CALCULATION OF FISSION PRODUCTS TRANSPORT IN A POOL TYPE REACTOR USING CATHARE2

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Obiectivul lucrării este de a dezvolta un model pentru transportul produşilor de fisiune în circuitul primar şi în anvelopa reactorului de tip piscină, reactorul TRIGA SSR 14 M utilizând codul CATHARE. Modelul, creat pentru reactorul TRIGA intenţionează să calculeze evoluţia a o serie de componente radio-chimice, având caracteristici definite în CATHARE2 (Kripton-87, Xenon-133, Iod-131 si Cesiu-137). Modelul conţine atât circuitul primar cât şi clădirea reactorului, implicând zone de apă şi gaze necondensabile. Produşii de fisiune sunt introduşi în piscina reactorului utilizând un operator CATHARE2 de tip SOURCE, concentraţiile acestora fiind calculate cu sistemul de coduri SCALE. Utilizând module integrate din SCALE incluzând ORIGEN-S, se obţine inventarul produşilor de fisiune în caseta TRIGA cu 25 pini de combustibil.

Acest tip de calcule sunt suport în studiile de nivel 2 PSA pentru reactorul TRIGA SSR, pentru a determina împraştierea speciilor radioactive atât în circuitul primar cât şi în anvelopă.

The objective of the paper is to develop a model for fission products transport in the primary circuit and in the containment of a pool type reactor, TRIGA SSR 14 MW reactor, using CATHARE code. The model, created for the TRIGA reactor, aims to calculate the evolution of a series of radio-chemical components, the characteristics of which are predefined in CATHARE (KRYPTON-87, XENON-133, IODE-131 and CESIUM-137). It contains both the primary circuit and the reactor building, involving water zones and non-condensable gases. The fission products are introduced in the pool using a SOURCE operator, the concentrations being calculated with SCALE code system. Using SCALE integrated modules including ORIGEN-S the fission products inventory are obtained in a TRIGA bundle with 25 fuel pins.

This kind of calculations is intended as support studies for Level 2 PSA for TRIGA SSR, in order to determine the spreading of radioactive species both in the primary circuit and in the containment.

Keywords: Fission products, CATHARE, TRIGA

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1. Introduction

Romania TRIGA reactor was commissioned in 1980 (first criticality was reached on November 17th 1979). There are two independent cores sharing the same pool: a high-flux 14 MW Steady State Reactor (SSR), research and materials testing reactor and an independent (from operation point of view) Annular Core Pulsing Reactor (ACPR).

The SSR is a forced convection reactor cooled via a primary circuit with 4 pumps (2 pumps in operation at 14 MW) and 3 heat exchangers (2 exchangers in operation at 14 MW). The power is removed by a secondary circuit with forced convection towers. The reactor was used for CANDU fuel testing, structural material (steel, zircaloy) testing and isotopes production. Characteristics of TRIGA SSR used in the calculation of fission product transport are given in the Table 1.

Table 1

Characteristics of TRIGA SSR

Name	Characteristics
Reactor tank	310 m^3
Delay tank	110 m^3
Pump Head $Head_{tot}$ Speed NPSH	906 m^3/h (0.252 m^3 /s) 35mCA 42mCA 1475 rot/min 5.4 m
Total volume of water (including pool)	687 m^3
Heat exchanger Water volume primary side secondary side Number of pipes (primary) Surface	 6.1 m^3 9.1 m^3 1262, diameter 20x2 mm 890 m^2
Volume of containment	18500 m^3
Ventilation system air flow (emergency mode)	13600 m^3/h

The CATHARE code (Code for Analysis of Thermal-Hydraulics during an Accident of Reactor and Safety Evaluation) [1] is developed to perform best-estimate calculations of pressurized water reactor accidents: PWR loss of coolant (large or small break, primary and secondary circuit), reactivity insertion, steam generator tube rupture, etc. It is developed in GRENOBLE by the French Atomic Energy Commission (CEA) and it is owned by four partners: CEA, EDF, FRAMATOME-ANP and IRSN.

2. CATHARE range of application

CATHARE includes several independent modules that take into account any two-phase flow behavior:

- Mechanical non-equilibrium:
  - vertical: co- or counter-current flow, flooding counter-current flow limitation (CCFL), etc.
  - horizontal: stratified flow, critical or not critical flow co- or counter-current flow, etc.
- Thermal non-equilibrium: critical flow, cold water injection, super-heated steam, reflooding, etc.
- All flow regimes and all heat transfer regimes.

In order to take into account these phenomena the CATHARE code is based on a two-fluid and six equation model with a unique set of constitutive laws. Various modules offer space discretization adapted to volumes (0D), pipes (1D) or vessels (3D) ready to assemble for any reactor description. CATHARE is limited to transients during which no severe damage occurs to fuel rods; more precisely, fuel ballooning and clad rupture are assumed to have no major effect on water flow in the primary circuit.

3. CATHARE fission products treatment

Acquisition of radio-chemical component characteristics must be specified for each circuit using the RADCHEMI key word and then associated with the circuit (CIRCUIT).

There are 13 radio-chemical components, the characteristics of which are predefined in CATHARE, and it is also possible to enter new user-defined components. For these new components, the user has to provide all characteristic data: chemical or radioactive type, chemical constants for the two-phase treatment, emergency shutdown behavior. The type of component is defined by the “ira” flag (ira = 2: gaseous component; ira = 1: non-gaseous component). The following properties of predefined radioactive components are given in the table below: “vie” (half-life), “ka” (weight of one GBq), “henry” (HENRY constant of the gas) and “effi” (efficiency of the Chemical and Volume Control filters). Those four constants have to be specified for every user-defined radio-chemical component.

Table 2

Constants used by the radio-chemical components existent in CATHARE2

	Constant	ira	half-life	ka (kg/GBq)	henry (in MPa)	effi
	corresponding CCV	IRA_NB	HFLIFE	KA_CST	HENRY_	RCVEFF
Component	**key-word**					
Krypton 87 (FP)	KRYPTO87	2	78 minutes	9.5 D-13	1.55 D4	1
Xenon 133 (FP)	XENON133	2	5.3 days	1.5 D-10	1.04 D4	1
Iodine 131 (FP)	IODE131	1	8 days	2.2 D-10	1	100
Cesium 137 (FP)	CESIU137	1	30 years	3.1 D-7	1	10
Nitrogen 16 (activation product of water)	AZOTE16	2	7.35 seconds	2.82 D-16	1.55 D4	1. D10

Table 3

"Ke" (entrainment coefficient through vaporization), "kc" (entrainment coefficient through condensation), "dilu" (gas into liquid dissolution time constant) and "dega" (gas stripping time constant) used in CATHARE2

Constant	ke	kc	dilu	dega
Corresponding CCV	EVAPOR	CONDEN	DISSOL	DEGASA
Component				
Krypton 88	0.1	0.01	5000. D3	5000. D3
Xenon 133	0.1	0.01	5000. D3	5000. D3
Iodine 131	0.1	0.01	1	1
Cesium 138	0.1	0.01	1	1

4. CATHARE2 modelling

The nodalization of the problem is depicted in Fig. 1. The reactor hall and reactor pool were modelled using *Volume* type module from CATHARE2, while the delay tank and pipes are modeled with *Axial* components. The reactor hall has two boundary conditions for inlet and outlet of air, simulating the air circulation done by the ventilation system (13600 m^3/h in emergency mode).

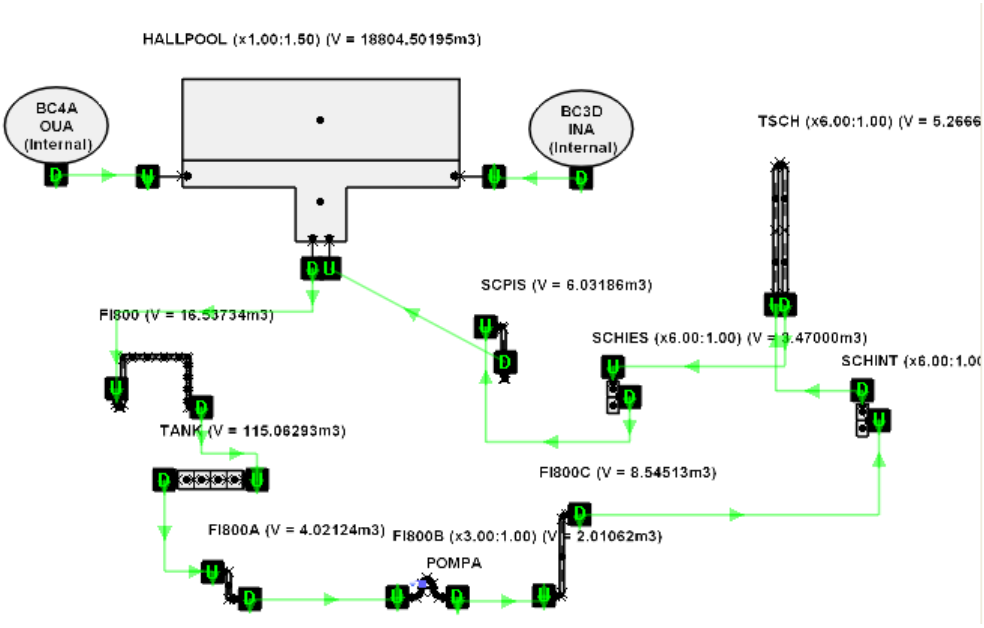

Fig. 1. Nodalization of TRIGA model for fission products transport (GUITHARE v1.5.1 [2])

The model represents the primary circuit components in a simplified manner. The water volume is preserved but some components have been collapsed: two pumps at nominal power are represented by a single pump with mass flow rate of 500 l/s (given by the combined action of the two actual primary pumps), the two heat exchangers are represented by only one composed of inlet volume (weight=2), individual thin tubes (weight=2x1262), outlet volume (weight=2). Since reactor core is not modeled, no heat transfer was considered and consequently there was no need for secondary system.

The purpose of the model is to calculate the fission products transport in the primary system and containment, and the evolution of the activity in different zones. The core damage is simulated by means of a radio-chemical components source (SOURCE operator) at the axial level of the core inside the volume representing the pool.

CATHARE2 is an evolved instrument capable of calculating the source of radio-elements coming from clad rupture of the defined fuel. Unfortunately, the model performed by CATHARE is dedicated exclusively to PWR (UO2 vertical fuel, inventory predefined in terms of activity for components at different times from emergency shutdown) and cannot be applied to TRIGA fuel. Thus, the source for the four radio-elements included in CATHARE2 (Kr-87, Xe-133, I-131

and Cs-137) had to be calculated by other means and included in the defined flow of the SOURCE operator as an activity concentration per kg of gas.

5. Calculation of the source of radio-elements

Inventory in TRIGA core with SCALE 4.4

An average TRIGA LEU bundle was modelled using SAS2H module from SCALE 4.4 [3]. One of the main utilizations of this module is to generate radiation and heat sources for depleted fuel. For each time dependent burnup composition, SAS2H does 1-D neutronic transport calculation using XSDRNPM for the assembly using a procedure with two distinct cell models. The first calculates the elementary cell of the fuel surrounded by water, obtaining the neutronic cross sections averaged on the cell neutron spectrum. The second model represents a larger cell from an infinite grid, model that can represent the TRIGA bundle. A schema of the SAS2H input model created for determining the radioactive inventory in TRIGA bundle is given in Fig. 2.

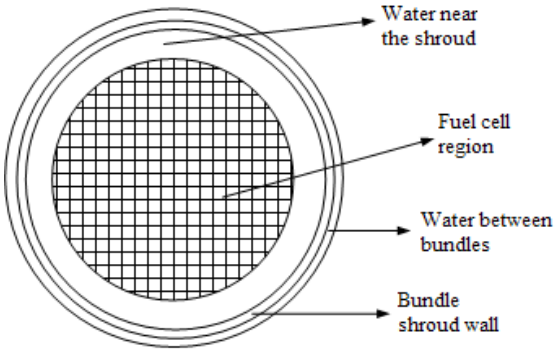

Fig. 2. Regions of the TRIGA bundle model for SAS2H

Neutronic spectrum inside fuel from the extended cell model is used to determine the cross sections for nuclides in the specified burnup composition. Neutronic cross sections resulted from transport calculations at each burnup time step are used in point calculations with ORIGEN-S which produces the burnup dependent composition for the next transport calculation. This sequence is repeated for the entire burnup history given by the user in the input deck.

We have introduced a simple and conservative burnup history of the reactor: 14500 MWD released without interruption (no ‘cooling’) by a 29 fuel bundle core (500 MWD per fuel bundle). The results of the SAS2H calculation are presented in Table 4, which gives the activity and mass of the four volatile

fission products that are included with CATHARE2. The last column in Table 4 gives the estimated releases, calculated as described as follows.

Table 4

Inventory and activity release for the CATHARE2 included fission products. Release fraction 0.057 corresponding to 940 °C fuel temperature

Fission product	Activity / bundle (Ci)	Mass / bundle (g)	Releases / core (29 bundles) (Ci)
I-131	1.21E+04	9.75E-02	2.00E+04
Kr-87	9.14E+03	3.22E-04	1.51E+04
Xe-133	2.65E+04	1.41E-01	4.38E+04
Cs-137	1.64E+03	1.89E+01	2.71E+03

Release of fission products from the TRIGA core

Work described in Ref. [4], produced by General Atomic Company, gives the correlation used to calculate the release of fission products from TRIGA fuel, both gaseous and volatile metals:

$$\psi = 1.5 \cdot 10^{-5} + 3.6 \cdot 10^{3} \cdot e^{-1.34 \cdot 10^{4} / T} \tag{1}$$

where T is the fuel temperature (K), stating that, though originally developed for HEU fuel, it is applicable to LEU as well.

The final Safety Analysis Report [5] points to the 1300-1400 °C fuel temperature domain for cladding rupture, as long as the cladding is kept below 300 °C and 940 °C when cladding temperature may be at the same value as the fuel, based on calculations of the stress on clad incolloy material due to temperature dependent dehydrading from $ZrH_{1.6}$. The most severe increase of the internal pressure of the fuel element are given by a step increase in temperature, that could be produced only by a strong reactivity insertion. As our previous analyses show that cladding will not be damaged for a credible reactivity insertion worth (2.4$ positive reactivity would be needed [6]), the reactivity insertion event was eliminated from the list of Initiating Events (IEs) that could lead to core damage. For a Loss of Flow Accident (LOFA) or a Loss of Coolant Accident (LOCA), where the fuel temperature evolution will not be so steep , therefore there is enough time for heat transfer inside the fuel element, and the 940 °C limit for cladding rupture is applicable.

Due to the mathematical form of the temperature dependence in (1), the release fraction will be higher at higher temperature, thus using 940 °C for fission products release will not be conservative if cladding rupture would actually be

produced at a higher temperature. But the operating history considered in accumulating the inventory is very conservative, because the TRIGA reactor was not operated continously and the average bundle energy released could not possibly reach 500 MWD because such a core would be subcritical. For 940 °C, using (1) we get a release fraction value of $5.7 \cdot 10^{-2}$ which is the release fraction entering the results in Table 4. Using results from [7], we estimated roughly the time for the release process as being 100 s. This was considered to be the time interval between hot pin failure and low power density pins failure in a LOFA without scram accident, taking into account that release from a broken clad is almost instantaneously for gases and volatile nuclides.

6. Calculation of the radio-elements transport

Using the CATHARE2 model described above, we investigated the capabilities of the code to calculate (first, with implicit data for the four radio-nuclides, Kr-87, Xe-133, I-131, Cs-137, existent in CATHARE) the activities of each fission product in different zones of the TRIGA facility, pool, primary lines, delay tank and containment (reactor hall).

In [5], the assumption is made that fission products apearing from damaged fuel will be found in the reactor hall and no transport calculation is done. Our purpose was to extend this analysis and to use CATHARE2 modelling for TRIGA reactor.

Using released activities given in Table 4, we launched two calculations with the CATHARE model described above and with implicit (hardwired in the code) radio-elements properties (see Table 2 and Table 3). The base case is with residual pump flow (50 l/s) in the primary circuit, and the other with nominal flow (504 l/s). The first is a more realistic assumption because one would expect fission products to be released when flow is lost (LOFA, LOPS), while the nominal flow rate was run to verify the effect of the possible entrainment of radio-nuclides in the primary circuit components. With TRIGA reactor, forced flow is downward, the largest water volume after the reactor pool (310 m^3) being the delay tank (115 m^3). In all cases that will be presented, the fission products source inside the reactor pool, placed 1.5 m above the pool floor, is activated at 10000 s of transient because of the time needed for the upper part of the volume representing the pool-containment to be filled with noncondensable (N2) by means of the inlet boundary condition. The lower part of this volume component is the pool, 10 m depth. The total time of the transient is 20000 s, consequently the fission products activity is tracked over a time interval of 10000 s. We mention that runtime for the time step used (maximum 10 s) is not prohibitive (a few minutes) because there is no fuel modelled and no heat transfer. Thus, the total duration of the transient could be increased if necessary.

Results with CATHARE radio-elements implicit properties

Fig. 3 shows the evolution of the total Kr-87 activity (in GBq) for residual flow in different zones of the TRIGA facility: containment-pool (a common volume, the upper, gas part, representing the containment), primary line at pool outlet (axial element „FI800" in Fig. 1) , delay tank (volume „TANK" in Fig. 1), primary line at pool return (axial element „SCPIS" in Fig. 1). Fig. 4 shows the same variables for the case with nominal flow rate. Fig. 5 and Fig. 6 gives the same evolutions, this time for Xe-133. Fig. 7 and Fig. 8 is for I-131, while Fig. 9 and Fig. 10 is for Cs-137.

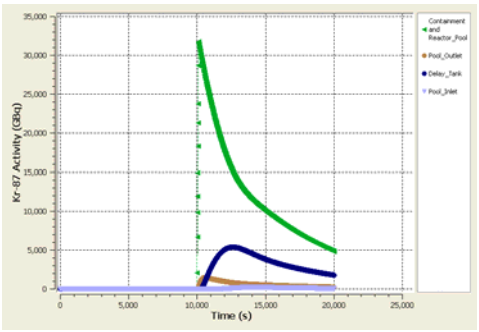

Fig. 3. Evolution of total Kr-87 activity in the primary circuit components (flow 50 l/s)

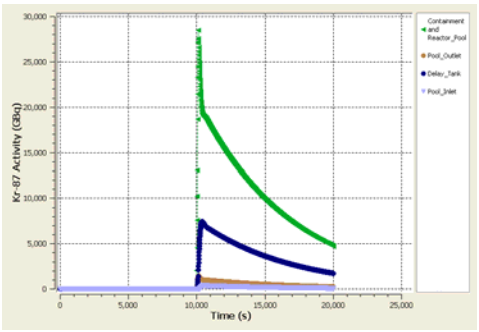

Fig. 4. Evolution of total Kr-87 activity in the primary circuit components (flow 504 l/s)

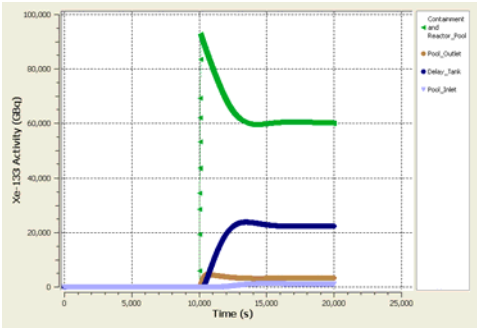

Fig.5. Evolution of total Xe-133activity in the primary circuit components (flow 50 l/s)

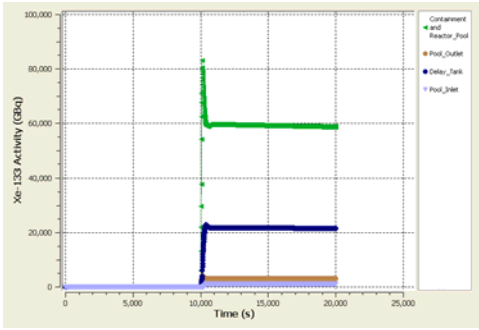

Fig.6. Evolution of total Xe-133 activity in the primary circuit components (flow 504 l/s)

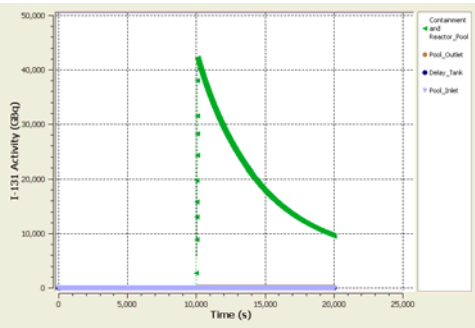

Fig. 7. Evolution of total I-131activity in the primary circuit components (flow 50 l/s)

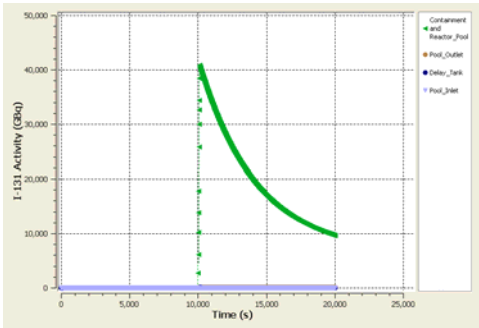

Fig. 8. Evolution of total I-131 activity in the primary circuit components (flow 504 l/s)

The total activity and its decrease trend are in each case dependent of the release activity and of the particular half time of the fission product. But there are specific properties of the CATHARE implicit radio-nuclides that make the behavior of Kr-87 and Xe-133 different from I-131 and Cs-137.

One can see in Fig. 3 through Fig. 6, that in case of the two noble gases studied there is a sizable activity in the delay tank and even in the primary lines, while this is not seen for I-131 and Cs-133.

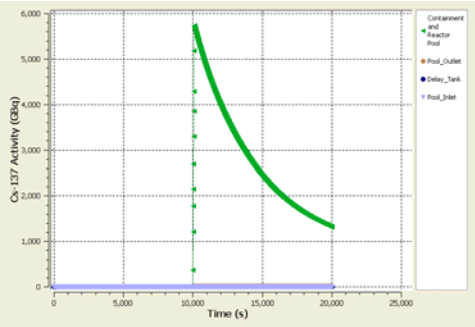

Fig. 9. Evolution of total Cs-137activity in the primary circuit components (flow 50 l/s)

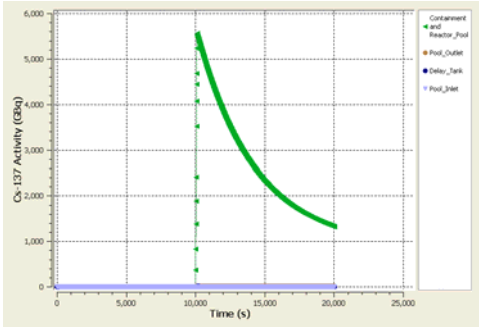

Fig.10. Evolution of total Cs-137 activity in the primary circuit components (flow 504 l/s)

For noble gases, the effect of the primary circuit flowrate becomes apparent, the decrease of the activity after the fission products source opening is steeper for large flowrate, suggesting that an important amount of these two species is entrained by the water inside the primary circuit components. Indeed, Fig.11 through Fig.14 shows that concentration of activity for Kr-87 and Xe-133 in water is much larger than in air, as opposed to what is expected since the source

inside the pool was entered as gaseous and the noble gases are known as noncondensable gases.

The flag (IRA=1) in CATHARE properties for I-131 and Cs-137 is not for gases, still they appear as highly volatile, Fig. 15 through Fig. 18 demonstrating that basically all their radioactivity is generated by the gas phase, being independent of the primary water flowrate, but subject to the ventilation gas entrainment from the reactor hall (or containment).

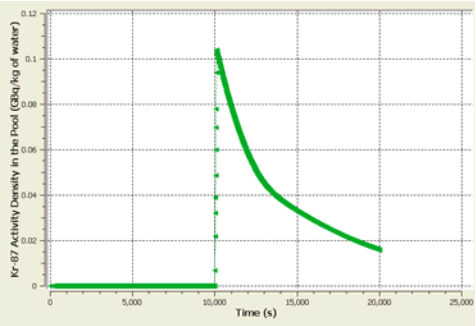

Fig. 11. Evolution of total Kr-87activity density in the pool (flow, per kg of water)

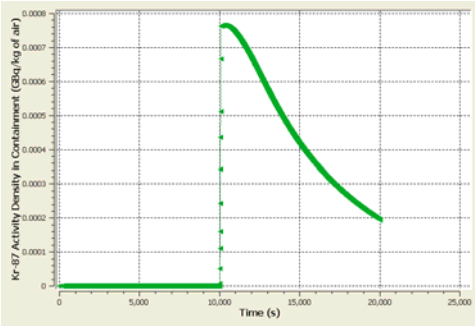

Fig. 12. Evolution of total Kr-87activity density in the containment (flow, per kg of gas in containment)

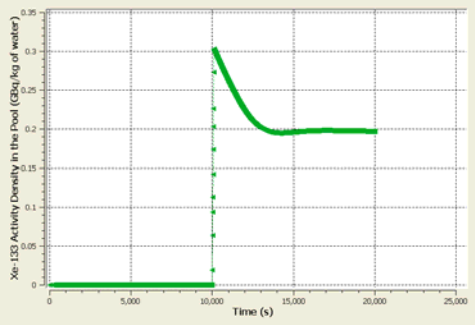

Fig. 13. Evolution of total Xe-133 activity density in the pool (residual flow, per kg of water)

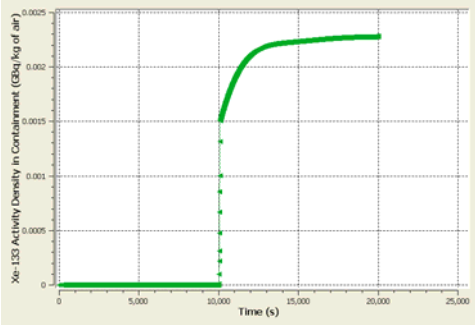

Fig. 14. Evolution of total Xe-133activity density in the containment (residual flow, per kg of gas in containment)

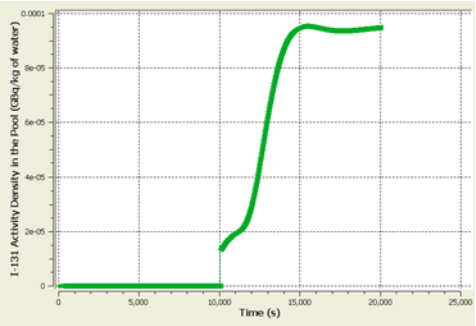

Fig.15.Evolution of total I-131 activity density in the pool (residual flow, per kg of water)

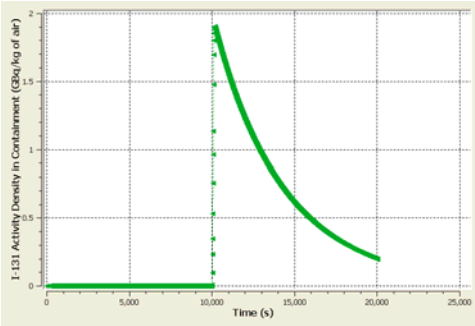

Fig. 16. Evolution of total I-131activity density in the containment (residual flow, per kg of gas in containment)

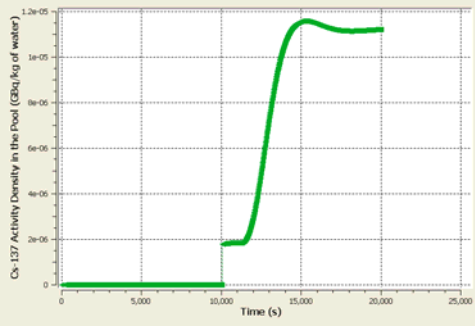

Fig. 17. Evolution of total Cs-137 activity density in the pool (residual flow, per kg of water)

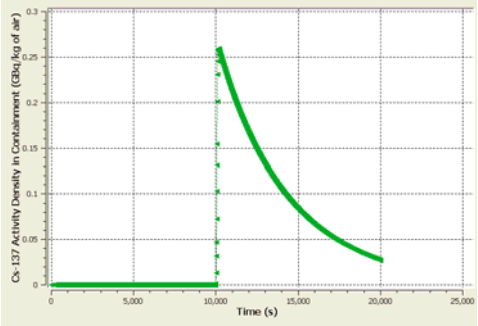

Fig. 18. Evolution of total Cs-137 activity density in the containment (residual flow, per kg of gas in containment)

Results with user specified CATHARE radio-elements properties

The behavior of noble gases resulted from calculations with CATHARE implicit properties for radio-elements lead to the ideea that an abnormally large condensation process is produced. The „DILU" constant in Table 3 seemed to be given very large (5000.E+3) to simulate a large gas into liquid dissolution time. We modified this time constant to be much larger (5000.E+9), but in order to do this, we had to specify completely the properties needed by CATHARE for radio-elements. Consequently, we calculated the „ka" constant and searched in the literature for the Henry constants of the gases. Table 5 gives our calculated „ka" for Kr-87, Xe-133, I-131 and Cs-137 using SCALE 4.4 results and the Henry constant found in [8] for Krypton, Xenon and Iodine (molecular I2) compared to CATHARE implicit values. Iodine was changed to gas (IRA=2) while Cesium is still treated as non-gaseous radio-species.

The evolution of total activity for Kr-87 and Xe-133 with modified parameters are given in Fig.19 through 22, for residual primary and nominal flowrate respectively (for comparison with implicit parameters see Fig.3 through Fig.6). The behavior of these gases is modified compared to the implicit properties behavior: primary flowrate is no more important because the concentration in water drastically diminishes while activity in containment becomes largely dominant, as proven by Fig.23 through Fig. 26.

I-131 and Cs-137 behave in the same manner as with implicit properties, remaining highly volatile. Our calculations for I-131 and Cs-137 do not take into account the chemical speciation of Iodine and Cesium, simply assuming the released fraction of these radio-nuclides as being pure species. Actually, in severe accidents, Cesium and Iodine interact with each other forming CsI which may be the main released chemical species, while molecular Iodine (I_2) will be only a fraction of the total Iodine inventory [9]. In the Final Safety Analysis Report for TRIGA SSR [5] , it is assumed that 10% of the halogens form organic compounds which are insoluble in water and 90% of the halogens are in elemental or particulate form of which all but 1% are released in the water. In case of LOCA, the above would not be valid, and the release will not be affected by water solubility or entrainment of the concentration in the circuit by means of primary flow.

Table 5

Implicit and user-defined constants for radio-nuclides

Component	ka (kg/GBq)		Henry (MPa)	
	implicit	calculated	implicit	modified
Kr-87	9.5E-13	9.52E-13	1.55E+4	2.234E+3
Xe-133	1.5E-10	1.44E-10	1.04E+4	1.299E+3
I-131	2.2E-10	2.18E-10	1.	1.862E+0
Cs-137	3.1E-7	3.1E-7	1.	1.

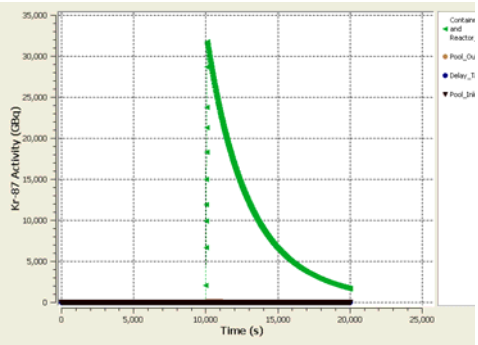

Fig. 19. Evolution of total Kr-87 activity in the primary circuit components (residual flow 50 l/s, modified properties)

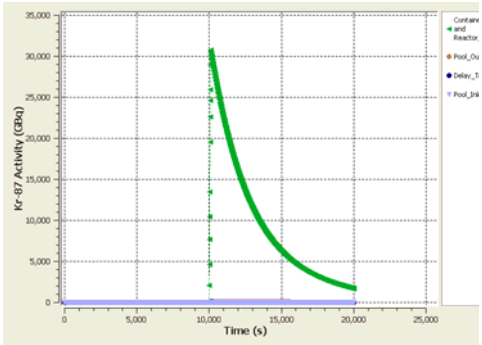

Fig. 20. Evolution of total Kr-87 activity in the primary circuit components (flow 504 l/s, modified properties)

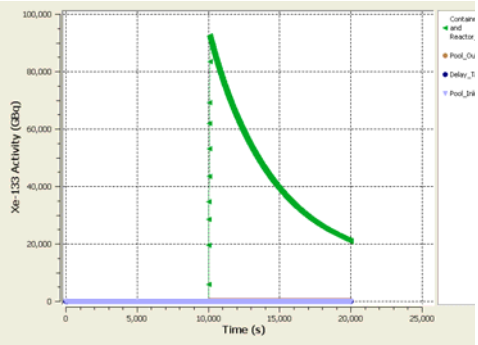

Fig. 21. Evolution of total Xe-133 activity in the primary circuit components (residual flow 50 l/s, modified properties)

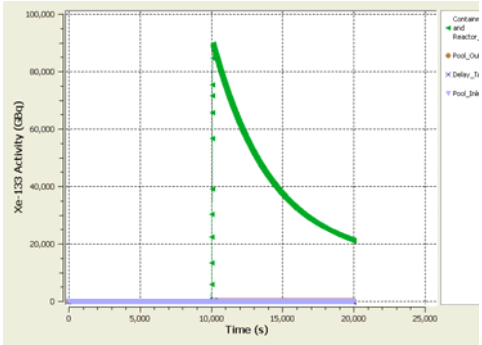

Fig. 22. Evolution of total Xe-133 activity in the primary circuit components (flow 504 l/s, modified properties)

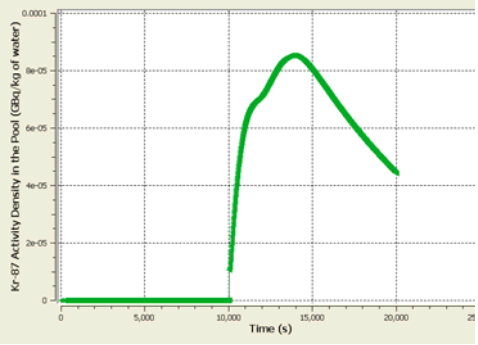

Fig. 23 Evolution of total Kr-87activity density in the pool (residual flow, per kg of water, modified properties)

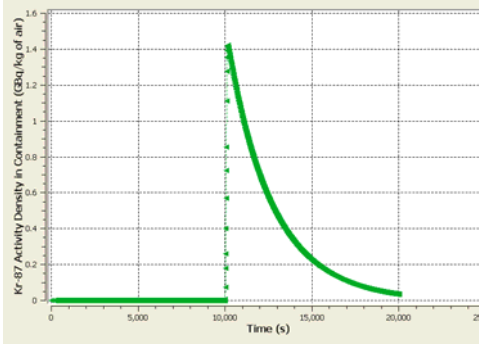

Fig. 24. Evolution of total Kr-87activity density in the containment (residual flow, per kg of gas in containment, modified properties)

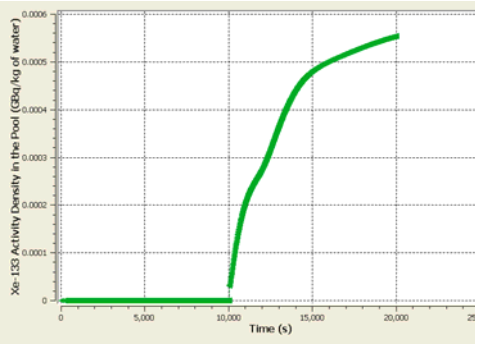

Fig. 25 Evolution of total Xe-133 activity density in the pool (residual flow, per kg of water, modified properties)

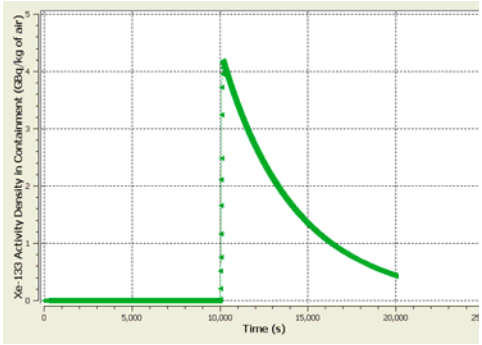

Fig. 26. Evolution of total Xe-133 activity density in the containment (residual flow, per kg of gas in containment, modified properties)

7. Conclusions

The paper presents a model for TRIGA reactor and investigation analyses of radio-chemical components behavior in the TRIGA facility with CATHARE2. It focuses on the four existent components in CATHARE2 (Kr-87, Xe-133, I-131 and Cs-137) for which calculations are performed with implicit and user defined properties.

The present work constitutes the start in Level 2 PSA support analysis. The internal Initiating Events that can lead to fuel damage and fission products release in PSA Level 1 for TRIGA reactor are Loss of Flow, Loss of Power Supply or Loss of Coolant Accident. The analyses with CATHARE2 study the transport of radio-chemical components inside the primary circuit and containment, and track their time evolution of concentration affected by disintegration, water entrainment and ventilation flowrate inside the reactor hall. A more extended list of radio-chemical components is needed for taking into consideration other gaseous or volatile fission products that contribute to the activity (and dose) that possibly result in case of an accident.

CATHARE2 implicit properties for the noble gases analyzed seem to lead to an abnormally large condensation process that makes important their activity in the primary circuit water. On the contrary, user defined properties (or specific parameters that control the behavior) introduced in the model produce large activities inside the containment atmosphere. I-131 and Cs-137 are highly volatile and mostly found in the containment both with implicit and user defined properties. CATHARE2, and consequently our calculations do not take into account the chemical interactions of the released species, assuming the inventory existent inside the fuel at the moment of cladding rupture will be released only with respect to the temperature.

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