Update of the Nuclear Criticality Slide Rule: Review of the estimation of the number of fissions

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ABSTRACT

IRSN (France), LLNL (USA) and ORNL (USA) began a long-term collaboration effort in 2015 to update the nuclear criticality Slide Rule (https://ncsp.llnl.gov/analytical-methods/criticality-sliderule) for the emergency response to a nuclear criticality accident. The Slide Rule permits the estimation of neutron and gamma dose rates and integrated doses based upon estimated fission yields, as a function of distance from the fission source, and time after criticality accidents for different critical systems. This paper presents the review of the section "estimation of the number of fissions", named "fission yield" in the Slide Rule document. This estimation is important because the other information provided by the Slide Rule is directly proportional to the number of fissions. The previous models, based on Hansen and Barbry formulae, will be presented with their associated assumptions and limitations. Then, new proposals will be presented with a comparison to past criticality accidents and experimental data. The new formulae, based on the heat energy equation, cover many types of criticality accidents (solution, dry or low-moderated powder, rods/assemblies in water and dry metal) and require limited information in an emergency situation.

***Key Words*: Criticality accident, Slide Rule, number of fissions**

1 INTRODUCTION

In 1997 Oak Ridge National Laboratory published the report "An Updated Nuclear Criticality Slide Rule" [1-2] as a tool for emergency response to a nuclear criticality accident. A similar document was produced by the Institut de Radioprotection et de Sûreté Nucléaire in 2000 [3]. According to [1], this kind of document "permits continued updating of information during the evolution of emergency response, including exposure information about accident victims, estimates of potential exposures to emergency response re-entry personnel, estimates of future radiation field magnitudes, and number of fissions (fission yield) estimate" without precisely knowing the initial conditions leading to the criticality accident. This document gives order of magnitude estimates of key parameters, useful for emergency response teams and public authorities.

This paper presents the update of the section "estimation of the number of fissions", named "fission yield" in the Slide Rule document. This estimation is important because the other information provided by the Slide Rule is directly proportional to the number of fissions. The previous models, based on Hansen [4] and Barbry [5] formulae, will be presented with their associated assumptions and limitations. Then, new proposals will be presented with a comparison to past criticality accidents and experimental data.

2 PRESENTATION OF SECTION "ESTIMATION OF THE NUMBER OF FISSIONS"

In the Functional Slide Rule [2], slide 6 and the page after are the operational documents used to estimate the number of fission. The §2.2 of reference [2] explains how to use this slide whereas §6 and §7 of reference [1] present the models and comparison with measurements. The models used are based on Hansen and Barbry formulae.

2.1 Hansen formula

The Hansen model is presented in [4] and is mainly used to estimate the First Pulse Fission Yield. It is based on an equation relating the probability of initiating the first persistent reaction as a function of time after the system reaches delayed criticality. The formula, giving the number of fissions of the first pulse, requires the following information:

- ramp reactivity insertion rate ($pcm.s^{-1}$),
- neutron lifetime (s),
- equivalent neutron source ($neutron.s^{-1}$),
- b = negative reactivity feedback of the system ($pcm.fission^{-1}$).

The document provides estimates for the three last parameters, based on information that must be provided by the user of the document (fissile concentration and volume). In particular, the "b" value as a function of system critical volume was based on some CRAC experiments. In the Slide Rule, the Hansen model is applied for the first spike and for Highly Enriched Uranium (HEU) or Low Enriched Uranium (LEU) solution only.

The main limitations noticed are first of all that the "b" value was empirically determined with only limited CRAC experiments, not necessarily representatives of all kind of solutions and configurations. Even with the CRAC experiments selected, the comparison of the estimation with the CRAC experiments results could lead to an underestimation, up to a factor 2. Then, the model is based on a weak neutron source assumption, which might not be applicable for plutonium systems. And finally, the model needs information about the kind of vessel (horizontal or vertical), the volume of solution, the fissile concentration and the addition rate of solution. It might be difficult to obtain all these parameters during the emergency, in particular the two last parameters.

2.2 Barbry formula

The Barbry formula is presented in [5] but not explicitly used (in particular, there is no operational slide). It is presented in the "conversion factors and equalities" section of the Functional Slide Rule [2] as an estimation of the total fission yield for a continuing aqueous solution criticality. It is an empirical equation that only needs the solution volume and the duration of criticality accident as parameters.

As presented before [6], the main limitations of the Barbry formula are linked to the fact that it is an empirical formula, so that the model is not applicable for conditions outside the conditions area defined by CRAC and SILENE experiments. In particular, the model is only applicable for a limited duration

(< 10 min) and without boiling of the solution. In addition, the formula does not give a bounding value in case of a premature stop of the accident/experiment.

2.3 Conclusion and objectives of the update

Based on the review performed of the Hansen and Barbry models, both models seem to be applicable only for solutions systems whereas the Slide Rule are supposed to cover several kinds of system. The formula only covers the first part of the criticality accident (first spike for Hansen model, less than 10 minutes and before boiling for Barbry model). The Hansen model requires several pieces of information to be used (in particular the addition rate of solution) and needs a weak neutron source assumption. Consequently the update of the section "estimation of the number of fissions" will need to:

- expand the kind of system (solution, powder or rods in water, metal),
- estimate the total number of fissions for the entire criticality accident duration (including boiling for system with water),
- require limited information because information from the field in case of an emergency won't be very precise,
- provide bounding deterministic approach.

3 UPDATE OF THE SECTION "ESTIMATION OF THE NUMBER OF FISSIONS"

The estimation of the number of fissions is an important issue in criticality accident studies or in case of a criticality accident emergency. Because of the interest, an ISO standard, ISO 16117, was published in 2013 to provide a methodology to estimate a reasonably maximal value of the total number of fissions of a postulated criticality accident. This standard, which was not for emergency response purpose, includes valuable information about the estimation of the maximal number of fissions: it is dedicated to the consequences of a criticality accident, and not to the detection of a criticality accident. In particular, annex D lists a wide range of formulae without making any recommendation. For the update of the section "estimation of the number of fissions", appropriate formulae were chosen in order to fulfill the objectives mentioned above.

Because of lessons learned from exercises or past criticality accidents, emergency response teams and users of the Slide Rule do not use it instantaneously as the criticality accident has occurred. A delay (many minutes or hours) occurs before the emergency organization is put in place and information arrives from the affected facility. So during this time, only limited information is available about the potential location and material characteristics of a criticality accident. That is why, the following update suggests two phases of the emergency response.

3.1 First phase of the emergency response

For all kinds of criticality accidents (solution, powder, rods/assemblies, metal), during the first phase of the emergency response, a value of 5×10^{18} fissions can be used in order to estimate a reasonably bounding value for the total number of fissions, taking into account the paucity of precise information about the accident. This value, which can be exceeded or far too bounding depending on the situation, seems reasonable. Based on lessons learned from past criticality accidents in nuclear fuel processing plants, it was exceeded for one criticality accident only ($\sim 4 \times 10^{19}$ fissions in about 20 min), which occurred with a HEU system in a very large vessel (800 liters of fissile solution), and seems unlikely to be met again. On the contrary, three criticality accidents exceeded 10^{18} fissions in about 20 min, 2 hours and 20 hours, respectively. In a more anecdotal way, the value of 5×10^{18} fissions corresponds to the maximal number of fissions obtained for CRAC and SILENE experiments (CRAC 18 experiment). This experiment lasted 6 hours, but a value close to 2.5×10^{18} fissions was already obtained after 1 hour.

This first estimate might be quickly updated as soon as possible during the second phase of the emergency response.

3.2 Second phase of the emergency response

Beyond the first phase of the emergency response, the estimation of the total number of fissions can be updated using specific documentation from the facility or formulae if information concerning the situation is available. The use of these formulae requires to know some input data, whose reliable/penalizing character must be analyzed. The suggested formulae are based on the "heat energy formula". A more detailed description of these formulae are given in appendix A with comparisons to experiments and past criticality accidents. For criticality accidents with liquid systems (solution or water), various formulae are suggested, depending whether boiling is reached or not. Nuclear Criticality Safety specialists can rely on possible temperature measurements of the process to determine if boiling has occurred or will take place. If not, boiling formula should be considered by default to estimate the total number of fissions. Formulae are presented for criticality accidents with fissile solution (§3.2.1), with a dry or low-moderated powder (§3.2.2), with rods/assemblies in water (§3.2.3) and with dry metal (§3.2.4).

3.2.1 Criticality accident with fissile solution

Boiling of the solution

With boiling of the solution, the total number of fissions (Nf), whatever the duration of the accident, is estimated with the following formula:

$$Nf = 1.3 \times 10^{16} . V . d_{sol} + 8 \times 10^{16} . \left(V - \check{V}_{critical(geom)}\right) \tag{1}$$

where V is the total volume of the solution (in liters), d_{sol} is the density of the solution (no unit) and $\check{V}_{critical(geom)}$ is the minimum critical volume of solution for the considered geometry (in liters). The minimum critical volume of solution can be obtained from standards or with a specific criticality calculation.

Without boiling of the solution

Without boiling of the solution, the total number of fissions, as a function of the duration of the accident, taken into account heat loss, is estimated with the following formula:

$$Nf = 1.3 \times 10^{16} . V . d_{sol} + 3.2 \times 10^{12} . h . S . t \tag{2}$$

where V is the total volume of the solution (in liters), d_{sol} is the density of the solution (no unit), h is the convection heat transfer coefficient (in $W.m^{-2}.°C^{-1}$), S is the heat transfer surface area (in m^2) and t is the duration of the criticality accident (in seconds). The recommended values for the convection heat transfer coefficient (h) are summarized in the following table.

Table I. Recommended values for the convection heat transfer coefficient (h)

Case		**h ($W.m^{-2}.°C^{-1}$)**
Free convection	Gases (air)	10
	Liquids	50 - 1000
Forced convection	Gases	25 - 250
	Liquids	100

For equipment surrounded by air (the most likely case), a value of 10 $W.m^{-2}.°C^{-1}$ for the convection heat transfer coefficient is recommended. For equipment surrounded by a cooling system (as for example the Tokai-mura criticality accident), a value of 100 $W.m^{-2}.°C^{-1}$ is recommended. Because the equipment can be considered as a simple geometry (sphere or cube), the following formulae are suggested in order to quickly calculate the heat transfer surface area S, keeping the previous units (S in m² and V in liters):

- for a sphere: $S = k.V^{\left(\frac{2}{3}\right)} = 4.836 \times 10^{-2}.V^{\left(\frac{2}{3}\right)}$
- for a cube: $S = k.V^{\left(\frac{2}{3}\right)} = 6 \times 10^{-2}.V^{\left(\frac{2}{3}\right)}$

The value of 6×10^{-2} for the parameter k is recommended for this formula (although it can be exceeded for geometries less compact than the cube or the orthocylinder).

3.2.2 Criticality accident with a dry or low-moderated powder

Boiling of water

With boiling of the water, the total number of fissions is estimated with the following formula, applicable for UO_2:

$$Nf = 1.2 \times 10^{16}.m_{water} + 8 \times 10^{16}.\left(m_{water} - \breve{m}_{water\_critical(geom)}\right) + 4 \times 10^{16}.m_{powder} \quad (3)$$

where m_{water} is the total mass of water (in kg), $\breve{m}_{water\_critical(geom)}$ is the minimum critical mass of water for the considered geometry (in kg) and m_{powder} is the total mass of UO_2 powder (in kg). The minimum critical mass of water for the considered geometry can be obtained from standards or with a specific criticality calculation.

Without boiling of water

Without boiling of water, the total number of fissions is estimated with the following formula, applicable for UO_2:

$$Nf = 1.2 \times 10^{16}.\left(m_{water} + 3.3 \times m_{powder}\right) \quad (4)$$

where m_{water} is the total mass of water (in kg) and m_{powder} is the total mass of UO_2 powder (in kg).

3.2.3 Criticality accident with rods/assemblies in water

The suggested formulae consider UO_2 rods (with Zircaloy cladding) in water. The following hypotheses are made, considering a UO_2 PWR "17x17" or "15x15" assembly:

- the UO_2 mass of an assembly is about 600 kg;
- the ratio between Zircaloy mass and UO_2 mass is about 0.17 (i.e. for each kg of UO_2, there is 0.17 kg of Zy).

Boiling of water

With boiling of the water, the total number of fissions is estimated with the following formula:

$$Nf = 1.2 \times 10^{16}.m_{water} + 8 \times 10^{16}.\left(m_{water} - \breve{m}_{water\_critical(geom)}\right) + 4 \times 10^{16}.m_{pellet} + 2.7 \times 10^{16}.m_{cladding} \quad (5)$$

where m_{water} is the total mass of water (in kg), $\breve{m}_{water\_critical(geom)}$ is the minimum critical mass of water for the considered geometry (in kg), m_{pellet} is the total mass of UO_2 pellet (in kg) and $m_{cladding}$ is the total mass of Zircaloy cladding (in kg). The minimum critical mass of water for the considered geometry can be obtained from standards or with a specific criticality calculation.

Without boiling of water

Without boiling of water, the total number of fissions is estimated with the following formula:

$$Nf = 1.2 \times 10^{16}.\left(m_{water} + 3.3 \times m_{pellet} + 2.2 \times m_{cladding}\right) \quad (6)$$

where m_{water} is the total mass of water (in kg), m_{pellet} is the total mass of UO_2 pellet (in kg) and $m_{cladding}$ is the total mass of Zircaloy cladding (in kg).

3.2.4 Criticality accident with dry metal

For this kind of criticality accident, the suggested formula takes into account the kind of dry medium, considered as metal systems (plutonium, uranium, alloy of uranium and molybdenum). Melting of the system is not considered*. The total number of fissions is estimated with the following formula:

$$Nf = 6 \times 10^{15}.k.m_{metal} \quad (7)$$

where m_{metal} is the total mass of metal (in kg) and k is a parameter depending of the kind of metal. The values for the parameter k are 1 for U-Mo systems, 0.77 for U systems and 0.5 for Pu systems.

4 CONCLUSIONS

This paper presents the update of the section “estimation of the number of fissions”, previsously based on Hansen [4] and Barbry [5] formulae. The new proposals expand the kind of system (solution, powder or rods in water, metal), estimate the total number of fissions for the entire criticality accident duration (including boiling for systems with water), require limited information, compatible with an emergency context, and provide bounding deterministic approach. The next step should be the development of other sections of the document such as slides regarding actions to stop an on-going criticality accident (for example, standards with neutron poison). The final task would be the development of a functional Slide Rule document based on all the previous work achieved since 2015, such as a document and/or an "application" for a handheld device (e.g. smartphone).

5 ACKNOWLEDGMENTS

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* It is considered that the entire fissile system reaches the melting temperature but without melting of the metal.

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APPENDIX A

Suggested formulae come from references [6] to [8]. They are based on the "heat energy formula". In the case of a liquid system without heat loss (including a phase before boiling and eventually a boiling phase), the two following formulae are used:

- phase before boiling: the number of fissions necessary to bring the system from its initial temperature (T_0) to the boiling temperature ($T_{boiling}$) is defined by:

$$Nf_1 = \varepsilon.m.C_p.\left[T_{boiling} - T_0\right] \tag{8}$$

where ε is the conversion factor between fission and Joule equal to 3.51×10^{10} fissions.J^{-1} (1 fission = 180 MeV of thermal energy), m is the total mass of solution (in kg), C_p is the specific heat (in $J.kg^{-1}.°C^{-1}$) (4,184 $J.kg^{-1}.°C^{-1}$ for water), $T_{boiling}$ is the boiling temperature (in °C) (110 °C for nitrate solution or 100 °C for water) and T_0 is the initial temperature of the solution (in °C) (arbitrary set to 20 °C),

- boiling phase: the number of fissions necessary to evaporate a given mass of water within the solution is defined by:

$$Nf_2 = \varepsilon.\Delta H_{vap}.\Delta m_{water} \tag{9}$$

where ε is the conversion factor between fission and Joule equal to 3.51×10^{10} fissions.J^{-1} (1 fission = 180 MeV of thermal energy), ΔH_{vap} is the vaporization enthalpy of the solution (2.26×10^6 $J.kg^{-1}$ for water) and Δm_{water} is the evaporated mass of water during the boiling (in kg).

If the boiling of the solution is reached, the total number of fissions is given by the following formula:

$$Nf = Nf_1 + Nf_2 = \varepsilon.m.C_p.\left[T_{boiling} - T_0\right] + \varepsilon.\Delta H_{vap}.\Delta m_{water} \tag{10}$$

Without boiling of the solution, the total number of fissions is given by the following formula:

$$Nf = Nf_1 = \varepsilon.m.C_p.\left[T_{boiling} - T_0\right] \tag{11}$$

The formulae (and associated assumptions) are then adapted to each kind of system and in order to take into account the heat loss (for solution systems). In particular, for the formulae with powder, rods and dry metal, the final temperatures are equal to the melting temperatures for theses media. For more information, one should read reference [6] for criticality accident with fissile solutions, reference [7] for criticality accident with powders or rods/assemblies within water and reference [8] for criticality accident with dry metals. These formulae present the advantage of estimating the total number of fissions for every kind of geometry and system. No information linked to the neutronic features of the system is necessary, because these formulae are based on thermal considerations. As a result, they could lead to important overestimations. On the other hand, despite the fact that the assumptions taken to determine these formulae are bounding, some phenomena (heat loss for formulae with powder, rods/assemblies in water, dry metal, absence of fusion for dry metal formula, no recondensation of the solution or the water during boiling) are not considered. Depending on the importance of these phenomena compared to other bounding assumptions considered, an underestimation of the total number of fissions is possible. The following figures and information illustrate the use of these formulae:

- for solutions, in comparison to CRAC 300 mm experiments (figure 1), CRAC 800 mm experiments (figure 2), SILENE experiments (figure 3), TRACY experiments (figure 4) and past criticality accidents (tables 1 and 2),
- for metal systems, in comparison to past criticality accidents (figure 5).

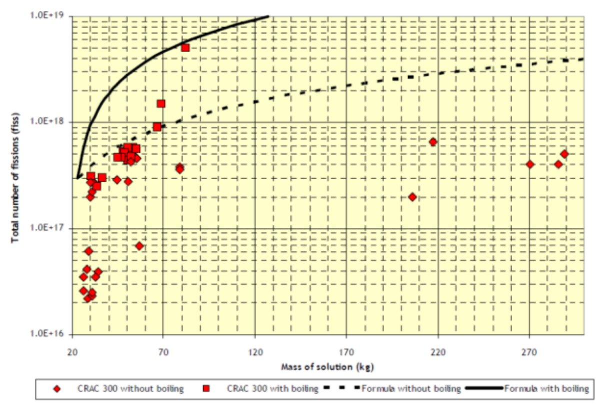

Figure 1. Comparison of formulae (1) and (2) to CRAC 300 mm experiments.

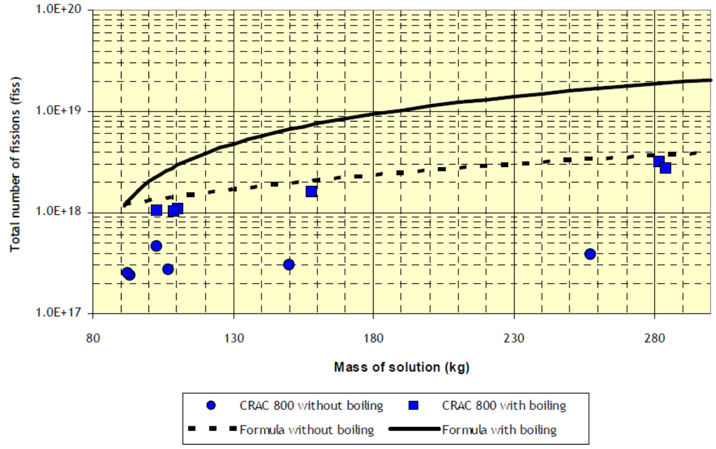

Figure 2. Comparison of formulae (1) and (2) to CRAC 800 mm experiments.

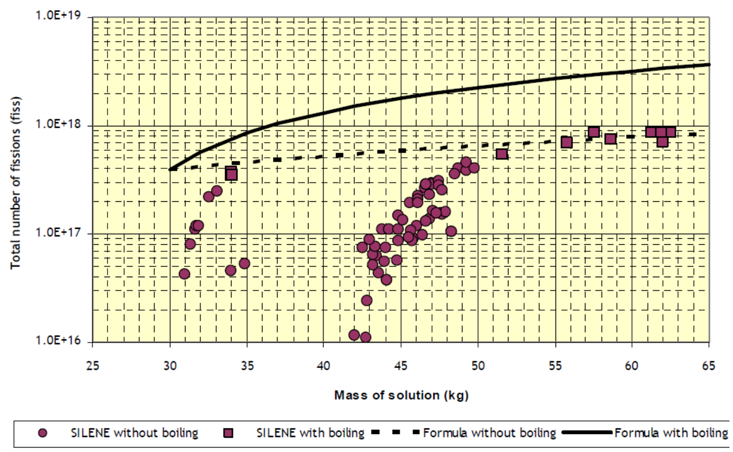

Figure 3. Comparison of formulae (1) and (2) to SILENE experiments.

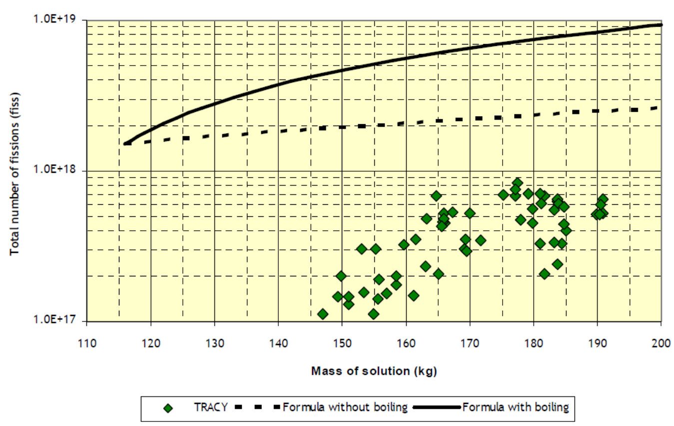

Figure 4. Comparison of formulae (1) and (2) to TRACY experiments.

No.	Site	Fissile media	Fuel Volume (litre)	Total U or Pu conc. (g/l)	Estimated mass of solution (kg)	Vessel diameter (cm)	Minimum critical mass of solution (kg)	Total fissions (fiss)	Estimated total fissions (fiss) without boiling	Estimated total fissions (fiss) with boiling	Duration
1	**Mayak**	Pu	31	27.5	**32.2**	40	24	2.0×10^{17}	4.2×10^{17}	-	< 1min
3	**Mayak**	U(90)	58.4	418	**91**	75	59	2.0×10^{17}	1.2×10^{18}	-	< 1min
4	**Y-12**	U(93)	56	40	**59**	55.2	34	1.3×10^{18}	$\mathbf{7.7\times10^{17}}$	2.8×10^{18}	20min
5	**LASL**	Pu	160	19.4 (*)	**164**	100	108	1.5×10^{17}	2.1×10^{18}	-	< 1min
6	**ICPP**	U(91)	800	42.5	**846**	-	*390*	4.0×10^{19}	$\mathbf{1.1\times10^{19}}$	4.7×10^{19}	20min
7	**Mayak**	Pu	19	47	**20.3**	34.8	21	2.5×10^{17}	2.6×10^{17}	-	1h50min
8	**ICPP**	U(90)	40	200	**51**	61	41	6.0×10^{17}	6.6×10^{17}	-	< 3min
10	**Hanford**	Pu	45	30.2	**47**	45.7	29	8.0×10^{17}	$\mathbf{6.1\times10^{17}}$	2.0×10^{18}	37.5h
11	**Mayak**	Pu	80	16.6	**82**	45	28	2.0×10^{17}	1.1×10^{18}	-	1h40min
12	**Tomsk**	U(90)	35.5	71	**39**	39	21	7.9×10^{17}	$\mathbf{5.1\times10^{17}}$	-	10h20min
13	**Tomsk**	U(90)	64.8	31.4	**67.8**	50	30	1.6×10^{16}	8.8×10^{17}	-	16h
14	**Wood River**	U(93)	51	55	**55**	45.8	26	1.3×10^{17}	7.1×10^{17}	-	1.5h
16	**Mayak**	U(90)	28.6	77	**31.6**	45	26	5.5×10^{17}	$\mathbf{4.1\times10^{17}}$	-	7h
17	**Mayak**	Pu	28.8	54.8 (*)	**31.1**	37.4	22.5	1.3×10^{17}	4.0×10^{17}	-	> 15min
18	**Windscale**	Pu	40	54.5 (*)	**43.2**	61	45	1.0×10^{15}	5.6×10^{17}	-	10s
19	**ICPP**	U(82)	315.5	23.5	**325**	61	41	2.7×10^{18}	4.2×10^{18}	-	1.5h
22	**Tokai-mura**	U(19)	45	370	**67.5**	45	*36*	2.5×10^{18}	$\mathbf{8.8\times10^{17}}$	-	19h40min

Table 1: Comparison of formulae (1) and (2) (without heat loss) to past criticality accidents.

No.	Site	Total fissions (fiss)	Estimated total fissions (fiss) without boiling	Estimated total fissions (fiss) without boiling (with heat loss)
12	Tomsk	7.9×10^{17}	5.1×10^{17}	1.3×10^{18}
16	Mayak	5.5×10^{17}	4.1×10^{17}	8.5×10^{17}
22	Tokai-mura	2.5×10^{18}	8.8×10^{17}	1.8×10^{19}

Table 2: Comparison of formula (2) to past criticality accidents (using heat loss formula for long duration accidents).

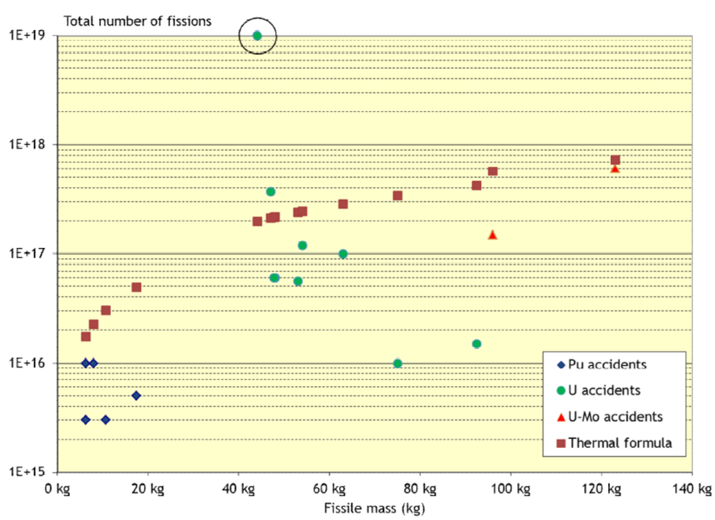

Figure 5. Comparison of formula (7) to past metal criticality accidents.