for the time 30 and 1800 second following the beginning of the accident.

Table (12) Accident severity, Percentage Release R(t,T) for I at different times

Temperature (^{0}C)	**Severity level**	**Time(sec)**				
		30	**60**	**600**	**1800**	**3600**
600	1	4.71	9.42	21.179	27.026	35.796
650	2	6.999	13.979	30.314	36.106	44.795
700	3	10.4	20.8	43.32	47.62	54.08
750	4	15.45	30.91	61.81	61.81	61.81
800	5	22.96	45.92	91.84	91.84	91.84
850	6	4.71	9.42	21.179	27.026	35.796

Table (13) Accident severity, Percentage Release R(t,T) for Cs at different times

Temperature (^{0}C)	**Severity level**	**Time(sec)**				
		30	**60**	**600**	**1800**	**3600**
600	1	0.244	0.489	0.907	0.737	0.408
650	2	0.290	0.581	1.1195	1.014	0.856
700	3	0.345	0.69	1.37	1.344	1.304
750	4	0.41	0.82	1.681	1.823	2.018
800	5	0.487	0.975	2.077	2.398	2.88
850	6	0.578	1.155	2.548	3.121	3.994

ii-Accidental fission products concentration in coolant $C_w(t)$

Accidental concentration $C_w(t)$ of fission products I and Cs in reactor coolant at different temperatures and times are calculated and given in Table (15).

Table (14) Concentration (Ci/mm) of fission products I and Cs in reactor coolant at different times

Temperature (^{0}C)	Severity level	I		Cs	
		30 (sec)	60 (sec)	30 (sec)	60 (sec)
600	1	----------	----------	0.43554E+5	0.7283E+5
650	2	11.718E+5	89.06E+5	0.51765E+5	1.528E+5
700	3	17.414E+5	111.45E+5	0.6158E+5	2.328E+5
750	4	25.875E+5	134.55E+5	0.732E+5	3.602E+5
800	5	38.4396E+5	153.783E+5	0.8693E+5	5.1408E+5
850	6	57.124E+5	228.497E+5	1.0317E+5	7.1293E+5

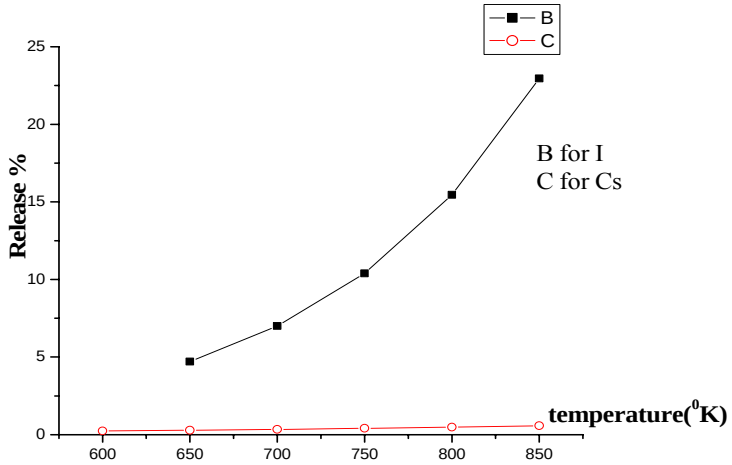


Fig. (12) Variation of release (%) of Iodine and Cesium with temperature at t=30 sec

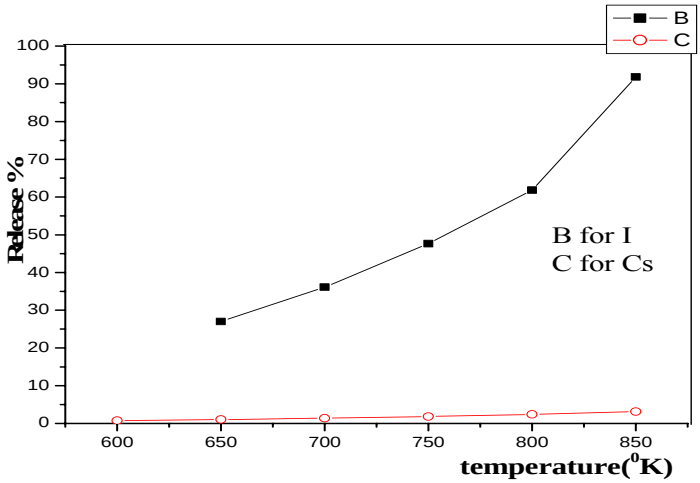


Fig. (13) Variation of release (%) of Iodine with temperature at t=1800 sec

CONCLUSIONS

1. The fission products activities in fuel matrix (source term) reaches the saturation levels after a considerable operating times (days) of many radionuclides as: I-131, Te-127, I-133, Te-131m, Kr-85m, Br-83, Xe-135. While the others are increasing with times as: Te-127m, Kr-81m, Br-82, Cs-137. These are due to the variation of the fission products nuclear parameters as fission yield, cross section, and decay constant.

2. The releases amount of different isotopes during an hypothetical accident varied in values depending on the type of the isotope, fuel temperature, and the released period. As an example Cs isotopes released with a percentage amount lower than that for I .

3. Very trace element of activation products as Ar^{41} , Na^{24} , and Mg^{27} could be found during power operation in the coolant.

4. Small amount of noble gases Xe^{135}, Xe^{133} ,and Kr^{85} could be found in reactor coolant during normal operation. The amount of Kr^{85} (long lived) is very small Than the short lived Xe^{135},and Xe^{133} .

REFERENCES

[1] NRC DOSE 2.3.2 ,"Evaluation of Routine Radioactive Effluents from Nuclear Power Plants "CCC-0684 NRC DOSE 2.3.2,NRC , April 2002.

[2] Croff ,"ORIGEN2 : A versatile Computer Code for Calculating the Nuclide Compositions and Characteristics of Nuclear Materials ", Nuclear Technology, Vol.62, Sep.1982,

[3] Abdel Nabi A.,"Safety calculations of Nuclear Reactors ", M.Sc. dissertation , Faculty of Engineering ,Alexandria University, 1998 .

[4] IAEA"Radioactive inventories, activity retention and releases", IAEA, Tec Doc 400, pp. 47, (1986).

[5] B.J. Lewis, et.al," Fission product release from Uranium-Aluminum alloy fuel in slowpokE-2 reactors" , nuclear technology, vol.105, Mar.1994, page 366-380.

[6] Taleyarkhan, R.P., "Analysis and Modeling of Fission Product Release from Various Uranium-Aluminum Plate-Type Reactor Fuels, Nuclear Safety," vol. 33. No.1, January-March 1992.