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Preliminary Study on Implementing a Simplified Source Terms Estimation Program for Early Radiological Consequences Analysis

Theo Alvin Ryanto[1)], **Jupiter Sitorus Pane**[1*)], **Muhammad Budi Setiawan**[1)], **Ihda Husnayani**[1)], **Anik Purwaningsih**[1)], **Hendro Tjahjono**[1)]

[1)] *Research Center for Nuclear Reactor Technology, South Tangerang, Banten 12710, Indonesia.*

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Indonesia possesses numerous potential sites for nuclear power plant development. A fast and comprehensive radiological consequences analysis is required to conduct a preliminary analysis of radionuclide release into the atmosphere, including source terms estimation. One simplified method for such estimation is the use of the Relative Volatility approach by Kess and Booth, published in IAEA TECDOC 1127. The objective of this study was to evaluate the use of a simple and comprehensive tool for estimating the source terms of planned nuclear power plants to facilitate the analysis of radiological consequences during site evaluation. Input parameters for the estimation include fuel burn-up, blow-down time, specific heat transfer of fuel to cladding, and coolant debit, using 100 MWe PWR as a case study. The results indicate a slight difference in the calculated release fraction compared to previous calculations, indicating a need to modify the Relative Volatility method for high-fuel burn-up implementation.

1. INTRODUCTION

Source term estimation is one of the essential assessments that need to be conducted for evaluating radiological consequences analysis of Nuclear Power Plant (NPP) construction [1-3]. The source terms represent the mix and magnitude of the radionuclides measured in becquerels (Bq) from the fraction of fuel inventory that could be released to the environment. Predicting and modeling the source terms during nuclear accidents is vital for NPP emergency responses [4,5].

The estimated source terms will be used as an input for atmospheric radioactive dispersion modeling of radionuclide releases during severe nuclear reactor accidents. It is vital to measure the radiological consequences of the fission products released into the environment as an input to the radiological safety of the environment in the nuclear installation site vicinity in case of emergency [6]. Along with that, the source terms estimation could also be used to evaluate the design of a radiation safety system for a power reactor.

Various methods are commonly employed to estimate source terms, including those found in FASTGRASS, SCDAP, VICTORIA, MAAP, MELCOR, ICARE, ELSA, and KESS, as well as a simplified Booth-type model. However, it is important to note that not all of these models consider the correlations between the effects of fuel burn-up. Alternatively, the RelVol model, based on Booth-type kinetics, is one of the source term

[*] Corresponding author
E-mail: theo001@brin.go.id
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estimation methods that consider the correlations. An example of RelVol model calculation for the PWR reactor is the SURRY plant source terms modeling.

This preliminary study aimed to develop a simple and comprehensive tool to estimate the source terms of installed NPPs to facilitate the analysis of radiological consequences during site evaluation at various locations in Indonesia. The aim is for the simplified program to be incorporated into the radiological consequences assessment as a comprehensive tool for site evaluation.

As a case study, the calculation was applied to SMART, a330 MWth PWR-type reactor, since this reactor was adopted as an alternative nuclear plant that will be installed at Pantai Gosong. The calculated source terms can be compared to the estimated SMART source terms values published in Udiyani [7] to compare the accuracy of the calculated models. Table 1 shows the SMART reactor characteristic details.

Table 1. SMART Characteristic

GENERAL INFORMATION	
Reactor Name	PWR 100 SMART
Reactor Type	Integral PWR
Thermal Power (MWth)	330
Electrical Power (MWe)	100
Design Life Time (yr)	60
Fuel Assembly matrix	17×17 Square FA
Reactor Core System Rod material	
Fuel Material	Low-enriched U O_2
Core Power Density (w/cc)	62.6
Refueling Cycle (yr)	3
Average Fuel Enrichment wt.%	5%
Uranium mass first loading (ton)	22.70
Fissile mass Loading (ton)	1.13
Active Core Height (m)	2
Core Diameter (m)	1.72
No. of Fuel Assembly	57
RPV Height (m)	18.5
PRV outer diameter	6.5
Control rod material	Zircaloy-4
Cladding material	Zircaloy-2
Design pressure (MPa)	17
Design temperature (°C)	350
Operating pressure (MPa)	15
Core inlet temperature (°C)	296
Core exit temperature (°C)	323
Steam generator type	OTSG U Tube
Steam generators	8
Reactor Coolant pumps	4

2. METHODS

The calculation of source terms estimation begins with the release from coolant activity caused y a break or leak in the reactor coolant system. The second step is fuel cladding failure, causing the release of the activity between the fuel pellet and the fuel cladding. The third, fuel degradation, starts, leading to a loss of fuel geometry and gradual melting and slumping of the core materials to the bottom of the reactor pressure vessel. We call this an early in-vessel release phase, and most of the noble gases (Xe, Kr) and significant amounts of volatile nuclides such as iodine (I) and cesium (Cs) are released into containment. The fourth step is started when the molten core debris from the reactor pressure vessel may interact with the concrete structural materials of the cavity below the reactor, resulting in ex-vessel releases of some less-volatile nuclides (Sr, Ba, La, Ce, Te) into the containment.

The presence of water in the reactor cavity above the core debris can significantly reduce the ex-vessel releases into the containment by cooling the core debris or scrubbing the releases and retaining a significant fraction of the water. The late in-vessel releases of some of the volatile nuclides deposited in the reactor coolant system during the in-vessel phase will also occur and be released into the containment.

The release of fission products into the containment could also be affected by two other phenomena. The first is "high-pressure melt ejection" (HPME), and the second is a possible steam explosion resulting from interactions between molten core debris and water. For the first one, where the Reactor Cooling System (RCS) is at high pressure at the time of failure of the bottom head of the reactor pressure vessel, certain quantities of molten core materials could be injected into the containment at high velocities. This condition leads to a potentially rapid rise in containment temperature and a significant amount of radioactive material, primarily aerosols. The second phenomenon leads to fine fragmentation of some portion of the molten core debris with an increase in airborne fission products. Small-scale steam explosions are likely to occur but will not significantly increase airborne activity within containment. On the other hand, large-scale steam explosions could significantly increase airborne activity but are much less likely to occur. In any event, releases of particulates or vapors during steam explosions will also be accompanied by large amounts of water droplets, which tend to sweep released material from the atmosphere quickly.

Several engineering safety features provided within the containment automatically reduce the number of radionuclides. The radionuclides that remain in the containment are released into the environment at a specific leakage rate [8].

Therefore, the source terms calculation for the environment was divided into four main steps. The first step involved the calculation of core inventory. The second step was dedicated to the calculation of radionuclides that were released into the vessel, followed by the third step, which involved the calculation of radionuclide release into the containment. The final step was focused on the calculation of radionuclide release into the environment. A visual representation of the process can be found in Fig. 1.

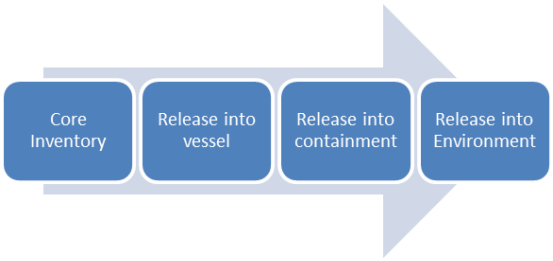

Fig. 1. Steps of radionuclide release to the environment

Core Inventory

The core inventory calculations represent the inventory of fissile radionuclides available from the fuel that might be released into the vessel. Several code programs were developed to estimate the core inventory that could be used for this research, namely the ORIGEN 2.1 or MCN. However, the core inventory activity data used in this research was published data from other sources, such as IAEA Publication or other researchers.

Release into Vessel

The simplification of release into vessel modeling in this study was calculated using the thermal-hydraulic relative volatility approach. A detailed process of this approach was explained in the IAEA Tecdoc 1127 [9]. In this approach, the reactor core is assumed to experience a loss of coolant due to a burst pipe in the primary system followed by a reactor shutdown.

The cooling period when an emergency cooling failure accompanies the water drops is called the blowdown time. The average peak of cladding temperature at that time is around 810 K or 538 °C. Even if the reactor is in the shutdown condition, the heating process would still occur due to residual heat in nuclear fuel, and the temperature would rise. When the temperature reaches about 1500 K or 1227 °C, an exothermic oxidation reaction occurs between water vapor and zirconium, producing hydrogen and heat. During this oxidation process, the heat would increase by the oxidation of zirconium. If water supply is insufficient, the temperature will continue to increase until it reaches the core melting temperature. This condition will last until everything melts or is stopped by the intervention of the safety system. The temperature in this condition is constant, called the hold time.

In the relative volatility method, the radionuclides fission product release modeling was commonly calculated using the spherical diffusion equation proposed by Kress et al.:

$$\partial C/\partial t = (D/r)[\partial^2 (rC)/\partial r^2] \quad (1)$$

where D = D_0 exp(-Q/RT), C is the concentration of the radionuclides, R is the universal gas constant (8.314 x 10^{-3} kJ mol-1K-1), T is the temperature in K, Do and Q is the Arrhenius correlation parameter (Cal/mol) which is taken from transient experiment data. The empirical correlation could be calculated by numerical time integration methods for various fission product species. The temperature transient, D_0, Q, and a need to be specified for the calculation. The solution for the numerical time integration approach was proposed by Booth and Rymer:

$$for\ Dt/a^2 \leq 0.1, \quad f = 6(Dt/\pi a^2)^{1/2} - 3Dt/a^2 \quad (2)$$

$$for\ Dt/a^2 \leq 0.1,\ f = 6(Dt/\pi a^2)^{1/2} - 3Dt/a^2 \quad (3)$$

where f is the cumulative fraction for every radionuclide released from the fuel at a certain time (t), a is the effective spherical parameter in cm, and D is the effective diffusion parameter in cm^2/s.

In severe accident conditions, the fuel temperature and D value would be varied over time. Thus, the approach to calculate the transient D value is by summing the D value in the short time period (Δt), using equation 4:

$$Dt = \sum D(t)\Delta t \quad (4)$$

by using the experimental data for the release of Cs and Sb from the ONRL, the "best-fit" approach was obtained as:

<u>for Cs</u>

$D_0 = (2.6833\ x\ 10^5)\ exp[-6.052\ x\ 10^{-4}(BU)]$
$Q = (2.065\ x\ 10^5) - (3.629)(BU)$
$a = 6.0\ \mu m$

<u>for Sb</u>

$D_0 = (3.4608\ x\ 10^6)\ exp[-6.052\ x\ 10^{-4}(BU)]$
$Q = (2.494\ x\ 10^5) - (3.629)(BU)$
$a = 6.0\ \mu m$

in which *BU* is the burn-up in MWd/t, D_0 is in cm^2/s and Q is in cal/mole.

Table 2. Relative Volatility Scale

GROUP	RV
NG	1.10
Te	1.07
I	1.03
Cs	1.00
Sb	0.68
Ba	0.42
Sr	0.34
Ru	0.25
La	0.14
Ce	0.085

Table 2 shows the relative volatility scale used to establish the fractional release model of all radionuclides as being the Kress/Booth RelVol model. The releases fraction of radionuclides could be determined by this interpolation equation:

$$f(i) = f(Cs)\left[\frac{f(Cs)}{f(Sb)}\right]^{\left(\frac{RV(i)RV(Cs)}{RV(Cs)RV(Sb)}\right)} \quad (5)$$

where f(i) is the fraction release of radionuclide i, f(Cs) and f(Sb) is the fraction release of Cs and Sb, RV(Cs), RV(Sb) and RV(i) is the relative volatility of Cs, Sb and i.

Release into Reactor Cooling System

The RCS's impact on fission products' behavior is significant, as it can attenuate their release, change their timing, and affect their chemical forms during severe accidents. However, these effects are minimal in low-pressure scenarios where the fission product residence time in the RCS is brief. Future LWRs will probably use depressurization as a strategy, which simplifies regulatory expectations and the development of a reference source terms by excluding credit for the effects of the RCS. Although this exclusion does not introduce any unacceptable distortion into the design basis "representative" source terms, it should be noted that it may not necessarily provide a maximum value. This condition was due to materials like control rod alloy and boric acid that could influence iodine's behavior in the containment during accidents, mitigating or exacerbating their consequences. Especially for activation product which is affected by corrosion rate [7]:

$$C_i(t) = \frac{P_{i,n}}{\lambda_i WP}\{1 - \exp(-\lambda_i t)\} + C_i(t_{n-1})\exp(-\lambda_i t) \quad (6)$$

where $\boldsymbol{C_i}$ is the radionuclide activity in the reactor coolant in Ci/kg, $\boldsymbol{P_i}$ is the rate of release of nuclei i from the core to the reactor cooler in Ci/s, WP is the reactor cooling capacity in kg, t is time in dt, $\boldsymbol{\lambda_i}$ is the nuclear transformation constant of radionuclide i in dt-1.

Release into Containment

Despite the differences in the design of the nuclear power plant, a general release group to the containment phases was derived according to the degree of fuel melting and relocation. There are four main release phases. The first one is the gap release phase which was the fuel cladding failure that would cause a release of radioactivity in the gap between the fuel pellet and fuel cladding. When the fuel degradation begins, the early in-vessel release phase is started. The gradual melting and slumping of core materials to the bottom of the vessel would also happen in this phase. The highly volatile nuclides, including all noble gases and a major fraction of Iodine and Cesium, were assumed to be released into the containment. The third phase, ex-vessel release, would occur when the bottom head of the reactor vessel failed, and the molten core debris interacted with the structural materials below the reactor. The less volatile nuclide would be released to the containment in this phase. The last phase was the late in-vessel release, which would begin when the molten core debris exits the reactor vessel and finish when the debris cooled sufficiently. Some volatile nuclides deposited in the reactor coolant system during the in-vessel phase would be released into the containment.

The radionuclides were divided into seven major groups based on similarity in chemical behavior. The release fraction for each phase on these radionuclides group is shown in Table 3. It is noted that the following set of radionuclides can affect human beings for the whole body such as noble gases (particularly ^{88}Kr, ^{135}Xe, and ^{133}Xe); Thyroid: iodines (particularly ^{131}I, ^{133}I); Lung/internal: volatile nuclides (e.g., ^{131}I, ^{132}Te, ^{106}Ru, ^{134}Cs, ^{137}Cs) and, for scenarios of high core temperatures (>1000°C), ^{90}Sr.

Table 3. Release Fraction of PWR Vessel

Elements	*gap release*	*early in vessel*	*ex-vessel*	*late invessel*
Kr, Xe	0.0500	0.9500	0.0000	0.0000
I	0.0500	0.3500	0.2500	0.1000
Rb, Cs	0.0500	0.2500	0.3500	0.1000
Te	0.0000	0.0500	0.2500	0.0050
Ba, Sr	0.0000	0.0200	0.1000	0.0000
Ru	0.0000	0.0025	0.0025	0.0000

In-containment behavior of fission products

The behavior of fission products within a containment structure determines the in-containment source terms and subsequent release of radioactivity. Regulatory requirements specify design-basis source terms and acceptable dose criteria at the site boundary, given a pressure source in the containment coupled with a design leak rate. Engineered safety features such as sprays and suppression pools are allowed for containment cooling and removal of gaseous iodine. The in-containment behavior of fission product aerosols is evaluated using reliable aerosol behavior phenomena, agglomeration, gravitational settling, and diffusional plate out. The use of engineered safety features is given credit in proportion to their reliability during severe accidents. Mechanistic aerosol codes can be used to determine transient aerosol behavior, and simple algorithms are proposed to capture the essence of these mechanical models. Specific Safety Guide (SSG-53) provides guidance on the design of reactor containment and associated systems for nuclear power plants to ensure their safety. Specific Safety Guide (SSG-53) provides guidance on the design of reactor containment and associated systems for nuclear power plants to ensure their safety. The containment system includes the reactor building, which provides primary containment, and auxiliary buildings, which provide secondary containment.

Release into the environment as a Source term

The radionuclide that reaches the containment is assumed to pass to the environment with a certain leakage rate. The leakage rate to the environment for a fraction of radionuclide could be calculated using Eq. 7 [10].

$$L(t) = \frac{B_0 X}{2400} \cdot \frac{1}{\left(\lambda + \frac{X}{2400}\right)} \cdot \left[1 - exp\left[-\left(\lambda + \frac{X}{2400}\right) t\right]\right] \quad (7)$$

where $L(t)$ is the leakage rate in Bq/hour, B_0 is the radionuclide concentration in Bq/m^3, X is the leakage percentage per day, λ is the half-life time of the radionuclide in hours, and t is the duration in hours.

3. RESULTS AND DISCUSSION

This study uses the core inventory from 100 MWe SMART calculated by Udiyani [7]. The entire fuel core inventory is shown in Table 4. However, in this research, the melted fuel fraction was assumed to be the same with Udiyani [7], which was 33% melted, to make the results comparable.

Table 4. Core Inventory

Elements	Half-life	Activity Released (Bq)
^{85m}Kr	4.4 hours	8.76 x 10^{16}
^{88}Kr	116 days	2.45 x 10^{17}
^{133}Xe	5.3 days	3.76 x 10^{17}
^{131}I	8.0 days	3.37 x 10^{17}
^{134}Cs	2.0 years	4.48 x 10^{16}
^{137}Cs	30.0 years	3.38 x 10^{16}
^{132}Te	78.0 hours	4.85 x 10^{17}
^{90}Sr	28.0 years	2.55 x 10^{16}
^{140}Ba	12.8 days	6.19 x 10^{17}
^{106}Ru	39.6 days	1.53 x 10^{17}

In-Vessel Release Fraction Calculation

The in-vessel source terms release rate was calculated based on the relative volatility method described in IAEA TECDOC 1127. Based on Kim et al. [11] the result of blowdown analysis for the SMART using the MARS-KS code, the blowdown time is 9 seconds for a double-ended guillotine break scenario. The normal operating power is 330 MWth with an average burn-up of approximately 40,000 MWd per metric ton of uranium (MTU). The melting point of the cladding material (Zircaloy-4) used in a SMART 100 MWe reactor is approximately 3,362 °F or 1851.11 °C or 2,124.26 K. Residual water mass to the top of the core is 15,794 kg and the residual water to core uncovery is 1,133 kg. Table 5 shows the Parameter Input for RelVol Calculation, and Table 6 shows the fractional activity release for each radionuclide with a 33% fuel melting assumption.

Containment Release Fraction Calculation

The release from the vessel to containment release rates calculation was based on general containment release phases according to the degree

of the meltdown. Because the calculation assumed that the core meltdown occurred, the fractional release from gap release to the late in-vessel release was summed. Table 7 shows the fractional release rates at the containment.

To minimize the release of radionuclides into the environment, a containment system is utilized to ensure that the radionuclides remain within the containment. In addition, various engineering safety features, such as agglomeration and gravitational settling, aerosol removal via spray, and pool scrubbing decontamination factors, are implemented. These measures can reduce the concentration of radionuclides by up to 60% [7].

Table 5. Input Parameters for RelVol Calculation

Parameter	Value
The average core burn-up	40,000 MWd/t
Normal operating power	330 MWth
Blowdown time	9 s
Core fuel/clad MCp	25,428 BTU/°F
Residual water mass to top of the core	15,794 Kg
Residual water to core uncovery	1,133 Kg
Runaway zr oxidation heatup rate	38 °F/s

Table 6. In-Vessel Fractional Activity Released

Elements	Fractional Releases	Activity Released (Bq)
^{85m}Kr	0.9674	2.80×10^{16}
^{88}Kr	0.9674	7.82×10^{16}
^{133}Xe	0.9674	1.20×10^{17}
^{131}I	0.5846	6.50×10^{16}
^{134}Cs	0.4711	6.96×10^{15}
^{137}Cs	0.4711	5.25×10^{15}
^{132}Te	0.7796	1.25×10^{17}
^{90}Sr	0.0041	3.45×10^{13}
^{140}Ba	0.0072	1.47×10^{15}
^{106}Ru	0.0021	1.06×10^{14}

Release to Environment Calculation

The fractional activity of the radionuclide which reached the containment would be released to the environment with a leakage rate that could be calculated using Eq. 7. In this calculation, the containment volume is needed to calculate the radionuclide concentration from the released activity and would be assumed to have a value of 50,625 m^3. The leakage percentage would be assumed to be 0.1% per day. Table 8 shows the calculated leakage rate for each radionuclide.

Table 7. Containment Fractional Activity Released

Elements	Fractional Releases	Activity Released (Bq)
^{85m}Kr	1	2.80×10^{16}
^{88}Kr	1	7.82×10^{16}
^{133}Xe	1	1.20×10^{17}
^{131}I	0.75	1.95×10^{16}
^{134}Cs	0.75	2.09×10^{15}
^{137}Cs	0.75	1.58×10^{15}
^{132}Te	0.305	1.52×10^{16}
^{90}Sr	0.12	1.66×10^{12}
^{140}Ba	0.12	7.06×10^{13}
^{106}Ru	0.005	2.12×10^{11}

Table 8. Leakage Rates to The Environment

Elements	Half-life (hour)	Leakage Rates (Bq/m3)	Published Source Term[7]
^{85m}Kr	4.4	2.80×10^{16}	8.76×10^{16}
^{88}Kr	2804	7.82×10^{16}	5.51×10^{15}
^{133}Xe	127.2	1.20×10^{17}	2.43×10^{15}
^{131}I	192.0	4.23×10^{9}	7.10×10^{9}
^{134}Cs	17520.0	4.97×10^{6}	2.22×10^{8}
^{137}Cs	262800.0	2.50×10^{5}	1.40×10^{9}
^{132}Te	78.0	8.13×10^{9}	2.00×10^{10}
^{90}Sr	245280.0	2.81×10^{2}	1.86×10^{10}
^{140}Ba	307.2	9.58×10^{6}	2.56×10^{10}

Table 8 shows the leakage activity rates for all radionuclides into the environment. From the table, all of the noble gases' elements, such as Xe and Kr, that are released to the vessel would be fully released to the environment due to the noble gases' high volatility. Another major environmental release with high activity is the I, Cs, and Te elements, even though the activity release is significantly lower than Xe. Cs isotopes have lower leakage rates due to the half-life time of this element being significantly higher than I and Te. The other fission products, such as Sr and Ba element, has a significantly lower activity.

The estimated source term leakage rates were compared with the published source term at large break LOCA accident for SMART PWR-100 MWe calculated with ORIGEN-2 [7]. The results show that several of the estimated source terms have

dissimilar leakage rates due to different leakage rates calculation method used. The noble gases group, such as Kr and Xe, have a higher leakage rate than the published data, except for ^{85m}Kr, which has a slightly lower rate. The ^{131}I and ^{132}Te also had lower rates than the published data. In this research, the source terms are assumed to not be fully released in one moment but leaked hourly. The half-life time of the radionuclides would affect the leakage rates, and the radionuclides with a longer half-life time would have significantly lower leakage rates. The ^{134}Cs, ^{137}Cs, and ^{90}Sr had lower rates than the published data due to this condition.

4. CONCLUSION

A preliminary study for the source terms estimation code that was released to the environment was developed for radiological consequences study. This method could estimate source terms from the SMART reactor fuel inventory that would be leaked to the environment during severe accidents. The estimation was calculated from the fuel melting, releases to the vessel and containment, and leakage rates to the environment. Based on the estimation results, the noble gases elements such as Xe and Kr have a significantly higher leakage rate due to the noble gases' high volatility.

AUTHOR CONTRIBUTION

All of the authors are equally contributed as the main contributors of this paper. All authors read and approved the final version of the paper.

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