RELEASABLE ACTIVITY AND MAXIMUM PERMISSIBLE LEAKAGE RATE WITHIN A TRANSPORT CASK OF TEHRAN RESEARCH REACTOR FUEL SAMPLES

by

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Containment of a transport cask during both normal and accident conditions is important to the health and safety of the public and of the operators. Based on IAEA regulations, releasable activity and maximum permissible volumetric leakage rate within the cask containing fuel samples of Tehran Research Reactor enclosed in an irradiated capsule are calculated. The contributions to the total activity from the four sources of gas, volatile, fines, and corrosion products are treated separately. These calculations are necessary to identify an appropriate leak test that must be performed on the cask and the results can be utilized as the source term for dose evaluation in the safety assessment of the cask.

Key words: transport cask, cask releasable activity, cask leakage rate, TRR fuel samples

INTRODUCTION

Technical aspects of mechanical, thermal, shielding, criticality, and containment requirements during both normal and accident conditions should be considered in design of a transport or storage cask [1-7]. The containment requirements are established by both national and international standards [8, 9]. According to the IAEA Regulations for the Safe Transport of Radioactive Material [10], limits on the releasable materials under normal and accident conditions are specified as a function of the contents and the actual quantity of material that may be released is dependent upon its isotopic constituents [11]. Since absolute containment cannot be guaranteed, the purpose of specifying maximum allowable activity leak rates is to permit the specification of appropriate and practical test procedures which are related to acceptable radiological protection criteria [12].

In order to transport irradiated fuel samples of Tehran Research Reactor (TRR), a transport cask which is categorized as a Type B(U) package, is intended to be designed based on the IAEA Regulations. In this study, releasable activity and maximum permissible volumetric leakage rate within the cask containing TRR fuel samples enclosed in an irradiated capsule are calculated. The schematic of the cask and the capsule is shown in fig. 1.

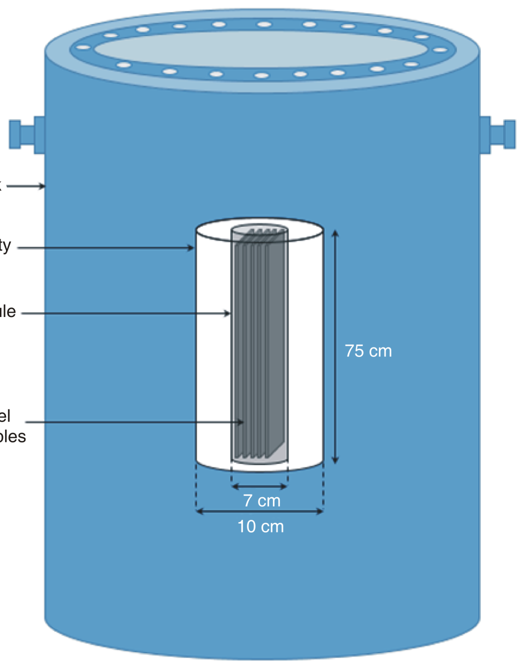

Figure 1. The schematic of the cask and capsule

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METHODOLOGY

According to the paragraph 659 of the IAEA Regulations, the cask shall be so designed that it would restrict the loss of activity to not more than $10^{-6}A_2$ per hour in normal conditions of transport (NCT) where A_2 (Bq) is dependent upon the isotopes being transported and it is tabulated in the IAEA Regulations. Also, the cask shall be so designed that it would restrict the accumulated loss of activity in a period of one week to not more than $10A_2$ for krypton-85 and not more than A_2 for all other radionuclides in hypothetical accident conditions (HAC). As the maximum permissible release rate for normal conditions of transport (R_N) is $A_2 \times 10^{-6}$ per hour, therefore

$$R_N = \frac{A_2 \times 10^{-6}}{3600} [\mathrm{Bqs}^{-1}] \quad (1)$$

On the other hand, as the maximum permissible release rate for accident conditions (R_A) is A_2 per week ($10A_2$ per week for krypton-85), therefore

$$R_A = \frac{A_2}{604800} [\mathrm{Bqs}^{-1}] \quad (2)$$

where mixtures of different radionuclides are present, $A_{mixture}$ shall apply, except for the krypton-85 that an effective $A_2(i)$ value may be used which is equal to $10A_2$.Thus, A_2 in eqs. (1) and (2) is equal to

$$A_{2,\,\mathrm{mixture}} = \frac{1}{\sum \dfrac{f(i)}{A_2(i)}} \quad (3)$$

where $A_2(i)$ and f_i are the appropriate A_2 value for nuclide *i* and the fraction of releasable activity of nuclide i in the mixture, respectively.

At first, the A_2 value is used to calculate the maximum permissible release rate and then the maximum permissible leakage rate for both NCT and HAC. For time-averaged volumetric concentrations of the suspended radioactivity within the cask C_N [Bqcm^{-3}], the maximum permissible leakage rates from the transport cask for the normal conditions of transport, L_N [$\mathrm{cm}^3\mathrm{s}^{-1}$], could be calculated by

$$L_N = \frac{R_N}{C_N} = \frac{A_2 \times 10^{-6}}{C_N \times 3600} [\mathrm{cm}^3\mathrm{s}^{-1}] \quad (4)$$

For time-averaged volumetric concentrations of the suspended radioactivity within the cask C_A [Bqcm^{-3}], the maximum permissible leakage rates from the transport cask for the accident conditions, L_A [$\mathrm{cm}^3\mathrm{s}^{-1}$], could be calculated by

$$L_A = \frac{R_A}{C_A} = \frac{A_2}{C_N \times 604800} [\mathrm{cm}^3\mathrm{s}^{-1}] \quad (5)$$

The activity per unit volume of the releasable radioactive material within the cavity under normal conditions, C_N [Bqcm^{-3}] is equal to

$$C_N = \frac{\text{Total releasable activity for NCT}}{\text{Free volume of the cavity}} = \frac{RA_N}{V_C} \quad (6)$$

Also, the activity per unit volume of the releasable radioactive material within the cavity under accident conditions, C_A [Bqcm^{-3}] is equal to

$$C_A = \frac{\text{Total releasable activity for HAC}}{\text{Free volume of the cavity}} = \frac{RA_A}{V_C} \quad (7)$$

where *RA* is the releasable activity and V_C is the free volume of the cavity. The free volume of the cavity in eqs. (6) and (7) is equal to the difference between the volume of the cavity and the volume of the capsule.

There are four sources of radioactive material that may become airborne during transportation. These sources are gases, volatiles, fines, and CRUD [13]. CRUD is a corrosion product whose activity comes primarily from cobalt. The principal volatile radionuclide is cesium [11]. If a cladding failure occurs, a large fraction of the gap inventories of the filled gas and gaseous fission products (tritium, iodine, krypton, and xenon) will be introduced into the free volume of the transport cask. Tritium and krypton-85 are typically the major sources of radioactivity among the present gases [14]. Volatile species, such as cesium, strontium, and ruthenium, can also be released from a fuel rod as a result of a cladding breach. Although some of these isotopes may only be volatile under the hypothetical accident conditions, they are included in the analysis for normal transport conditions as a conservative approach. The CRUD activity associated with the oxide layer of the cladding material. The fines are characterized as particles with diameters less than about 100 μm [14]. The fuels fines are particulate materials composed of the fuel compounds and are produced as a result of the mechanical stresses at the fuel-cladding interface. Fines can be ejected from the fuel rod through the cladding failure by the purging action of filled gas and gaseous fission products. A sudden release of the fuel fines occurs when the cladding integrity is initially lost, but no additional releases occur once equilibrium conditions are reached. The contributions to the total activity density in the shipping cask free volume from the four sources are treated separately. Therefore, the A_2 values of the four sources are determined separately. The A_2 for each of the four groups is calculated by eq. (3).The releasable activity of each nuclide is determined using set of factors. These factors are specified for each of four groups of nuclide (gas, volatile, fines and CRUD) [13]. Therefore, the releasable activity (*RA*) is determined using the factors as follows

$$RA \quad TA\, f_{\mathrm{b}}\, f_{\mathrm{i}} \tag{8}$$

where TA is the total activity which is calculated by ORIGEN, f_b and f_i are the fraction of breaching in the cask which includes the breaching in the fuel ($f_{b,f}$) and the capsule ($f_{b,c}$) and the fraction of gas or volatile that escapes from the breached fuel and cask, respectively. f_b is determined using

$$f_{\mathrm{b}} \quad f_{\mathrm{b,f}}\, f_{\mathrm{b,c}} \tag{9}$$

There are different factors for the fines group instead of f_i. Moreover, the determination of the releasable activity for the CRUD has a different method. Both fines and CRUD releasable activity calculations will be introduced later.

The source term associated with the contents of the irradiated TRR fuel samples are determined by use of ORIGEN code. Because an ORIGEN calculation provides output for over 800 radionuclides, the number of radionuclides in the ORIGEN output can be reduced by dividing the amount of each cooled radionuclide by its A_2 value and then selecting the smallest set of these normalized curie amounts that yielded a sum greater than 99.9 percent of the sum of all of these normalized curie amounts. This procedure reduces the ORIGEN output to a much smaller set of radionuclides that are important for the estimation of radiological health effects. In addition, in most fuels five radionuclides including ^{106}Ru, ^{134}Cs, ^{144}Ce, ^{147}Pm, and ^{154}Eu, available for release, were added so that the most of the fuel inventories contained the same set of radionuclides. In order to have a noble gas in the inventory, ^{85}Kr which is the noble gas with the largest curie amount in the ORIGEN inventory for irradiated fuel samples, is also added to the reduced set of radionuclides, although eliminated by the A_2 screen [15].

RESULTS AND DISCUSSION

The maximum permissible leakage rates are calculated for both normal condition of transportation (NCT) and hypothetical accident condition (HAC). $f_{b,c}$ is assumed to be equal to 1.00 during the failure of the capsule and it means that in this case, all of the radioactive nuclide is leaked out. In the normal condition, the $f_{b,f}$ is conservatively assumed to be equal to 1.00 because of the fact that the fuels are research samples which are purposed for irradiation condition evaluation in the TRR core and the simultaneous failure of all samples are probable.

Gas calculation

A mixture of A_2 is determined by eq. (3). The factors and releasable activity values for the gas group, A_{2mix}, are calculated and illustrated in tab. 1. The releasable activity is calculated using eq. (8). The f_G is the fission gas release fraction. The releasable activity density inside the cavity due to the release of gas is described by eqs . (4) and (5). Based on these calculations, $C_{gas,N}$ = 4.1366 10^4 Bq/cm^3 and $C_{gas,A}$ = = 2.63033 10^7 Bq/cm^3.

Volatile calculation

The released volatile which is considered in this analysis includes Cs, Sr, and Rb. The fractional release of volatiles (f_v) is estimated less than 10^{-6} based on the experimental data on the release of fission products during fuel melting experiments [13]. Those volatiles which occupy the volume fraction of fuel meat, are released as fines and it is also included in the fines calculation. A mixture of A_2 is determined by the eq. (3). The factors and releasable activity values for the fines group, A_{2mix}, are calculated and illustrated in tab. 2. The releasable activity for volatile is calculated using eq. (8) and is presented in tab. 2. The f_V is the fines release fraction. The releasable activity densities inside the cavity due to the release of gas are $C_{volatile,N}$ = 1.83816 10^1 Bq/cm^3 and $C_{volatile,A}$ = 1.16846 10^4 Bq/cm^3 for normal and accident conditions, respectively.

Fines calculation

A mixture of A_2 is determined by eq. (3). The factors and releasable activity values for the fines group

Table 1. Mixture A_2 determination for gases in TRR fuel samples

Nuclide	A_2 [Bq]	Activity* [Bq]	f_G	$f_{b,f}$	$f_{b,c}$	RA [Bq]	F_i**	F_i/A_{2i} [Bq^{-1}]
Normal condition								
^{85}Kr	9.99 10^{12}	2.63403 10^{11}	3.00 10^{-1}	1.00	1.57 10^{-3}	1.2395 10^8	1.00	1 10^{-13}
$\Sigma F_i/A_{2i}$ = 1.001 10^{-13} Bq^{-1} RA = 1.2395 10^8 Bq $A_{2gas,N}$ = 9.99 10^{12} Bq								
Accident condition								
Nuclide	A_2 [Bq]	Activity* [Bq]	f_G	$f_{b,f}$	$f_{b,c}$	RA [Bq]	F_i	F_i/A_{2i} [Bq^{-1}]
^{85}Kr	9.99 10^{12}	2.63403 10^{11}	3.00 10^{-1}	1.00	1.00	7.9032 10^{10}	1.00	1 10^{-13}
$\Sigma F_i/A_{2i}$ = 1.001 10^{-13} Bq^{-1} RA = 7.9032 10^{10} Bq $A_{2gas,A}$ = 9.99 10^{12} Bq								

*The activity calculated by ORIGEN: **The releasable activity fraction

Table 2. Mixture A_2 determination for volatile in TRR fuel samples

Normal condition								
Nuclide	A_2 [Bq]	Activity [Bq]	f_V	$f_{b,f}$	$f_{b,c}$	RA [Bq]	F_i	F_i/A_{2i} [Bq^{-1}]
^{89}Sr	7.4 10^8	3.64931 10^{11}	10^{-6}	1.00	1.57 10^{-3}	4.42150 10^5	8.009 10^{-1}	1.83 10^{10}
^{90}Sr	7.4 10^8	2.76057 10^{11}	10^{-6}	1.00	1.57 10^{-3}	3.27339 10^4	5.928 10^{-2}	4.06 10^{10}
^{137}Cs	7.4 10^8	2.19077 10^{11}	10^{-6}	1.00	1.57 10^{-3}	3.53905 10^4	6.409 10^{-2}	4.39 10^{10}
^{134}Cs	7.4 10^8	8.7727 10^{12}	10^{-6}	1.00	1.57 10^{-3}	4.181 10^4	7.570 10^{-2}	1.48 10^9
$\Sigma F_i/A_{2i}$ = 1.0431965 10^{-11} Bq^{-1} RA = 5.5204 10^5 Bq $A_{2\text{volatile,}}$ = 1.31239 10^{11} Bq								
Accident condition								
Nuclide	A_2 [Bq]	Activity [Bq]	f_V	$f_{b,f}$	$f_{b,c}$	RA [Bq]	F_i	F_i/A_{2i} [Bq^{-1}]
^{89}Sr	7.4 10	3.64931 10^{11}	10^{-6}	1.00	1.00	2.81 10^8	8.009 10^{-1}	1.83 10^{10}
^{90}Sr	7.4 10	2.76057 10^{11}	10^{-6}	1.00	1.00	2.08088 10^7	5.928 10^{-2}	4.06 10^{10}
^{137}Cs	7.4 10	2.19077 10^{11}	10^{-6}	1.00	1.00	2.24997 10^7	6.409 10^{-2}	4.39 10^{10}
^{134}Cs	7.4 10	8.7727 10^{12}	10^{-6}	1.00	1.00	2.65734 10^7	7.570 10^{-2}	1.48 10^9
$\Sigma F_i/A_{2i}$ = 1.0431965 10^{-11} Bq^{-1} RA = 3.510449 10^8 Bq $A_{2\text{volatile,}}$ = 1.31239 10^{11} Bq								

are calculated. All the nuclides are in the fines group except for the gases [16]. As mentioned previously, there are additional factors for calculating the releasable activity and explained in releasable activity relationship as presented [13]

$$RA \quad Fines\ activity \quad f_b \quad ESA \quad P\frac{T_F}{V_M} \qquad (10)$$

where ESA is the amount of exposed meat surface area per cask and is 1 % of outside plates for normal and accident conditions [cm^2], P [cm] – the depth of corrosion attack, T_F – the oxide spallation fraction, V_M [cm^3] – the volume of the meat region of the fuel per cask.

For the TRR samples, the ESA is the surface of two largest side 1830 cm^2 of each samples multiply by 1 % which is determined by experimental data [13]. Therefore, ESA = 1.83 10^3. The factor P is the depth of corrosion attack and it is assumed to be 5 10^{-4} cm [13] and depends on the materials of the fuel samples and the thermo-hydraulic and the chemical conditions of the coolant. In this case although the fuel samples are in enclosed irradiated capsule and are not prone to the coolant, conservatively it is assumed that the fuels are in the corrosion conditions due to the capsule failure. The term $T_{F,N}$ = 0.15 and $T_{F,A}$ = 1.0 for NCT and HAC, respectively. The aluminum oxide films are very tenacious and resistant to spallation. However, fuel handling can cause scratches in the coating, resulting in breach of the oxide and providing sites susceptible to initiation of pitting corrosion. If the water chemistry is aggressive, pitting can occur [16]. The V_M = 640.5 cm^3 is used here to obtain the fraction of corrosion respect to the fuels meats. Therefore, the calculated releasable activity densities inside the cavity are $C_{\text{fines,N}}$ = 8.51 Bq/cm^3 and $C_{\text{fines,A}}$ = 2.31065 10^7 Bq/cm^3.

CRUD calculation

Aluminum spent fuel do not acquire CRUD in the same manner as commercial spent nuclear fuel (SNF). The surface activity of Al-SNF is primarily a result of storage in radioactively contaminated water. The $A_{2\text{mix}}$, the factors and releasable activity values for the fines group are calculated and illustrated in tab. 3. The calculated A_2 value is 9.99 10^9 Bq [13]. The RA inside the containment vessel due to the release of fines is described by [13]

$$RA \quad f_C S_C S_A \qquad (11)$$

where $f_C = f_i$ is the CRUD spallation fraction ($f_{C,N}$ = = 0.15, $f_{C,A}$ = 1.0), S_C – the CRUD surface activity (5.143 10^3 Bq/cm^2), S_A – the sum of the surface areas of all mini-plate plus outer surface of capsule. For TRR fuel samples, S_A = 29241.338 cm^2. Therefore, RA densities are $C_{\text{CRUD,N}}$ = 7.5073 10^3 Bq/cm^3 and $C_{\text{CRUD,A}}$ = 5.0061 10^4 Bq/cm^3.

Combining the sources of radioactive material in cask free volume

The contributions to the total activity density in the shipping cask free volume from the four sources are combined by

$$C_{\text{total}} \quad C_{\text{gas}} \quad C_{\text{volatile}} \quad C_{\text{fines}} \quad C_{\text{CRUD}} \qquad (12)$$

The total releasable activity density inside the cavity due to the release of gases, volatiles, fines, and CRUD is thus $C_{\text{total,N}} = C_N$ = 4.8914 10^3 Bq/cm^3 and $C_{\text{total,A}} = C_A$ = 4.9469 10^7 Bq/cm^3. The mixture A_2 values derived previously are combined to determine a group A_2 for normal and accident conditions of transport, respectively, using eq. (3). The tab. 4 illustrates

Table 3. Mixture A_2 determination for CRUD in TRR fuel samples

Normal condition						
A_2 [Bq]	f_C	S_A [cm^2]	S_C [Bqcm^{-2}]	RA [Bq]	F_i	F_i/A_{2i} [Bq^{-1}]
9.99 10^9	0.15	2.924 10^4	5.143 10^3	2.25552 10^7	1.00	1 10^{-10}
$\Sigma F_i/A_{2i}$ = 1.001 10^{-10} Bq^{-1} RA = 2.25552 10^7 Bq $A_{2CRUD,N}$ = 9.99 10^9 Bq						
Accident condition						
A_2 [Bq]	f_C	S_A [cm^2]	S_C [Bqcm^{-2}]	RA [Bq]	F_i	F_i/A_{2i} [Bq^{-1}]
9.99 10^9	1.00	2.924 10^4	5.143 10^3	1.50368 10^6	1.00	1 10^{-10}
$\Sigma F_i/A_{2i}$ = 1.001 10^{-10} Bq^{-1} RA = 1.50368 10^6 Bq $A_{2CRUD,A}$ = 9.99 10^9 Bq						

Table 4. A_2 and releasable activity density in both NCT and HAC in TRR fuel samples

Source	A_{2i} value [Bq]	C_i value [Bqcm^{-3}]	Fraction (C_i/C_t)	Fraction/A_{2i}
Gas	9.99 10^{12}	4.14 10^4	8.459 10^{-1}	8.46747 10^{-13}
Volatile	1.31 10^{11}	1.84 10^1	3.758 10^{-4}	2.86348 10^{-14}
Fines	5.54 10^{10}	8.51	1.740 10^{-4}	3.13932 10^{-14}
CRUD	9.99 10^9	7.51 10^3	1.535 10^{-1}	1.53654 10^{-10}
ΣC_i = 4.890019 10^4 Bqcm^{-3} F_i/A_{2i} = 1.54560428390183 10^{-10} Bq^{-1}				
Group $A_{2,N}$ = 6.4676 10^{10} Bq				
Source	A_{2i} value [Bq]	C_i value [Bqcm^{-3}]	Fraction (C_i/C_t)	Fraction/A_{2i}
Gas	9.99 10^{12}	2.63 10^7	5.317 10^{-1}	5.32232 10^{-13}
Volatile	1.31 10^{11}	1.17 10^4	2.362 10^{-4}	1.79977 10^{-14}
Fines	5.38 10^{10}	2.31 10^7	4.670 10^{-4}	8.68062 10^{-14}
CRUD	9.99 10^9	5.01 10^4	1.012 10^{-3}	1.01301 10^{-12}
ΣC_i = 4.94715456 10^7 Bqcm^{-3} F_i/A_{2i} = 1.65004914507402 10^{-12} Bq^{-1}				
Group $A_{2,A}$ = 1.13146 10^{11} Bq				

the A_2 and releasable activity density in normal and accident conditions.

Maximum permissible release rate and maximum permissible leakage rate

Assuming that the release rate is independent of time, the maximum permissible release rates for normal and accident conditions of transport can be expressed using eqs. (1) and (2). Therefore, R_N = = 1.79783 10^1 Bq/s and R_A = 1.86702 10^5 Bq/s. The maximum permissible leakage rate is calculated using eqs. (4) and (5). The results of these calculations are L_N = 3.675 10^{-4} cm^3/s and L_A = 3.774 10^{-3} cm^3/s for normal and accident conditions, respectively. The maximum permissible leakage rates, L_N and L_A, must be converted to an equivalent air-leakage rate under standard conditions to identify possible leak-test methods. Something that is often overlooked is that the regulations specify that the package leak tightness must be demonstrated to a sensitivity of A_2 10^{-6} Bq/h. Since test sensitivity is specified as R_N/2 in the ANSI N14.5 Standard, the allowable leak rate is then R_N = = 2A_2 10^{-6} Bq/h. Ordinarily, the leak rate is calculated, and reported, as A_2 10^{-6} Bq/h, and the test sensitivity is set to one-half of this value.

CONCLUSIONS

Releasable activity and maximum permissible volumetric leakage rate within the transport cask containing TRR fuel samples enclosed in an irradiated capsule are calculated. Four sources, which are treated separately, have been considered to the total activity density in the free volume of the shipping cask. Based on the calculations, the maximum permissible volumetric leakage rates for normal and accident conditions are L_N = 3.675 10^{-4} cm^3/s and L_A= 3.774 10^{-3} cm^3/s, respectively.

The calculated maximum permissible leakage rates and releasable activities for NCT and HAC can be utilized for identifying an appropriate leak test that must be performed on the cask and dose evaluation in the safety assessment of the cask. Also, the calculated releasable activities of normal and accident conditions can be utilized for dose evaluation in the safety assessment of the cask.

AUTHOR CONTRIBUTIONS

The methodology of calculations was performed by M. Rezaeian. Three separate ORIGEN calculations were carried out by M. Rezaeian, A. Moosakhani, and

E. Noori. The results of calculations were checked by M. Roshanzamir. The analysis of results was a joint undertaking of all authors. J. Kamali provided overall guidance during the study and interpreting the results. The manuscript was written by M. Rezaeian and was reviewed by M. Roshanzamir and J. Kamali. The figures and the tables were prepared by A. Moosakhani and E. Noori.

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ОСЛОБОЂЕНА АКТИВНОСТ И МАКСИМАЛНА ДОЗВОЉЕНА ЈАЧИНА ЦУРЕЊА У ТРАНСПОРТНОМ БУРЕТУ СА УЗОРЦИМА ГОРИВА ТЕХЕРАНСКОГ ИСТРАЖИВАЧКОГ РЕАКТОРА

У нормалним условима рада као и у току акцидената, изоловање транспортног бурета важно је ради здравља и безбедности становништва и оператора. У сагласности са прописима Међународне агенције за атомску енергију, израчунати су ослобођена активност и максимална дозвољена запреминска јачина цурења унутар бурета које садржи узорке горива Техеранског истраживачког реактора затвореног у озраченим капсулама. Доприноси укупној активности из четири различита извора, гаса, паре, праха и производа корозије, разматрани су засебно. Ови прорачуни потребни су да би се утврдио одговарајући тест цурења бурета који се мора спровести, а резултати употребити као изворни члан за одређивање дозе у процени безбедности бурета.

Кључне речи: транспортно буре, ослобођена активност из бурета, цурење из бурета, TRR узорци горива