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Applicability of Simplified Methods to Evaluate Consequences of Criticality Accident Using Past Accident Data

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Applicability of four simplified methods to evaluate the consequences of criticality accident was investigated. Fissions in the initial burst and total fissions were evaluated using the simplified methods and those results were compared with the past accident data.

The simplified methods give the number of fissions in the initial burst as a function of solution volume; however the accident data did not show such tendency. This would be caused by the lack of accident data for the initial burst with high accuracy. For total fissions, simplified methods almost reproduced the upper envelope of the accidents. However several accidents, which were beyond the applicable conditions, resulted in the larger total fissions than the evaluations. In particular, the Tokai-mura accident in 1999 gave in the largest total specific fissions, because the activation of cooling system brought the relatively high power for a long time.

KEYWORDS: criticality accident, simplified method, accident evaluation, total fissions, fissions in the initial burst, duration

1. Introduction

Several simplified methods to evaluate the consequences of criticality accidents in fissile solutions were proposed in the 1970s to the mid 1990s[1-4]. Those methods are very useful to estimate the scale of the accident quickly using the limited information. Most of those methods are based on the experimental data using highly enriched uranium (HEU) solutions, such as CRAC[5], SILENE[6], and KEWB[7]. Although some of them were applied or verified for the past criticality accidents occurred in the US and UK, the number of the accidents was 8 or less at that time and it was insufficient to examine the applicability of those methods to the real accident.

In the late 1990s, the Russian federation disclosed the technical information about the past criticality accidents, and two accidents were happened in the Russian federation (1997) and in Japan (1999). Consequently, the number of the accident became 22, which is more than twice as that of earlier. Thus, we will examine the applicability of the proposed simplified methods using all the available data of past accidents in this study.

2. Criticality Accidents

Up to the present, twenty-two criticality accidents in nuclear fuel processing plants have been reported[8]. Table 1 shows the main characteristics of the accidents, which are quoted from Ref. 8. One of them was occurred in handling of plutonium metal. The other 21 accidents were occurred with solution or slurry systems. Total fission numbers were between 1×10^{15} and 4×10^{19}. Most of them (17 cases) resulted in 1×10^{16} to 3×10^{18} fissions. Figure 1 shows the chronological record of the accidents.

In the present paper, 21 accidents, except one with Pu metal, were evaluated using the simplified methods, because the proposed simplified methods were developed for solution systems.

3. Simplified Methods

We will investigate the applicability of four simplified methods, proposed by Tuck, Olsen, Barbry, and Nomura. In Table 2, the evaluation formulas and the applicable conditions of each method are described. The main features of each method are as follows.

3.1 Tuck's Method

This method is based on the past 6 accidents with fissile solutions, which were occurred in the US and UK until 1965, and experiments of CRAC and KEWB. The evaluated items are: maximum fissions in a 5-sec interval, maximum specific fission rate in a 5-sec interval, total fissions, maximum pressure, and average power during boiling. Different formula was used for uranium system and plutonium system, respectively, to evaluate the maximum fissions in a 5-sec interval. For the evaluation of total fissions, evaporation of all the solution with 20% heat loss is assumed.

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Table 1 Summary of criticality accidents in nuclear fuel processing plants

No.	Site[1]	Date	Fuel type	Fissile	Fuel Volume (l)	Vessel Volume (l)	Fissions in Iinitial Burst (fiss)	Specific Fissions in Initial Burst (fiss/liter)	Total Fissions (fiss)	Specific Total Fissions (fiss/liter)	Duration
1	Mayak	1953/3/15	Solution	Pu	31	40	unknown	unknown	2.0×10^{17}	6.5×10^{15}	1min[3)]
2	Mayak	1957/4/21	Slurry	U(90)	30	100	unknown	unknown	1.0×10^{17}	3.3×10^{15}	10min
3	Mayak	1958/1/2	Solution	U(90)	58.4	442	2.0×10^{17}	3.4×10^{15}	2.0×10^{17}	3.4×10^{15}	1min[3)]
4	Y-12	1958/6/16	Solution	U(93)	56	208	1.0×10^{16}	2.0×10^{14}	1.3×10^{18}	2.3×10^{16}	20min
5	LASL	1958/12/30	Solution(Org)	Pu	160	982	1.5×10^{17}	9.4×10^{14}	1.5×10^{17}	9.4×10^{14}	1min[3)]
6	ICPP	1959/10/16	Solution	U(91)	800	18900	1.0×10^{17}	1.0×10^{14}	4.0×10^{19}	5.0×10^{16}	20min
7	Mayak	1960/12/5	Solution	Pu	19	40	unknown	unknown	2.5×10^{17}	1.3×10^{16}	1h50min
8	ICPP	1961/1/25	Solution	U(90)	40	461	6.0×10^{16}	1.5×10^{15}	6.0×10^{17}	1.5×10^{16}	3min[4)]
9	Tomsk	1961/7/14	Solution(Org)	U(22.6)	42.9	65	none	none	1.2×10^{15}	2.8×10^{13}	1min[3)]
10	Hanford	1962/4/7	Solution	Pu	45	69	1.0×10^{16}	2.0×10^{14}	8.2×10^{17}	1.8×10^{16}	37.5h
11	Mayak	1962/9/7	Solution	Pu	80	100	none	none	2.0×10^{17}	2.5×10^{15}	1h40min
12	Tomsk	1963/1/30	Solution	U(90)	35.5	49.9	unknown	unknown	7.9×10^{17}	2.2×10^{16}	10h20min
13	Tomsk	1963/12/2	Solution(Org)	U(90)	64.8	100	none	none	1.6×10^{16}	2.5×10^{14}	16h
14	Wood River	1964/7/24	Solution	U(93)	51	103.7	1.0×10^{17}	2.4×10^{15}	1.3×10^{17}	2.5×10^{15}	1.5h
15	Electrostal	1965/11/3	Slurry	U(6.5)	100	300	none	none	1.0×10^{16}	1.0×10^{14}	1min[3)]
16	Mayak	1965/12/16	Solution	U(90)	28.6	100	none	none	5.5×10^{17}	1.9×10^{16}	7h
17	Mayak	1968/12/10	Solution(Org)	Pu	28.8	62.1	3.0×10^{16}	1.0×10^{15}	1.0×10^{17}	3.5×10^{15}	15min[5)]
18	Windscale	1970/8/24	Solution(Org)	Pu	40	156	none	none	1.0×10^{15}	2.5×10^{13}	10s
19	ICPP	1978/10/17	Solution	U(82)	315.5	315.5	unknown	unknown	2.7×10^{18}	8.6×10^{15}	1.5h
20	Tomsk	1978/12/13	Metal	Pu	0.54	3.2	3.0×10^{15}	5.6×10^{15}	3.0×10^{15}	5.6×10^{15}	1min[3)]
21	Novosibirsk	1997/5/15	Slurry	U(70)	*	700	none	none	5.5×10^{15}	*	27h5min
22	Tokai-mura	1999/9/30	Solution	U(18.8)	45	100	5.0×10^{16}	1.1×10^{15}	2.5×10^{18}	5.6×10^{16}	19h40min

1) **Mayak**: Mayak Production Association (Russia), **Y-12**:Oak Ridge Y-12 Plant (USA), **LASL**: Los Alamos Science Laboratory (USA), **ICPP**: Idaho Chemical Processing Plant (USA), **Tomsk**: Siberian Chemical Combine (Russia), **Hanford**: Hanford Works (USA), **Wood River**: United Nuclear Fuel Recovery Plant (USA), **Electrostal**: Electrostal Machine Building Plant (Russia), **Windscale**: Winscale Works (UK), **Novosibirsk**: Novosibirsk Chemical Concentration Plant (Russia), **Tokai-mura**: JCO Fuel Fabrication Plant (Japan).

2) Number in parentheses shows enrichment of ^{235}U. 3) Set 1min for "a short time" or "single excursion."

4) Set 3min for "a few minutes." 5) Set 15min for "15min + alpha."

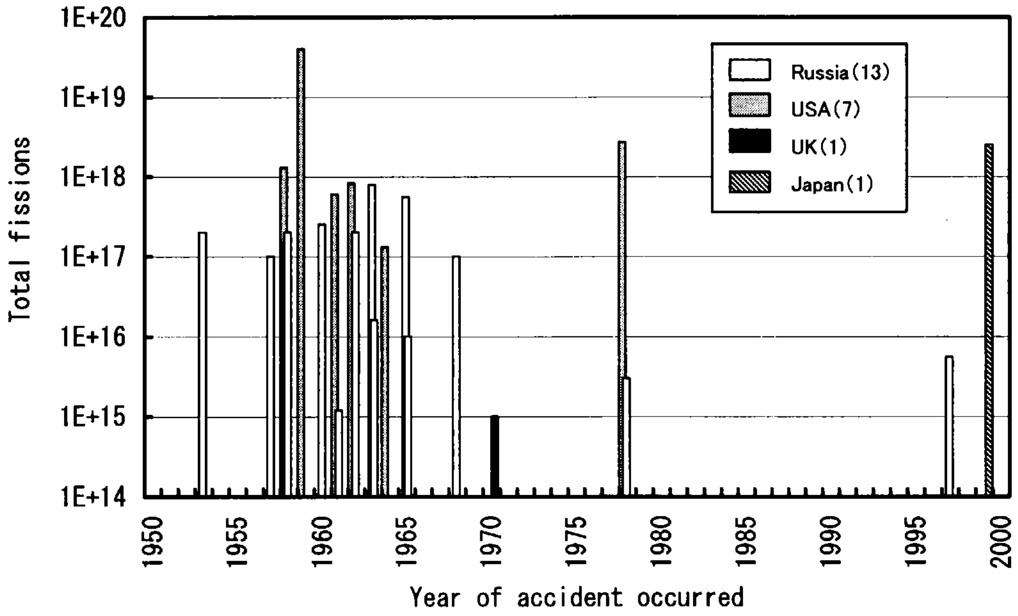

Fig.1 Chronological record of criticality accidents in nuclear fuel processing plants

Table 2 Simplified Methods

	Items	Evaluation formulas
Tuck	Maximum fissions in the initial burst (Maximum fissions during a 5-sec interval)	$F_B = 2.4\times10^{15}\cdot V\cdots$ U system (+70%, -90%) $F_B = 4.6\times10^{16}a^{1/4}\exp\left[0.0177D-\frac{0.8\cdot(150-H)a}{D}\right]\cdots$ Pu system (+100%, -70%) where V: tank volume (ℓ), a: solution feed rate (ℓ/s), D: tank diameter (cm), H: tank height(cm), if H>150, then use 150.
	Maximum specific fission rate in the initial burst (fission/ℓ·s)	$f_r = \frac{7.7\times10^{23}}{H^3D^2}\cdot k\cdot a\cdot\left(\frac{L}{S}\right)^{1/2}$ (+110%, -70%) where k: source initiating parameter (U system: 1～100, Pu system: 1), L: longest dimension(H or D in cm), S: shortest dimension (H or D in cm), H = 20cm for D = 29～40cm, H = 10cm for D > 40cm.
	Total fissions	$F_T = V\times10^{17}$ (±20%) Fissions needed to evaporate the solution with about 20% heat loss.
	Applicable conditions	1) Solution feed rate is the maximum design capacity of the system. It must be between 0.47 and 0.006 ℓ/s. 2) Tank volume refers to the largest tank in the system. The diameter must be between 28 and 152 cm. 3) Fissile solution volume is the maximum one for normal operations plus that which could credibly be inadvertently added. 4) The tank bottom is 30cm or more above any reflecting material such as concrete. 5) The fuel concentration is that which will produce the worst accident for the conditions involved. 6) After a criticality alarm, any operating equipment, such as pumps or valves, is not shut off by the operator. 7) The heat transfer from the tank to the environment must be not greater than a bare tank cooled by room air. Little venting steam will condense in the vent lines and drain back into the tank.
Olsen	Fissions in the initial burst	$F_B = 2.95\times10^{15}\cdot V_B^{0.85}$ where V_B: solution volume at the time of the burst. This model is mainly applicable to highly enriched uranium. For Pu systems, F_B becomes half or less. It is not estimated for slightly enriched uranium system.
	Fissions in the plateau	$F_P = 3.2\times10^{18}\cdot(1-t)^{-0.15}$ where t: duration of the plateau.
	Total fissions	$F_T = F_B + F_P$
	Applicable conditions	1) Tank diameter must be between 30 and 80 cm. 2) Solution feed rate must be between 0.027 and 0.52 ℓ/s.
Barbry	Total fissions	$F_T = \frac{V\cdot t}{3.55\times10^{-15}+6.38\times10^{-17}\cdot t}$ where V: solution volume (ℓ), t: duration (s) that should be less than 600 s.
	Applicable conditions	1) The model is valid for uranyl nitrate solutions in a homogeneous media with highly enriched uranium, and having an appreciable nominal internal neutron source. 2) The model should be applicable to Pu solutions. 3) Experimental data with the following conditions are used. Fuel concentration: 20～360gU/ℓ, Tank: cylindrical shape with diameters of 30, 80 cm (CRAC) and 36 cm (SILENE).
Nomura	Total fissions	$F_T = 2.6\times10^{16}\cdot V\cdots$ for non-boiling (Case #1). $F_T = 6\times10^{16}\cdot V\cdots$ for boiling (Case #2). where V: solution volume (ℓ).
	Applicable conditions	1) Excursion will terminate by the temperature rise from 20 to 110°C for Case #1. 2) Excursion will terminate by the evaporation of 25 % of solution volume during boiling for Case #2.

In these evaluations, vessel (tank) volume is used, and solution feed rate is also required for the evaluation of several items

3.2 Olsen's Method

Formulas to evaluate the fissions in the initial burst and the plateau are proposed. For the plateau, fissions as a function of duration are evaluated. This method is developed based on the data of CRAC experiments, thus the method is applicable for HEU system. It is estimated that the fissions in the initial burst for plutonium system will be half of that for HEU system. There is no estimation for slightly enriched uranium system.

3.3 Barbry's Method

Formula gives the time change of fissions for the first 600 seconds of excursion. This method is based on the CRAC and SILENE experimental data. It is applicable for the HEU solution having a significant neutron source. It is estimated that the formula is also applicable for Pu solutions.

3.4 Nomura's Method

Two formulas to evaluate the total fissions for non-boiling case and for boiling one are proposed. For non-boiling case, it is assumed that the temperature rise of 90°C terminates the excursion. For boiling case, evaporation of 25% of solution volume is assumed. The formulas are verified for the CRAC experiments and past accidents in the US and UK.

4. Evaluation of Accidents

4.1 Fissions in the Initial Burst

Figure 2 shows the evaluated fissions in the initial burst using Tuck's and Olsen's formulas, in comparison with the past accidents. In Tuck's formula for Pu system, the tank height was changed as a parameter (for H > 150 cm, H = 150 cm), while the diameter was set to the maximum one (152 cm), and the maximum solution feed rate of 0.47 liter/s was employed.

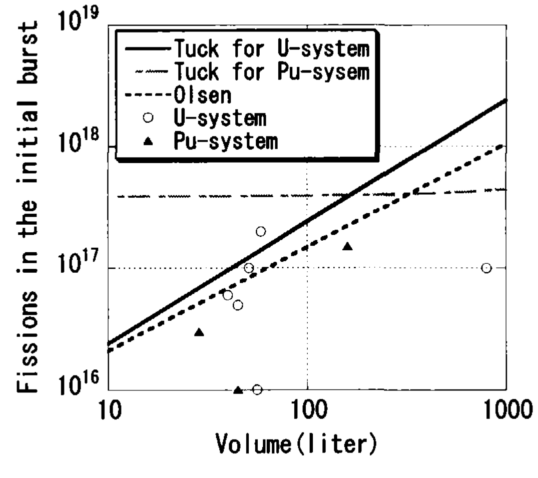

Fig.2 Fissions in the initial burst estimated by Tuck and Olsen

Although those formulas show the dependence on the solution volume, the accident data have no significant relation with the volume. We can say that it is difficult to determine or evaluate the power behavior in the initial burst of the past accidents, and then the accuracy of the reported value is not enough. Therefore, we can not judge the applicability of the simplified methods using the present results.

4.2 Total Fissions as a Function of Volume

Figure 3 shows the evaluated total fissions as a function of solution volume using Tuck's and Nomura's formulas. Nomura's formula #1 (for non-boiling case) gives the upper envelope value except two cases. These are the Tokai-mura accident (No.22 in Table 1) and the ICPP one (No.6). In the Tokai-mura accident, the cooling system was activated and that resulted in the relatively high power level for long time. In the ICPP accident, half of the solution was evaporated due to the violent boiling.

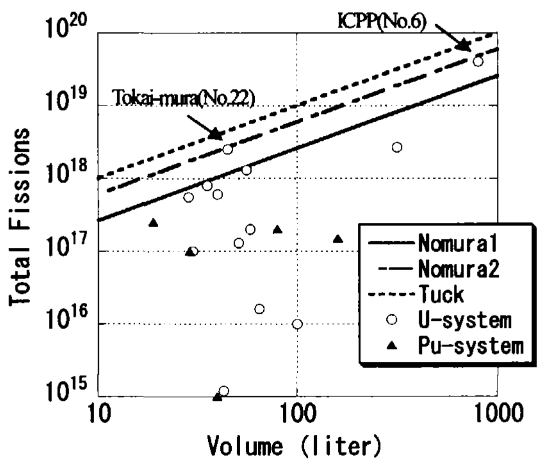

Fig.3 Total fissions as a function of fuel volume estimated by Tuck and Nomura

Both cases are beyond the applicable conditions for Nomura's formula #1. On the other hand, Nomura's formula #2 (for boiling case) reproduces those two accidents well. However, the Tokai-mura accident is also beyond the conditions, thus we think that this agreement is caused by coincidence. Tuck's formula overestimates all the accidents; this indicates that the assumption of all volume evaporation is too conservative.

After all, Nomura's two formulas give good results for all the cases except the Tokai-mura accident.

Figure 4 shows the evaluated total fissions as a function of solution volume using Olsen's and Barbry's formulas with infinitive duration. In Olsen's formula, fissions in plateau (3.2×10^{18} for t →∞) are dominant, and hence the result becomes almost constant. Barbry's formula gives the almost same result with Nomura's formula #1; however it underestimates the fissions for several cases.

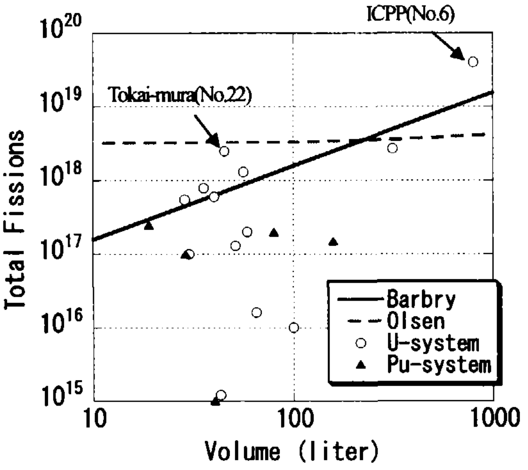

Fig.4 Total fissions as a function of fuel volume estimated by Olsen and Barbry

4.3 Total Fissions as a Function of Duration

Figure 5 shows the evaluated total fissions as a function of duration using Olsen's formula. As previously mentioned, fissions in the plateau becomes dominant with increasing duration in Olsen's formula. The evaluation shows the upper envelope for the total fissions except one case of the ICPP accident (No.6), in which the violent boiling was occurred and large numbers of fission yielded in a short time.

Figure 6 shows the evaluated total specific fissions as a function of duration using Barbry's formula. The evaluation almost reproduces the upper values with the difference of 50% except two cases, i.e., Tokai-mura(No.22) and ICPP(No.6). It is found from the figure that these two accidents give the quite larger total specific fissions than the others. In particular, Tokai-mura accident gives the largest total specific fissions in the past accidents. This is due to the solution cooling effect, which brought the relatively high power for a long time. For the evaluation of the Tokai-mura accident, it is necessary to develop a new formula including the cooling effect.

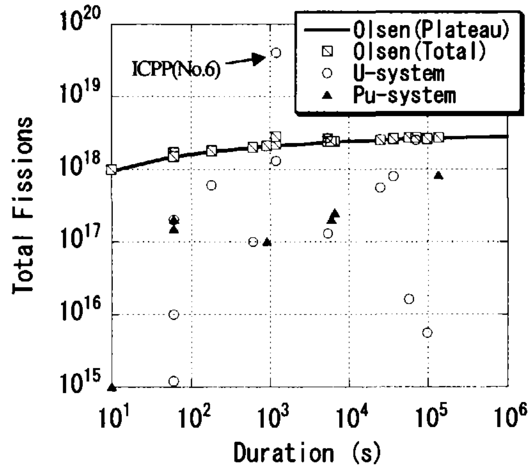

Fig.5 Total fissions as a function of duration estimated by Olsen

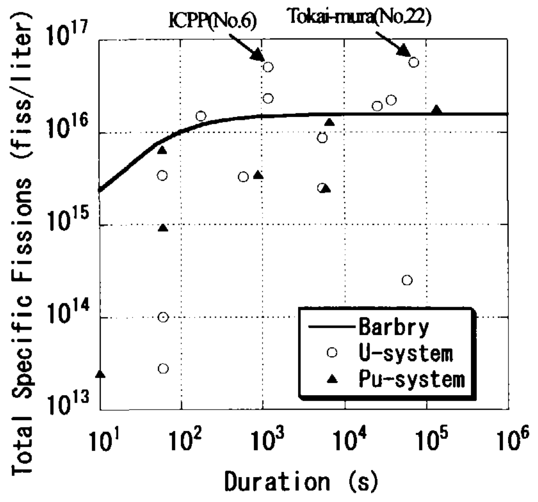

Fig.6 Total specific fissions as a function of duration estimated by Barbry

5. Conclusions

Applicability of four simplified methods, proposed by Tuck, Olsen, Barbry and Nomura, to evaluate the consequences of criticality accident was investigated. Fissions in the initial burst and total fissions were evaluated using those simplified methods and the results were compared with the past accident data. The following conclusions were obtained in the present study.

- The simplified methods give the number of fissions in the initial burst as a function of solution volume; however the accident data did not show such tendency. This would be caused by the lack of accident data for the initial burst with high accuracy.
- For the total fissions as a function of solution volume, Nomura's formula #1 reproduced the upper envelope of the accidents for non-boiling cases. Nomura's formula #2 predicted well the total fissions of the boiling accident of ICPP(No.6). For the Tokai-mura accident, in which the cooling system was activated, Nomura's formula #2 showed good agreement, however it would be caused by coincidence. Barbry's formula with infinitive duration showed the similar tendency with the Nomura's formula #1.
- For total fissions as a function of duration, Olsen's formula shows the upper envelope for the total fissions except one case of the ICPP accident (No.6), in which the violent boiling was occurred and large numbers of fission yielded in a short time. Barbry's formula almost reproduces the upper values of total specific fissions with the

difference of 50% except two cases, i.e., Tokai-mura(No.22) and ICPP(No.6). These two accidents gave the quite larger total specific fissions than the others. In particular, Tokai-mura accident gave the largest total specific fissions in the past accidents. This is due to the solution cooling effect, which brought the relatively high power for a long time. For the evaluation of the Tokai-mura accident, it is necessary to develop a new formula including the cooling effect.

References

1) G. Tuck, "Simplified Methods of Estimating the Results of Accidental Solution Excursions," Nucl. Technol., **23**, 177 (1974).
2) A. R. Olsen et al., "Empirical Model to Estimate Energy Release from Accidental Criticality," Trans. Am. Nucl. Soc., **19**, 189 (1974).
3) F. Barbry et al., "Criticality Accident Studies in France: Experimental Programs and Modelisation," Proc. Int. Seminar on Nuclear Criticality Safety, ISCS'87, Tokyo, Japan, October 19-23, 1987, p. 423 (1987).
4) Y. Nomura and H. Okuno, "Simplified Evaluation Models for Total Fission Number in a Criticality Accident," Nucl. Technol., 109, 142 (1995).
5) P. Lecorche et al., "A Review of the Expriments Performed to Determine the Radiological Consequences of a Criticality Accident," Y-CDC-12 (1973).
6) F. Barbry, "A Review of the SILENE Criticality Excursions Experiments," Proc. 1993 Topical Meeting on Physics and Methods in Criticality Safety, Nashville, TN, USA, September 19-23, 1993, p. 34 (1993).
7) M. S. Dunenfeld et al., "Summary Review of the Kinetic Experiments on Water Boilers," NAA-SR-7087 (1963).
8) T. P. McLaughlin et al., "A Review of Criticality Accidents, 2000 Revision," LA-13638 (2000).