# Critical and Subcritical Parameters of the System Simulating Plutonium Metal Dissolution

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Dissolution of plutonium metal was simulated using the Monte Carlo computer code to calculate criticality safety limits for the process. Calculations were made for the constant masses of plutonium charged to the dissolving vessel considering distribution of plutonium in metal and solution phases. Critical parameters and limits were calculated as a function of dissolving vessel volume and plutonium metal mass. $^{240}$Pu content was assumed to be from 0% to 10% (mass). Critical parameters were evaluated for the system with a water reflector. Results of this paper may be used in the designing process equipment for plutonium metal dissolution.

***KEYWORDS: criticality safety, $K_{eff}$, plutonium metal dissolution, critical parameters of dissolving vessel, plutonium ingot, $^{240}$Pu content***

## 1. Introduction

One of the urgent tasks today consists in utilization of accumulated weapons grade plutonium stockpiles by using it as nuclear fuel for NPPs. This generates the need to dissolve a significant amount of plutonium metal for its purification from radiogenic impurities and further reprocessing.

The paper presents calculations of critical and subcritical parameters for dissolving vessel (further on – "dissolver") with spherical geometry and plutonium metal dissolution in nitric acid. Consideration was given to a two – isotope model of plutonium: $^{239}$Pu and $^{240}$Pu; the $^{240}$Pu mass fraction in the mixture varied from 0% to 10%. Dissolvers of big volumes (dangerous from the point of view of criticality safety) necessary to meet process requirements have been examined.

## 2. Criticality Safety Requirements

In accordance with Russian Criticality Safety Rules, when a dangerous equipment is in operation, the following requirements must be simultaneously met:[1)]

- Requirement «A». The $K_{eff}$ value of a isolated unit of equipment must not exceed 0.95 under the normal operation conditions.
- Requirement «B». The fissile material charge must not exceed the value of minimum critical mass for this piece of equipment reduced 2.1 times (2.1 is a fissile material mass margin).
- Requirement «C». The $K_{eff}$ value of this piece of equipment must not exceed 0.98 at any single failure of the equipment or any single human error (emergency conditions).

In this paper the dissolver repeat charging (double charge) due to a human error is considered as an emergency situation. The consequences of other emergency situations can only be considered when the real dissolver design is analyzed in view of its use in the technological process.

Three criticality safety requirements («A», «B», «C») indicated above must be complied with both in the process of plutonium metal preparation for dissolution (i.e. the plutonium metal ingot outside of dissolver must be taken into account), and during its dissolution (i.e. the dissolver must be considered with plutonium metal loaded into it). As no process operations on metal preparation for dissolution have been considered in this paper, it is reasonable to be restricted to requirements «A» and «B» as regards plutonium metal which will be charged into the dissolver. As far as the dissolver is concerned all the three criticality safety requirements indicated above must be considered for it. However it should be mentioned that if the dissolver design eliminates any possibility to exceed the allowable charge the compliance with requirement «A» will be sufficient in respect of this dissolver.

## 3. Justification of calculation model and technique

During calculations the most conservative from the point of view of criticality safety geometrical model was assumed when the dissolver in the form of sphere was surrounded with a full water reflector (with the thickness of 25 cm), and plutonium metal to be dissolved (in the form of an ingot) was located in its center. The Pu mass remained constant in the dissolver during the whole process of dissolution up to

the complete plutonium metal conversion into the solution. This approach corresponds to the assumed conservative concept because in the real dissolution process the Pu concentration in the solution is limited by the process requirements of the next reprocessing stage and part of the solution can be removed during the dissolution process and a fresh nitric acid solution can be added to the dissolver.

A two – zone model of dissolution has been considered, when plutonium metal in the form of sphere (part of the charged metal that hadn't been dissolved) was surrounded by plutonium solution with Pu concentration homogenious in the entire solution volume. Strictly speaking, this model does not completely agree with the reality because, on the one hand, the metal dissolution process can have a different rate on its surface, and on the other hand, the solution concentration close to metal is always somewhat higher than in the peripheral part of the solution. However these effects impact $K_{eff}$ insignificantly due to gas release during the dissolution process and the use of forced solution circulation. These facts result in rather intensive solution stirring (with regard to a duration of dissolution process).

The applicability of the calculation model chosen for the criticality safety analysis with the dissolver being loaded with several fragments of plutonium metal (with the constant total charge mass) will be considered below.

The complex of MMKFK-2 codes was used for calculations.[2] The use of this complex for calculation of plutonium critical assemblies in the solution resulted in overestimating $K_{eff}$ up to 1% within the H/Pu-239 moderation range from 125 to 1000.[3]

The methodological error of this complex was also estimated by the authors when they calculated benchmark spherical critical assemblies with plutonium metal and plutonium nitric solution.[4] The main parameters of these assemblies and calculation results are given in Table 1. The results obtained show that the complex[2] overestimates the $K_{eff}$ values calculating the assemblies with plutonium metal and plutonium nitric solution. So it can be used for justification of criticality safety systems which simulate plutonium metal dissolution.

**4.Calculation results.**

For the calculation of nuclear concentrations in plutonium nitric solution the dependence of solution density, d (g/cm$^3$), on Pu concentration, $C_{Pu}$ (g/l), given in the JAERI Nuclear Criticality Safety Handbook, was used.[5] At the solution temperature of 25°C and in the absence of free nitric acid this dependence has the form of the following equation:

$$d=0.99708+1.65625*10^{-3}*C_{Pu}-3.418*10^{-8}*C_{Pu}^2 \quad (1)$$

Theoretical density of plutonium metal was assumed equal to 19.816 g/cm$^3$.[5]

The values of maximum mass of plutonium metal which can be loaded to the dissolver (according to requirements «A» and «B») were determined in the following order:

- critical mass $M_{CR}$ was calculated for the sphere of plutonium metal with a full water reflector;
- a preliminary value of safe Pu mass was determined $M_{2.1} = M_{CR} / 2.1$;
- $^{2,1}K_{eff}$ was calculated for the sphere of plutonium metal with the mass of $M_{2.1}$ and with a full water reflector;
- with the value of $^{2,1}K_{eff} \leq 0.95$ the safe Pu mass, $M_S$, was assumed equal to $M_{2.1}$.

Table 1. Principal parameters of benchmark assemblies and calculation results.

| Assembly type | | Pu-MET-FAST-011 | Pu-SOL-THERM-001 |
|---|---|---|---|
| Pu concentration, g/cm$^3$ | | 19.74 | 0.096 |
| Radius of its active part, cm | | 4.1217 | 14.5377 |
| Nuclear concentration of Pu isotopies, 1/barn*cm | $^{238}$Pu | – | $1.4571*10^{-8}$ |
| | $^{239}$Pu | $4.6982*10^{-2}$ | $2.2977*10^{-4}$ |
| | $^{240}$Pu | $2.5852*10^{-3}$ | $1.1242*10^{-5}$ |
| | $^{241}$Pu | $1.4915*10^{-4}$ | $7.3148*10^{-7}$ |
| | $^{242}$Pu | $9.9432*10^{-6}$ | $2.1495*10^{-8}$ |
| Steel wall thickness, cm | | – | 0.1245 |
| Water reflector thickness, cm | | 25.4 | 30.0 |
| $K_{eff} \pm \sigma$ | | 1.0146±0.0005 | 1.0073±0.0005 |

The calculation results of safe plutonium metal mass are given in Table 2.

The dependence of dissolver $K_{eff}$ on Pu concentration in the solution at various values of dissolver volume (V) is given in Figure 1, with dissolution of $^{239}Pu$ ingot with the mass of 2.4 kg as an example. The RMS deviation ($\sigma$) $K_{eff}$ calculations did not exceed 0.0013, thus it is not shown in Figure 1.

The graphs of $K_{eff}$ as a function of Pu concentration in the solution at other parameters of the dissolver (its charge and $^{240}Pu$ content) have a similar shape and differ in the Pu concentration value at which the maximum $K_{eff}$ is achieved, and in the value of $K_{eff}$.

Table 2. Calculation results of safe plutonium metal mass

| Calculated parameter | % $^{240}Pu$ | | |
|---|---|---|---|
| | 0 | 5 | 10 |
| $M_{CR}$, kg | 5.038 | 5.372 | 5.679 |
| $M_{2,1}$, kg | 2.399 | 2.558 | 2.704 |
| $^{2,1}K_{eff}$ | 0.8063±0.0007 | 0.8058±0.0008 | 0.8065±0.0006 |
| $M_S$, kg | 2.40 | 2.56 | 2.70 |

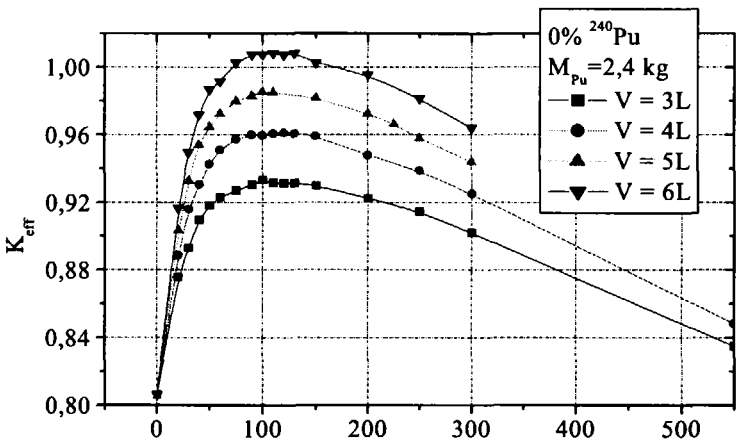


Fig.1. $K_{eff}$ of the dissolver as a function of Pu concentration in the solution and dissolver volume at 0% $^{240}Pu$.

## 5. Analysis of calculation results

The results of calculations made can be presented in the form of graphs of maximum $K_{eff}$ values as a function of dissolver volume at the given values of its charge ($M_{Pu}$) and $^{240}Pu$ content. Figure 2 shows these graphs for the case of $^{239}Pu$ dissolution (0%$^{240}Pu$).

Based on these graphs we can get the dependence of dissolver charge on its volume at the constant $K_{eff}$ values of the dissolver.

These functions are presented in figures 3, 4, 5 for $K_{eff}$ = 1.0; 0.98 and 0.95.

The following conclusions can be made as a result of the analysis of figures 3, 4 and 5 (as it was mentioned above, the dissolver double charge was considered as the emergency situation):

* When the charge norms for the dissolver with a dangerous volume are determined based on requirement «A» (dissolver $K_{eff}$ does not exceed 0.95), requirements «B» and «C» are not met.

For example, as graph $K_{eff}$ = 0,95 in Figure 4 shows, for the dissolver with the volume of 8.3 L the charge can be determined equal to 1.2 kg at 5% $^{240}Pu$. But in this case with double charge (2.4 kg) the value of dissolver $K_{eff}$ will exceed 1.0.

* When the charge norms of the dissolver with a dangerous volume are determined based on requirement «B» (the charge is 2.1 times lower than the minimum critical charge), requirements «A» and «C» are not met.

For example, as graph $K_{eff}$ = 1,0 in Figure 5 shows, for the dissolver with the volume of 11.1 L the charge can be determined equal to 1.1 kg at 10% $^{240}Pu$ (1.1 kg=2.31 kg / 2.1). But in this case with the normal charge (1.1 kg) the value of dissolver $K_{eff}$ will exceed 0.95, and with the double charge (2.2 kg) the value of its $K_{eff}$ will exceed 0.98. * When the charge norms for the dissolver with a dangerous volume is determined based on requirement «C» (the charge is twice as low as the charge at which $K_{eff}$ =0.98), requirements «A» and «B» are met.

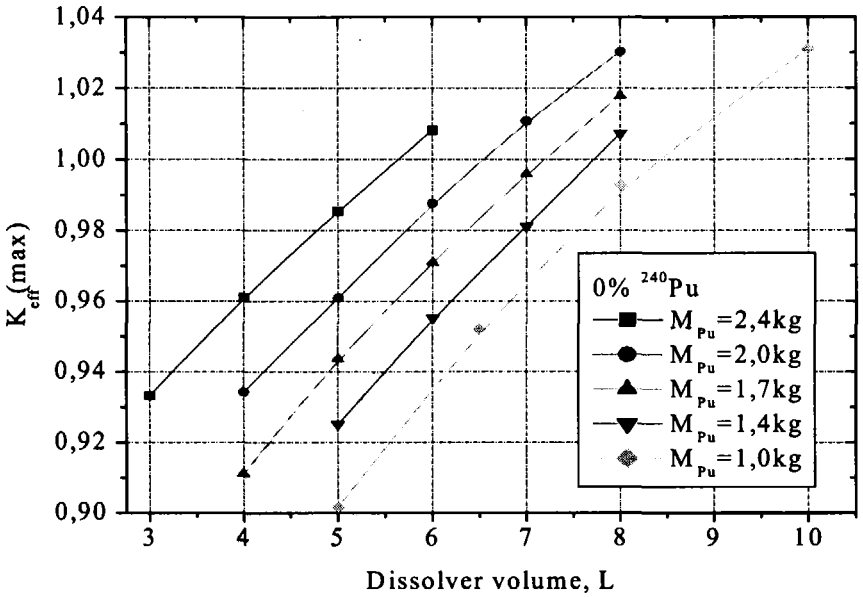


Fig.2. The maximum $K_{eff}$ value as a function of dissolver volume and its charge at 0% $^{240}$Pu.

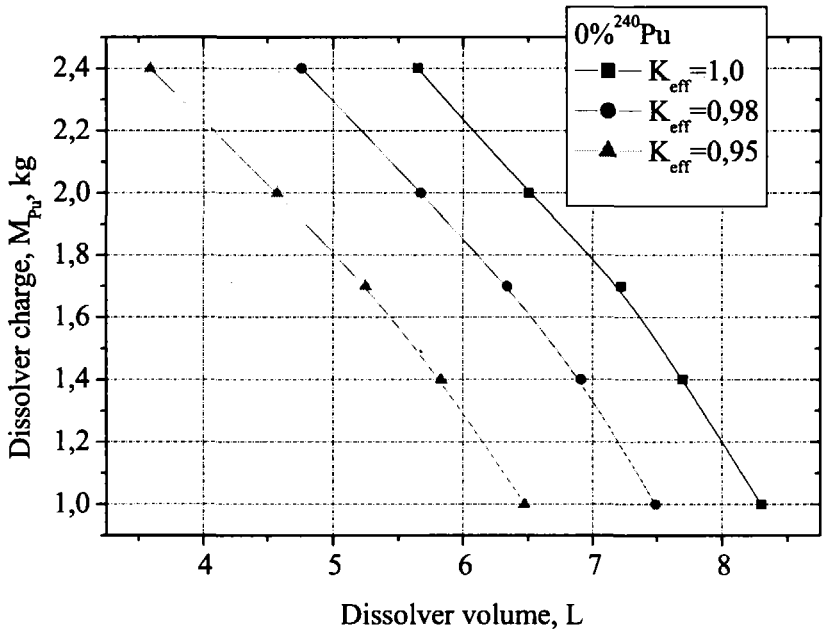


Fig.3. Dissolver charge as a function of its volume at 0% $^{240}$Pu.

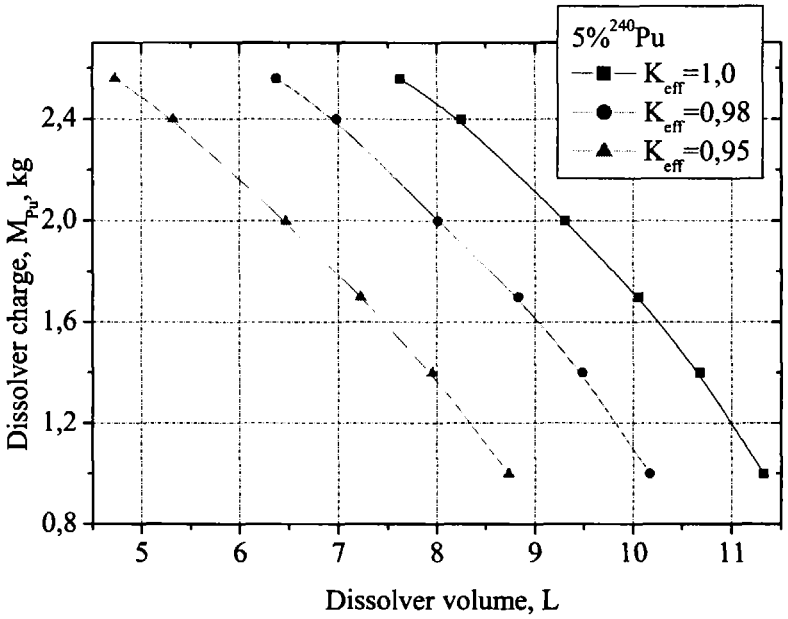


Fig.4. Dissolver charge as a functoin of its volume at 5% $^{240}$Pu.

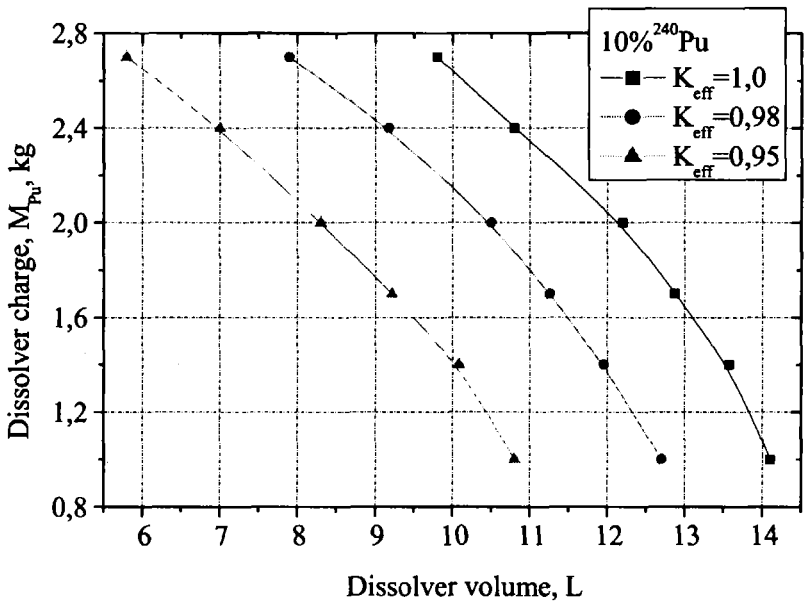


Fig.5. Dissolver charge as a function of its volume at 10% $^{240}$Pu.

For instance, as graph $K_{eff}$ = 0,98 in Figure 3 shows, for the dissolver with the volume of 5.65 L the charge can be determined equal to 1.0 kg at 0% $^{240}Pu$ (so that in case of the double charge equal to 2.0 kg $K_{eff}$ would not exceed 0.98). In this case, with the normal charge (1.0 kg) $K_{eff}$ does not exceed 0.95 (thus being in compliance with requirement «A»), and with the charge of 2.1 kg (2.1 kg=1.0 kg * 2.1) $K_{eff}$ will not exceed 1.0 (thus being in compliance with requirement «B»).

The conclusion that if requirement «C» is met, requirements «A» and «B» will be met simultaneously, is true only for the parameters of the dissiolver and metal under dissolution which have been considered in the given study.

For example, as graph $K_{eff}$ =0,98 in Figure 4 shows, for the dissolver with the volume of 9.0 L the charge norm is determined equal to 0.8 kg at 5% $^{240}Pu$ (in case of the double charge the value of dissolver $K_{eff}$ will be equal to 0.98), the maximum calculated value of $K_{eff}$ of this dissolver will be equal to 0.951 with the charge of 0.8 kg (i.e. requirement «A» is not met).

The applicability of the obtained results for the criticality safety analysis of the dissolver into which several fragments of plutonium metal are charged, was studied with dissolution of 2.4 kg of $^{239}Pu$ in the dissolver with the volume of 6 L as an example. Consideration was given to the dependence of $K_{eff}$ calculations for the case of dissolution of plutonium metal in the form of one fragment (ingot) can be used as preliminary results for the analysis of dissolution of several Pu fragments with the same total mass. In this case the main condition consists in limiting the volume in which plutonium metal fragments are located in the course of dissolution. In practice this condition can be met by means of the use of a charging basket of the adequate design.

## 6. Conclusion

Based on the calculation results and the analysis of $K_{eff}$ as a function of charge and volume of the dissolver during dissolution of plutonium metal with the mass content of $^{240}Pu$ from 0% to 10%, the following recommendations can be made:

1. Requirement «C» is the most conservative from the point of view of criticality safety when the dissolver charge is determined.
2. Based on requirement «A» a safe charge can be only determined with the special dissolver design which rules out the possibility to exceed the charge indicated for this dissolver.
3. The final decision about the safe charge size for the dissolver must be made in view of the analysis of the consequences of all possible emergency conditions, based on the condition of $K_{eff} \leq 0.98$.

Table 3.The effect of gaps between plutonium metal fragments on $K_{eff}$

| Interfragment gap, cm | 0 | 0.05 | 0.247 | 0.495 |
|---|---|---|---|---|
| $K_{eff} \pm \sigma$ | 1.0072±0.0007 | 1.0024±0.0012 | 0.9974±0.0011 | 0.9915±0.0011 |

dissolver $K_{eff}$ on the gap size between plutonium metal fragments at the Pu concentration in the solution which corresponds to the maximum value of $K_{eff}$ at dissolution of Pu in the form of one fragment (ingot) of the same mass. In accordance with figure 1 the maximum value of $K_{eff}$ was achieved at $C_{Pu}$=110 g/L with the dissolution of 2.4 kg of $^{239}Pu$ in the dissolver with the volume of 6 L. With this Pu concentration in the solution, the mass of metal part which had not been dissolved was equal to 1.75 kg. The following geometrical model was assumed for the calculations:

- the volume into which the fragments were located did not exceed 121.1 $cm^3$, thus corresponding to the volume of plutonium metal ingot with the mass of 2.4 kg;
- the number of fragments was assumed equal to eight, the fragments had the form of cubes of the same size determined by the total fragment mass (1.75 kg), which corresponded to the metal part which had not been dissolved.

The results of $K_{eff}$ calculation as a function of interfragment gap sizes are given in Table 3.

The indicated results show that the increase in the distance between plutonium metal fragments does not result in the growth of dissolver $K_{eff}$. So the results of

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