Uncertainty analysis of toxic gas leakage accident in cogeneration high temperature gas-cooled reactor

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Abstract
An uncertainty analysis method for control room habitability under toxic gas leakage accidents in cogeneration high temperature gas-cooled reactor (HTGR) is proposed to support risk-informed design of the plant. The method is applied to representative toxic gas leakage accidents in a hydrogen production plant by thermochemical Iodine-Sulfur water splitting method coupled to the HTTR gas turbine test plant. Variable parameters are successfully selected for the inputs to uncertainty propagation analysis by sensitivity analysis. Epistemic and aleatory uncertainties for each variable parameter are identified and are propagated using Latin hypercube sampling. The analyses show that the suggested method can successfully characterize and quantify uncertainties in the toxic gas concentration in control room. One important finding is that impact of uncertainty in surface roughness height on toxic gas concentration in control room is significant. The uncertainty is due largely to the simplification of the modeling of obstacles that exists between the reactor building and hydrogen production plant. The results lead us to the conclusion that toxic gas dispersion behavior analysis should combine two evaluation methods: dense gas dispersion model and computational fluid dynamics simulation.

Keywords: Uncertainty analysis, Toxic gas leakage accident, High temperature gas-cooled reactor, Risk-informed design, Hydrogen production

1. Introduction

High temperature gas-cooled Reactor (HTGR) is expected to extend the use of nuclear heat to a wider spectrum of industrial applications such as hydrogen production, process heat supply, due largely to high temperature heat supply capability as well as inherently safe characteristics. Towards the realization of the HTGR process heat application, a safety design of coupling an industrial plant to a nuclear reactor should be established. One important consideration for the design is toxic gas leakages from the industrial plant. The gas would spread to atmosphere by a leakage or a failure in the piping and equipment of industrial plant. The released gas may flow into the control room in the reactor building through ventilation systems and affect its habitability. A safe distance between the reactor building and industrial plant is required to prevent undue concentration increase of toxic gas in the control room. Traditionally, deterministic approaches are used to evaluate the adequacy of the distance with a worst leakage scenario for control room habitability, however, such approach may result in an unreasonable plant design.

With the aim of establishing a probabilistic approach for assessment of toxic gas leakage accidents in an industrial plant to support risk-informed design of cogeneration HTGR, the present study focusses on development of an uncertainty analysis method for toxic gas concentration in a control room.

The first section of this work explains our proposal for the PRA framework for control room habitability against toxic gas leakage accidents in an industrial plant. The following section describes a detail of uncertainty analysis methodology for toxic gas concentration assessment of a control room. The last section provides an application of the uncertainty analysis method to representative toxic gas leakage accidents in a hydrogen production plant by thermochemical Iodine-Sulfur water splitting method coupled to the HTTR gas turbine test plant (HTTR-GT/H2 plant) (Yan et al., 2017).
2. PRA framework for toxic gas leakage accident in industrial plant

We define sum of the product of the mean frequencies of accident sequences which leads to loss of control room habitability as risk metrics. The frequency of each accident sequence $F_{SF}$ can be assessed by the following expression.

$$F_{SF} = F_{IE} \cdot F_{MS} \cdot F_{CEF}$$

Here, $F_{IE}$ is initiating event frequency, $F_{MS}$ is mitigation system failure probability and $F_{CEF}$ is probability of exceeding evaluation criteria e.g. IDLH limits, etc. for control room habitability.

Figure 1 shows the task flow of control room habitability PRA. The overview of each step is as follows:

**Plant information review**

Define the reactor system and industrial plant of interest and clarify the system boundary. Review the information required to develop PRA model such as plant basic specifications, system configuration, plant layout, etc.

**Initiating event analysis:**

Identify initiating events which may leads to release of toxic gases from an industrial plant to the environment. Perform binning of the initiating events for efficient assessment and select a representative event in the group. The grouping should be such that events in the group have similar impact on control room habitability.

**Success criteria development:**

Identify a mitigation system and/or a combination of mitigation systems which are required to provide safety of personnel in the control room and assure the operability of reactor.

**Accident sequence analysis:**

Develop event trees to analyze accident sequence for toxic gas release events. Set headings based on the developed
success criteria.

System analysis:
Develop fault trees for systems corresponding to the headings in the event trees. The top event of the fault tree should be set based on the success criteria.

Data analysis:
Collect general reliability data from recognized sources. Develop a set of parameters, e.g. probabilities of basic events, component or system unavailabilities due to maintenance or repair, etc. required for accident sequence quantification. The frequency of each initiating events and/or initiating group should be also assessed.

Accident sequence quantification:
Quantify the frequency of accident sequence. Analyze uncertainties in the accident sequence quantification.

Release analysis:
Evaluate mass flow rate with duration time or released mass of toxic gas depending on release modes such as transient outflows and instantaneous releases. The assessment should consider phenomena relevant to the releases, e.g. flashing, evaporation, etc.

Dispersion analysis:
Evaluate dispersion behavior of toxic gas released from industrial plant to the environment for modeled accident sequences. The analysis should incorporate dense gas characteristics because toxic gases in the industrial plant generally have higher density than atmospheric air. Uncertainties in the dispersion analysis should also evaluated.

Control room concentration analysis:
Evaluate toxic gas concentration in control room. The analysis should consider closed loop operation in the control room ventilation system due to detection of toxic gas leakages, control room leakages and filtration of toxic gases. Uncertainties in the evaluation are needed to be considered. Assess the probability of exceeding toxic gas concentration limit.

Risk integration:
Evaluate integrated risk using the risk metrics for the control room habitability by Eq.1 using the frequencies obtained in aforementioned steps. Identify risk significant contributors by evaluating risk importance metrics.

3. Uncertainty analysis for toxic gas concentration in control room

The following section provides a methodology for uncertainty analysis for toxic gas concentration in the control room. The application to a representative cogeneration HTGR is also described.

3.1 Methodology
Table 1 provides summary of the uncertainty analysis process. The detail description follows the table.

Step 1: Uncertain parameter identification
This step identifies uncertain parameters influencing toxic gas concentration in a control room. Firstly, elements in release and transport pathways of toxic gases from industrial plant to control room should be investigated. Plausible phenomena in each element are then identified. Finally, uncertain parameters which corresponds to the identified phenomena are determined. Systematic approach e.g. Master Logic Diagram (MLD), etc. should be used to ensure comprehensiveness and transparency of the process.
Table 1 Procedure of uncertainty analysis for toxic gas concentration in control room.

| Step | Description                             |
|------|-----------------------------------------|
| 1    | Uncertain parameter identification       |
| 2    | Variable parameter derivation           |
| 3    | Uncertainty identification               |
| 4    | Sensitivity analysis                     |
| 5    | Uncertainty propagation analysis         |
| 6    | Uncertainty analysis result assessment   |

Step 2: Variable parameter derivation

Variable parameters which represent uncertain parameters are selected from input data of calculation methods such as empirical expression, and simulation codes.

Step 3: Uncertainty identification

Key sources of uncertainty in variable parameters should be investigated based on results of literature survey and knowledge of analyst. The epistemic and aleatory uncertainties should be separately identified in order to clarify contributions from each type of uncertainty in the results. Estimate probability distribution for each variable parameter based on various sources such as experimental data, detail simulation results, information from literature, etc.

Step 4: Sensitivity analysis

Perform sensitivity analysis for screening of variable parameters. The ranges of parameter variations are determined based on interval of uncertainty estimated in Step 3.

Step 5: Uncertainty propagation analysis

Develop a dataset for variable parameters by a random sampling method or an equivalent method based on probability density functions estimated in Step 3. Perform uncertainty propagation analysis using the dataset.

Step 6: Uncertainty analysis results assessment

Assess average and interval values of assessment results. The contribution of variable parameters to the results should be also analyzed.

3.2 Application

This section provides the results of application of the method to gas leakages of sulfuric dioxide (SO₂) and Iodine (I₂) from the IS hydrogen production plant in HTTR-GT/H₂ plant.

Step 1: Uncertain parameter identification

An MLD is developed starting with a top event of “operator poisoning” which is the safety issue regarding the toxic gas leakage accident. Fig. 2 shows release and transport pathways of toxic gas from the hydrogen production plant to control room. The following five elements are identified for the pathway;

(1) Leakage from hydrogen production plant to environment
(2) Dispersion in environment
(3) Inflow to control room
(4) Circulation in control room ventilation system
(5) Leakage in from environment to control room

Second level of the MLD corresponds to the identified pathways as shown in Fig.3. The elements are decomposed into contributing phenomena. For example, the pathway #1 is subdivided into three phenomena; jet flow, flashing, pool evaporation, depending on process conditions.

As a result of identification, nine uncertainty parameters are obtained.
Step 2: Variable parameter derivation

Input data of methods to evaluate each uncertainty parameters are investigated. Regarding the assessment of release quantity, flash rate and evaporation rate, expressions suggested in the guideline of disaster phenomena evaluation model issued by Fire and Disaster Management Agency in Japan is used. As for the evaluation of concentration at intake of control room ventilation system, SLAB code (Ermak, 1990), an atmospheric dispersion model for denser-than-air gas, is applied. The control room concentration is analyzed by solving an unsteady mass balance equation for lumped parameter system simulating the control room ventilation system. The remaining uncertainty parameters, i.e. toxic gas removal rate and amount of control room leakage, are used for calculation conditions in the equation. Removal of toxic gases by filter is not considered in the present simulation. The investigation results in identification of thirteen variable parameters (cf. Tables 2 and 3). Meteorological parameters are also extracted for the variable parameters, however, the present analysis did not consider uncertainty of those because the contributions are well studied in the field of radionuclide release for nuclear power plant accidents.

Table 2 Variable parameters and input parameters for uncertainty analysis of SO$_2$ leakage accident.

| ID | Variable parameters                              | Mean values | Standard deviations (2σ) |
|----|--------------------------------------------------|-------------|--------------------------|
| 1  | Discharge coefficient                            | 0.75        | 0.12                     |
| 2  | Exit cross sectional area [m$^2$]                | 1.2$\times$10$^{-3}$ | 4.6$\times$10$^{-5}$   |
| 3  | Operating pressure [MPa]                         | 5.0         | 0.018                    |
| 4  | Gas molar weight [kg/mol]                        | 6.4$\times$10$^{-2}$ | 4.3$\times$10$^{-6}$   |
| 5  | Operating temperature [K]                        | 1032        | 12                      |
| 6  | Gas heat capacity ratio                          | 1.2         | 0.040                    |
| 7  | Gas constant [J/mol K]                           | 8.31        | 4.6$\times$10$^{-7}$    |
| 8  | Leakage point elevation [m]                      | 3.0         | 0.041                    |
| 9  | Surface roughness height [m]                     | 0.55        | 0.23                     |
| 10 | Control room volume [m$^3$]                      | 747         | 0.95                     |
| 11 | Flow rate of control room ventilation system [m$^3$/h] | 300         | 14                       |
| 12 | Circulation flow rate of control room ventilation system [m$^3$/h] | 10          | 0.47                     |
| 13 | Control room leakage rate [1/h]                  | 0.038       | 0.011                    |

Step 3: Uncertainty identification

Epistemic and aleatory uncertainties for each variable parameter are identified as shown in Tables 2 and 3. The sources of aleatory uncertainty are natural variability of physical properties and meteorological conditions. The epistemic uncertainty can be subdivided into uncertainty due to model simplification, lack of data acquisition and
incompleteness. Key sources of the uncertainties are simplification of modeling for obstacles, manufacturing and installment accuracies, errors in instrumentation systems and operational variations. Normal distribution is assumed for probability density functions of each variable parameter. The basis to determine mean values and standard deviations are as follows:

Table 3 Variable parameters and input parameters for uncertainty analysis of $I_2$ leakage accident

| ID | Variable parameters                                      | Mean values | Standard deviations (2σ) |
|----|----------------------------------------------------------|-------------|--------------------------|
| 1  | Discharge coefficient                                    | 0.75        | 0.12                     |
| 2  | Exit cross sectional area [m$^2$]                        | $6.3\times10^{-2}$ | $3.3\times10^{-4}$     |
| 3  | Operating pressure [MPa]                                 | 5.0         | 0.018                    |
| 4  | Gas molar weight [kg/mol]                                | $2.5\times10^{-1}$ | $1.5\times10^{-4}$     |
| 5  | Operating temperature [K]                                | 464         | 5.5                      |
| 6  | Gas heat capacity ratio                                  | 1.3         | 0.046                    |
| 7  | Gas constant [J/mol K]                                   | 8.31        | $4.6\times10^{-7}$      |
| 8  | Leakage point elevation [m]                              | 2.2         | 0.03                     |
| 9  | Surface roughness height [m]                             | 0.55        | 0.23                     |
| 10 | Control room volume [m$^3$]                              | 747         | 0.95                     |
| 11 | Flow rate of control room ventilation system [m$^3$/h]   | 300         | 14                       |
| 12 | Circulation flow rate of control room ventilation system [m$^3$/h] | 10          | 0.47                     |
| 13 | Control room leakage rate [1/h]                          | 0.038       | 0.011                    |

- Discharge coefficient: Mean value is obtained by the evaluation guideline for jet release of gas (Fire Defense Agency, 2001). Standard deviation is estimated using data obtained from the literature (C. J. H. Van Den Bosh and R. A. P. M. Weterings, 2005).
- Exit cross sectional area: Design data of the HTTR-GT/H$_2$ plant is used for the mean value. Manufacturing accuracy for steel piping defined in Japanese Industrial Standard (JIS) is used for determining the standard deviation.
- Operating pressure and temperature: Operating condition for the HTTR-GT/H$_2$ plant is used for the mean value. Postulated variation in normal operation estimated in the HTTR design is applied for the standard deviation.
- Gas molar weight: Mean value and standard deviation are obtained by NIST database (NIST, 2015).
- Gas heat capacity ratio: Mean value and standard deviation are obtained by NIST database (NIST, 2018).
- Gas constant: Mean value and standard deviation are obtained by database of National Institute of Advanced Industrial Science and Technology (AIST) (AIST, 2018).
- Leakage point elevation: Mean value is obtained from the HTTR-GT/H$_2$ design. Standard deviation is estimated from the database of installment accuracies in HTTR.
- Surface roughness height: Mean value and standard deviation are determined based on SLAB code manual (Ermak, 1990).
- Control room volume: Mean value is obtained from the HTTR-GT/H$_2$ design. Standard deviation is determined based on Japanese Architectural Standard.
- Flow rate and circulation flow rate of control room ventilation system: Mean value is obtained from the HTTR-GT/H$_2$ design. Standard deviation is estimated based on the operational variation assumed in the HTTR design.
- Control room leakage rate: Mean value and standard deviation are estimated based on data listed in NRC guide (US NRC, 2001).

Step 4: Sensitivity analysis

Sensitivity analysis is performed for control room toxic gas concentration varying identified variable parameters using standard deviations. Ten seconds is assumed for the isolation time for control room ventilation system. The offset distance between leakage point and intake point of control room ventilation system is defined as 37m in accordance with layout design of HTTR-GT/H$_2$ plant. Meteorological parameters are set to the following values:

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- Ambient wind speed: 1.0 m/s
- Ambient temperature: 298 K
- Relative humidity: 72%
- Stability class: F (Stable)

A sensitivity factor $F_S$ defined in the following expression is used as an index to select parameters for further investigation.

$$F_S = \left| \frac{\delta C_{TX}}{C_{TX,ref}} \right|$$

Here, $\delta C_{TX}$ is deviation of maximum toxic gas concentration in control room and $C_{TX,ref}$ is maximum toxic gas concentration in control room using mean values for variable parameters.

Fig. 4 shows results of sensitivity analysis. Sensitivity factor of 1.0 is used for the criteria to screen variable parameters. The following parameters are selected for input parameters for uncertainty propagation analysis:

*Variable parameters correspond to ID number in Table 3*

- (a) SO$_2$ leakage
  - Discharge coefficient (ID#1)
  - Exit cross sectional area (ID#2)

*Variable parameters correspond to ID number in Table 3*

- (b) I$_2$ leakage

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**Fig.4 Sensitivity analysis results**
Step 5: Uncertainty propagation analysis

The Latin hypercube sampling (LHS) method is used to generate a dataset for selected variable parameters with the probabilistic distribution functions defined in Table 2. 100 cases are designed for uncertainty propagation analysis. Metrological parameters are the same with the conditions used in the sensitivity analysis.

Fig. 5 shows contours of SO\textsubscript{2} concentration in environment for the case using mean values for input parameters. Although SO\textsubscript{2} is heavier than air at room temperature, the plume of SO\textsubscript{2} moves upward as can be seen in Fig.5 because temperature of source material is extremely high. However, the direction of gas tends to flow down as the distance from leakage point increases. After a few ten seconds from completion of leakage, SO\textsubscript{2} is diluted by ambient air. The plume passes over the intake point of control room ventilation system.

(a) Contour during continuous leakage
(b) Contour after completion of leakage

Fig.5 Contours of SO\textsubscript{2} concentration in environment for the case using mean values for input parameters.

Fig. 6 depicts toxic gas concentrations in the control room for the case with mean values. Plume of toxic gas reaches the intake point of control room ventilation system approximately 30 seconds after the initiation of release. On the other hand, isolation of the ventilation system is completed within 10 seconds. Hence, increase rate of toxic gas concentration in the control room is small because the gas only intrudes into the control room by in-leakage. The increase continues until completion of release. The peak concentrations of SO\textsubscript{2} and I\textsubscript{2} are 6.0×10\textsuperscript{-3} and 5.0×10\textsuperscript{-5} mg/m\textsuperscript{3} which is far below the IDLH limit.

Step 6: Uncertainty analysis result assessment

Fig.7 shows histograms for distribution of uncertainty propagation analysis results. The analysis for peak concentrations of SO\textsubscript{2} and I\textsubscript{2} results in non-normal distribution with median values of 2.3×10\textsuperscript{-3} and 4.2×10\textsuperscript{-4}. Maximum values of the distributions are 6.4 and 1.4×10\textsuperscript{-1}, respectively. The inter-quartile ranges of distributions are 1.8×10\textsuperscript{-1} and 1.8×10\textsuperscript{-2}. The results show that variations of the toxic gas concentrations are significantly large.
Fig. 8 shows regression coefficients for variable parameters. The trends of the correlations obtained by multiple linear regression analysis are similar to sensitivity analysis results. (cf. Fig.4). The results clearly show that the surface roughness height (ID number 9) is the dominant input parameter for uncertainty of toxic gas concentration in the control room. The regression coefficients of remaining parameters are below 0.5, and therefore it can be concluded that the correlations between input and output uncertainties are weak.

Fig. 9 depicts SO₂ concentration contour in the environment for the case with maximum concentration at the intake point of control room ventilation system during continuous release. The surface roughness height for the case is approximately two times larger than mean value. It is clear that SO₂ concentration at the intake point for the present case is considerably higher than the case using mean values.

The results of study demonstrated that the suggested methodology can characterize and quantify uncertainty in toxic gas concentration in the control room.

### 3.3 Discussion

The focus of our study is to establish an uncertainty analysis method for toxic gas leakage accidents in an industrial plant to support risk-informed design of cogeneration HTGR. We suggest an uncertainty analysis method for toxic gas concentration in a control room. and applied to a SO₂ and I₂ gas leakage accidents in a hydrogen production plant in the
HTTR-GT/H₂ plant. It was found that reduction of uncertainty in surface roughness height is needed to improve the reliability of risk for toxic gas leakage accidents. The surface roughness height, the height above a surface at which the wind profile is assumed to be zero, is introduced in the code to characterize surface resistance to wind depending on surface geometry. The parameter is generally estimated using measurement data of wind profile obtained by field experiments under the circumstance of postulated surface conditions. The key source of uncertainty in the parameter is “simplification of modeling for obstacles” which can be classified into an epistemic uncertainty. Therefore, the uncertainty potentially has a room to be reduced. In reality, however, the surface conditions between an industrial plant and reactor building in a HTGR cogeneration plant cannot be fully modeled in the experiments. Based on the knowledge obtained in the present study, we came to a conclusion that toxic gas dispersion behavior analysis should combine two evaluation methods: dense gas dispersion model such as SLAB code and computational fluid dynamics (CFD) simulation. CFD tool should be applied to risk significant sequences (RSSs) in order to improve reliability of assessment results as well as secure practicability in the context of computational time.
4. Conclusion

With the aim of establishing a probabilistic approach for assessment of toxic gas leakage accidents in an industrial plant to support risk-informed design of a cogeneration HTGR, the present study suggests an uncertainty analysis method for toxic gas concentration in a control room. The method is then applied to SO_2 and I_2 gas leakage accidents in a hydrogen production plant in the HTTR-GT/H_2 plant. The results obtained in the application are summarized as follows:

• Variable parameters are successfully selected for the inputs to uncertainty propagation analysis by sensitivity analysis.
• Non-normal distribution is obtained for the output of uncertainty analysis. The variation of toxic gas concentration in control room is significantly large.
• Dominant contributor for the uncertainty of toxic gas concentration in control room is surface roughness height.
• Toxic gas dispersion behavior analysis should combine two evaluation methods: dense gas dispersion model such as SLAB code and CFD simulation.
• CFD tool should be applied to RSSs.

The approach presented in this study comply with technical requirements in ASME/ANS PRA standard for advanced Non-LWRs that is expected to be accepted by US NRC. Hence, it is expected to be directly applied to identification of significant accident sequence for licensing of cogeneration HTGR.

A further direction of the study is to develop a procedure for incorporating bifurcation of two dispersion analysis methods between RSSs and non-RSSs. In addition, validation of CFD tools for toxic gas dispersion simulations considering complex surface conditions should be performed. Furthermore, design considerations such as layout optimization of obstacles between a reactor building and an industrial plant is recommended to be performed using the developed method.

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References

AIST, Thermophysical database system (2018), (online) available from <https://tpds.db.aist.go.jp/tpds-web/statistics/metastatistics.aspx> (accessed on November 26, 2018).
C. J. H. Van Den Bosh, R. A. P. M. Weterings, TNO Yellow Book, Methods for the calculation of physical effects (2005) Ministerie van Verkeer en Wasterstaat.
Ermak, D.L., Users Manual for SLAB: An Atmospheric Dispersion Model for Denser-than-air Releases (1990) UCRL-MA-10.
Fire Defense Agency, Guideline in disaster prevention assessment for petrochemical complexes (2001)
NIST, NIST Chemistry WebBook (2018) (online), available from <https://webbook.nist.gov/chemistry/> (accessed on November 26, 2018).
NIST, Atomic Weights and Isotopic Compositions with Relative Atomic Masses (2015) (online) available from <https://www.nist.gov/pml/atomic-weights-and-isotopic-compositions-relative-atomic-masses> (accessed on November 26, 2018).
US NRC, Evaluating the Habitability of a Nuclear Power Plant Control Room during a Postulated Hazardous Chemical Release (2001), Reg. Guide. 1.78, Rev.1.
Yan, X.L., Sato, H., Sumita, J., Nomoto, Y., Horii, S., Imai, Y., Kasahara, S., Suzuki, K., Iwatsuki, J., Terada, A., Tachibana, Y., Oono, M., Yamada, S., Suyama, K., Design of HTTR-GT/H_2 test plant, Nucl. Eng. Des. Vol. 329, (2017), p.223–233.