Emergency evaluation model for nuclear criticality accidents of uranium enrichment facility

Linsheng Jia¹, Yapeng Yang, Renze Wang, Zongyang Feng, Ning Wang and Boning Liang
China Institute for Radiation Protection, Taiyuan, China

¹E-mail: jialinsheng2008@126.com

Abstract. Accident emergency evaluation technology is an important part of emergency preparedness and response for nuclear critical accidents. Moreover, emergency management also requires that facility with potential nuclear criticality accidents should have the emergency evaluation capability of the accident, so it can provide auxiliary decision support for early emergency decision-making. The paper introduces the emergency evaluation model for nuclear criticality accident of uranium enrichment facilities, including criticality fission number estimation, released source term estimation and dose estimation. The potential nuclear critical accident scenario of uranium enrichment facilities considered in the paper is uranyl fluoride solution criticality. Solution criticality is a complex accident scenario that may involve multiple criticality problems. In the paper, the criticality fission number is estimated based on the critical radiation monitoring data, which can give the results quickly and meet the emergency requirements. Estimation of criticality fission number is a technical difficulty, which can directly reflect nuclear criticality accident scale and provide a theoretical basis for conservative protective actions.

1. Introduction
According to foreign public reports, 22 nuclear critical accidents have occurred in the process of nuclear fuel [1]. Of these, 21 cases occurred when fissile material was in solution or slurry and 1 case occurred when fissile material was in metal ingot. A total of nine people died, four were severely exposed and more than 25 were exposed to high doses. Therefore, although the probability of nuclear critical accident is very small, once it happens, the accident will do great harm and affect a wide range. Thereafter, corresponding protective measures need to take.

Nuclear criticality accident is the design benchmark accident of uranium enrichment facilities. Operators and management attach great importance to this accident, try to eliminate the possibility of this accident in terms of design and management, and at the same time, make good emergency preparation and response. Accident emergency assessment is an important part of emergency preparedness and response for a nuclear criticality accident. Emergency assessment for a nuclear criticality accident includes estimation of criticality fission number, instantaneous dose estimation and the radiation dose caused by criticality fission products.

Criticality accident scenario of uranium enrichment facility is uranyl fluoride solution criticality. Solution criticality is a complex accident scenario, which may involve the problem of many criticality impulses. In the paper, the criticality accident emergency assessment model for uranyl fluoride solution is introduced, and criticality fission number estimation method based on the accident process is proposed. Criticality fission number is an important factor in the emergency assessment of a nuclear
criticality accident, and is also one of the technical difficulties. It reflects the size and scale of nuclear criticality accident and directly affects the decision of emergency protection action. There are many methods for criticality fission number estimation, each with its own applicable conditions. During a nuclear criticality accident, the information obtained at the beginning is limited and the evaluation method is conservative. With time lapse of the accident, abundant information can be obtained, and the estimation method is changed accordingly and should be more accurate.

2. Nuclear criticality accident emergency assessment process
Nuclear criticality accident emergency assessment mainly includes: (1) Criticality fission number calculation; (2) Instantaneous dose estimation; (3) Estimation of radiation dose caused by criticality fission products. The process of nuclear criticality accident assessment in the paper is shown in Figure 1.

![Figure 1. The flow of nuclear criticality accident assessment.](image)

3. Criticality fission number calculation
There are many methods to estimate fission number for uranyl fluoride solution criticality accident. These methods are mainly based on the readings of the criticality γ alarm monitor, conservative estimation of the system scenario and three simple formulas. Different fission number estimation methods have their own advantages and disadvantages and applicable scenarios.

3.1. Based on the readings of the criticality γ alarm monitor
The method can realize real-time assessment and meet the emergency requirements, but the uncertainty is large. Figure 2 shows the curve of γ dose rate with time and distance for uranyl fluoride solution criticality system, uranium enrichment is 4.95%, the ratio of hydrogen to uranium is 500, and criticality fission number is $10^{17}$, which was given in the literature NUREG/CR-6504 [2]. Based on the figure, the criticality fission number can be estimated from the actual γ dose rate meter readings of the facility. Firstly, the distance between the alarm monitor and the criticality accident point is determined, and the relationship between γ dose rate and time $10^{17}$ fission number under the distance is calculated. Then, the γ dose rate at a certain moment after the field accident is obtained and compared with the calculated relationship curve. Finally, the actual criticality fission number is estimated by extrapolation. Based on Figure 2, the paper fitted the formula for the relationship between γ dose rate and time $10^{17}$ fission number under different distances, as shown in Table 1. Interpolation calculation is required for other different distances.
Figure 2. The relation between $\gamma$ dose rate and time distance under the scenario of U(4.95)O$_2$F$_2$@H/$^{235}$U=410.

Table 1. Fitted formula of $\gamma$ dose rate and time at fission number $10^{17}$.

| $x$ (s) | $10^{10}$ | $10^{11}$ | $10^{12}$ | $10^{13}$ |
|---------|-----------|-----------|-----------|-----------|
| $<10$   | 225870/x | 346590/x  | 10630/x   | 8610/x    |
| $10$    | 20280/x  | 27110/x   | 955/x     | 753/x     |
| $100$   | 3019/x   | 4810/x    | 138/x     | 110/x     |
| $200$   | 682/x    | 999/x     | 33.1/x    | 28/x      |
| $500$   | 154/x    | 216/x     | 7/x       | 6.11/x    |
| $1000$  | 17/x     | 23.72/x   | 0.83/x    | 0.83/x    |

3.2. Based on system scenario conservative estimation
The method is simple and easy to operate, and is based on the historical criticality fission number and experimental experience. Table 2 shows the first fission number and total fission number corresponding to different volumes under solution system criticality scenario as listed in literature RASCAL 4: Description of Models and Methods [3]. RASCAL is an acronym for Radiological Assessment System for Consequence Analysis. The fission number given in this table is too conservative and is generally used in the design phase, where the assumed criticality accident fission number is used to make design requirements.

Table 2. Fission number corresponding to different system scenarios.

| system scenario | first fission number | total fission number |
|-----------------|----------------------|----------------------|
| Solution volume<379L | $1\times10^{17}$ | $3\times10^{18}$ |
| Solution volume>379L | $1\times10^{18}$ | $3\times10^{19}$ |

3.3. Based on three simplified formulas
Based on uranyl fluoride solution criticality accident scenario, there are three methods to estimate the criticality fission number conservatively. They are respectively Tuck equation, Nomura & Okuno,
equation and Oslen equation. Among them, the Nomura & Okuno equation is derived theoretically, and the Oslen equation is based on the empirical formula given by CRAC experiment [4].

1) Tuck equation [4, 5]

\[ F = \text{V} \times 10^{17} \quad (1) \]

Where: \( V \) is the volume of fuel solution, and unit is L.
Applicable conditions: a) Density <1.2 g/cm\(^3\); b) Natural cooling; c) No condensation occurs during boiling.

2) Nomura & Okuno equation [4, 6]

   1) If not boiling

\[ F_1 = 2.6 \times \text{V} \times 10^{16} \quad (2) \]

   2) If boiling

\[ F_2 = 6 \times \text{V} \times 10^{16} \quad (3) \]

Where: \( V \) is the volume of fuel solution, and unit is L.
Applicable conditions: a) Density <1.85 g/cm\(^3\); b) Evaporation during boiling is less than 25%; c) Natural cooling; d) No condensation occurs during boiling.

3) Oslen equation [4, 7]

\[ F = 2.95 \times 10^{15} \text{V}^{0.82} + 3.2 \times 10^{18} (1 - t^{-0.15}) \quad (4) \]

Where: \( V \) is the volume of fuel solution in the instantaneous explosion stage (L), and \( t \) is the duration of the power slowly decline stage (s).
Applicable conditions: a) High or low enrichment; b) Diameter of the cylindrical container is 30~80 cm; c) Feeding rate of the solution was 97~1872 L/h.

3.4. Based on accident progress to estimate criticality fission number

Each criticality fission number estimation method has its own advantage. The paper suggests that the method based on the readings of the criticality \( \gamma \) alarm is preferred when fuel solution volume cannot be obtained. If fuel solution volume is obtained, other methods are used to estimate criticality fission number. Figure 3 depicts the comparison of the curves of other methods. F1 is Nomura & Okuno equation when solution is not boiling, F2 is Nomura & Okuno equation when solution is boiling.

![Figure 3. Comparison of different estimation methods.](image-url)
Assuming that the volume calculation of the intersection points of Oslen equation curve with Tuck, F2 and F1 equation curve are respectively V1, V2 and V3, the conservative degree of different estimation methods is shown in Table 3.

**Table 3.** The conservative degree of different methods.

| No. | Volume | conservative degree comparison |
|-----|--------|--------------------------------|
| 1   | 0-V1   | Rascal > Olsen > Tuck > F2 > F1 |
| 2   | 30L- V2| Tuck > Rascal > Olsen > F2 > F1 |
| 3   | 30L- V2| Tuck > Rascal > F2 > Olsen > F1 |
| 4   | 30L- V3| Tuck > F2 > Rascal > Olsen > F1 |
| 5   | 30L- V3| Tuck > F2 > Rascal > F1 > Olsen |
| 6   | 30L- V3| Tuck > F2 > Rascal > F1 > Olsen |
| 7   | 30L- V3| Tuck > F2 > Rascal > F1 > Olsen |
| 8   | 30L- V3| Tuck > F2 > Rascal > F1 > Olsen |
| 9   | 30L- V3| Tuck > F2 > Rascal > F1 > Olsen |

Taking the fuel solution volume of 379-500L as an example, the criticality fission number estimation process can be summarized as follows in Figure 4.

![Criticality fission number estimation flow](image)

**Figure 4.** Criticality fission number estimation flow for uranyl fluoride solution with volume of 379-500L.

4. Estimation of radiation dose due to criticality fission product release

The formula for estimating the source term released to the environment is:

\[ S_i(n) = A_i(n) \times AF_i(n) \times RF_i(n) \times LF_i(n) \]  

where:
\[ S_i(n) \text{- Release amount of nuclide } i \text{ in the } n \text{ time step}; \]
\[ A_i(n) \text{- Inventory of nuclide } i \text{ in the } n \text{ time step}; \]
\[ AF_i(n) \text{- Airborne release fraction of nuclide } i \text{ in the } n \text{ time step}; \]
\[ RF_i(n) \text{- Reduction factor of nuclide } i \text{ in the } n \text{ time step}; \]
\[ LF_i(n) \text{- Leakage factor of nuclide } i \text{ in the } n \text{ time step}. \]

Table 4 lists initial inventory of fission products with a \(10^{19}\) fission number, which is given in *RASCAL 4* [3], and actual fission products are deduced through linear equal proportion.

| Nuclide   | Activity | Nuclide   | Activity |
|-----------|----------|-----------|----------|
| Kr-83m    | \(1.5 \times 10^2\) | I-131     | \(7.3 \times 10^0\) |
| Kr-85m    | \(8.9 \times 10^1\)  | I-132     | \(1.0 \times 10^3\) |
| Kr-85     | \(1.3 \times 10^{-5}\)| I-133     | \(1.7 \times 10^2\) |
| Kr-87     | \(1.1 \times 10^3\)  | I-134     | \(4.2 \times 10^3\) |
| Kr-88     | \(6.6 \times 10^2\)  | I-135     | \(5.0 \times 10^2\) |
| Kr-89     | \(4.6 \times 10^4\)  | Sr-91     | \(3.2 \times 10^2\) |
| Xe-133m   | \(1.9 \times 10^{-2}\)| Sr-91     | \(1.2 \times 10^3\) |
| Xe-133    | \(2.7 \times 10^{-3}\)| Ru-106    | \(2.0 \times 10^2\) |
| Xe-135m   | \(3.3 \times 10^2\)  | Cs-137    | \(1.0 \times 10^2\) |
| Xe-135    | \(5.2 \times 10^0\)  | Ba-139    | \(2.4 \times 10^1\) |
| Xe-137    | \(2.4 \times 10^4\)  | Ba-140    | \(1.1 \times 10^1\) |
| Xe-138    | \(1.0 \times 10^4\)  | Ce-143    | \(1.0 \times 10^2\) |

The calculation flow of criticality fission product release source term to the environment is shown in the following Figure 5.

**Figure 5.** The calculation flow of criticality fission product release source term to the environment.
Gaussian plume diffusion model can be taken as the atmospheric diffusion model. The radiation dose due to criticality fission product release to the environment mainly includes three parts: organ committed equivalent dose (internal irradiation) caused by inhalation, external dose of surface irradiation and external dose of plume irradiation.

5. Instantaneous dose estimation

Criticality instantaneous dose includes instantaneous gamma dose and the instantaneous neutron dose. The formula for calculating instantaneous dose is recommended in EJ/T 988-96.

Semi-empirical formula for instantaneous gamma dose (unshielded) [8]:

$$D_{\gamma} = 2.1 \times 10^{-22} N d^{-2} e^{-3.4d}$$  \hspace{1cm} (6)

Where:
- $D_{\gamma}$: γ dose equivalent, Sv;
- $N$: Fission number;
- $d$: Distance from criticality point, Km.

Semi-empirical formula for instantaneous neutron dose (unshielded) [8]:

$$D_{n} = 7.0 \times 10^{-22} N d^{-2} e^{-5.2d}$$  \hspace{1cm} (7)

Where:
- $D_{n}$: Neutron dose equivalent, Sv;
- $N$: Fission number;
- $d$: Distance from criticality point, Km.

When there is shield, it is necessary to consider reduction factors of the interaction between gamma rays and neutrons with matter. The reduction factor estimation formula in the paper is established according to the relation curve between the reduction factors of γ and neutrons and the shielding thickness (see Figure 6) in the literature NUREG/CR-6504 [2].

Figure 6. The relation of reduction and shield thickness.

6. Conclusions

Aiming at the uranyl fluoride solution criticality accident, one of the basic accidents in the nuclear fuel cycle facility design, the paper introduces its emergency evaluation model, which includes criticality fission number calculation, instantaneous dose estimation, and estimation of radiation dose caused by criticality fission products. At the same time, the paper proposes based on accident progress to estimate criticality fission number, which can not only estimate conservatively in the case of limited information to provide auxiliary support for emergency decision makers, but also it can also choose a relatively accurate estimation method when information is abundant to reduce unnecessary protective measures. The method solves the practical application problem of criticality accident emergency evaluation and the difficult problem of which evaluation method the user chooses.
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