

Conf-9204105--6

The submitted manuscript has been authored by a contractor of the U. S. Government under contract No. W-31-109-ENG-38. Accordingly, the U. S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U. S. Government purposes.

RESULTS OF AEROSOL CODE COMPARISONS WITH RELEASES FROM ACE MCCI TESTS

J. K. Fink  
Argonne National Laboratory, Argonne, IL, USA

ANL/CP--74553

M. Corradini  
University of Wisconsin, Madison, Wisconsin, USA

DE92 010510

A. Hidaka  
Japan Atomic Energy Research Institute,  
Tokai-mura, Naka-gun, Ibaraki-ken, Japan

E. Hontañón  
Polytechnical University of Madrid, Madrid, Spain

M. A. Mignanelli  
AEA Technology, Harwell, UK

E. Schrödl  
Gesellschaft für Reaktorsicherheit, Köln, Germany

V. Strizhov  
Institute for Nuclear Safety, Moscow, Russia

Received by OSTI  
APR 01 1992

ABSTRACT

Results of aerosol release calculations by six groups from six countries are compared with the releases from ACE MCCI Test L6. The codes used for these calculations included: SOLGASMIX-PV, SOLGASMIX Reactor 1986, CORCON.UW, VANESA 1.01, and CORCON mod2.04/VANESA 1.01. Calculations were performed with the standard VANESA 1.01 code and with modifications to the VANESA code such as the inclusion of various zirconium-silica chemical reactions. Comparisons of results from these calculations were made with Test L6 release fractions for U, Zr, Si, the fission-product elements Te, Ba, Sr, Ce, La, Mo and control materials Ag, In, and Ru. Reasonable agreement was obtained between calculations and Test L6 results for the volatile elements Ag, In and Te. Calculated releases of the low volatility fission products ranged from within an order of magnitude to five orders of magnitude of Test L6 values. Releases were over and underestimated by calculations. Poorest agreements were obtained for Mo and Si.

MASTER

## INTRODUCTION

The CSNI benchmark exercise on the chemical modelling of the release of radionuclides due to core-concrete interactions [1,2] identified five reasons for different calculated releases by VANESA and other codes, namely: (1) absence of direct interaction between the condensed phases; (2) use of the metal phase alone to set the oxygen potential rather than the oxygen conservation equation and allowing the oxide phase to set its own oxygen potential; (3) failure to include all species in the gas composition; (4) neglect of vaporization from the metal phase; and (5) choice of ionic versus molecular solution model for the condensed phase with failure to include certain condensed phase interactions such as Zr-SiO<sub>2</sub>. The differences in releases due to different treatments of the solution phases, choice of thermodynamic functions, and uncertainties in thermodynamic data for a specific problem were also assessed in the benchmark exercise. The CSNI report [2] pointed out that uncertainties in the modelling of releases from core-concrete melts may be further reduced by comparison of calculations with releases from experiments. The Advanced Containments Experiments (ACE) molten core concrete interactions (MCCI) test program provides opportunities to compare calculations with experimental releases. Tests L6 and L8 have been selected for comparison with blind posttest calculated releases. Code comparisons for Test L6, a siliceous concrete test, have been completed. Comparisons with Test L8, which used limestone/limestone concrete, are underway.

The focus of this paper is the results of the Test L6 code comparison. Results of twenty-two aerosol release calculations by six groups from six countries were submitted for comparison with test data. Fourteen of these calculations were posttest blind calculations. Only results of these blind calculations will be included in this paper. In cases where more than one calculation was done with one code, the calculation with the best agreement with Test L6 releases has been selected for comparison in this paper.

In the next section, Test L6 parameters required for the code calculations are given. The third section gives an overview of the codes used in the release calculations. The focus of the fourth section is comparison of results of calculations with releases from the experiment and discussion of the results. Conclusions and recommendations are given in the last section.

## TEST L6

Test L6 determined the releases from the interaction of a PWR corium with 30% Zr oxidation and a siliceous concrete basemat. The test apparatus and composition of the corium, concrete/metal inserts and concrete in the ACE MCCI experiments are described in other papers [3-5]. The corium contained  $UO_2$ ,  $ZrO_2$ ,  $CaO$ ,  $SiO_2$ , and oxides of fission-product elements at twice the reactor inventory. The concrete/metal inserts contained stainless steel, Zr, Ru, Ag, In, and  $ZrTe_2$ . The siliceous concrete basemat was 50.2 cm by 49.2 cm in area. At the time aerosol collection was terminated, about 8.8 cm of the basemat had been ablated. Four reinforcing rods located at 5.2 cm in the basemat had been incorporated in the melt.

Test data provided for the aerosol code calculations included: moles of each species of off gas released upward as a function of time, ablation rate, and melt temperature as a function of time. Because off gas from the concrete migrated downward during the test, the off gas detected above the melt was only about half of the gas available from the ablated concrete. All times were given relative to the surface of the concrete basemat reaching 1673 K. The melt temperature during insert ablation was assumed to be 2500 K. Melt temperature during basemat ablation varied from 2425 K at 12.5 min to 2263 K at 30 min.

Aerosol and off gas release began at -52 min ( 52 min prior to the basemat surface reaching 1673 K) when the concrete/metal inserts reached 373 K. Concrete/metal insert ablation began at -44 min. At 31 min, the diluter in the off gas line plugged and collection of aerosols was terminated. Release calculations were to be done from -52 to 31 min.

## PARTICIPANTS

The six participants and the codes used are given in Table 1. In order to minimize the number of comparisons shown in figures and tables, the calculation giving best agreement with experimental releases was chosen when more than one calculation was performed with one code or modifications of that code. Abbreviations for the calculations that are used in figures and tables in this

Table 1. Test L6 Aerosol Release Code Comparison Participants

Participant	Organization	Code	Total Release Calculations	Symbols
M. A. Mignanelli	AEA Technology Harwell, UK	SOLGASMIX REACTOR Version 1986	2	UK
E. Schrödl	GRS, Germany	SOLGASMIX - PV	3 <sup>a</sup>	GRS
E. Hontañón	Polytechnical University of Madrid, Spain	SOLGASMIX	1	S-S
		VANESA 1.01	2	SV2
A. Hidaka	JAERI, Japan	VANESA 1.01	1	JPN
V. Strizhov	Nuclear Safety Institute, Moscow, Russia	VANESA Mod 7/15/1985 CORCON Mod2.04	4 <sup>b</sup>	URC, URE
M. Corradini	University of Wisconsin, USA	CORCON.UW	1	WIS
			Σ 14	

<sup>a</sup>3 total releases were calculated. For one case, separate calculations gave the release for ablation of the concrete/metal insert and for the ablation of the basemat. The total release was the sum of the releases from these calculations.

<sup>b</sup>The release during the ablation of the concrete/metal inserts was determined in a separate calculation. Results of this calculation were used with 4 separate calculations of the ablation of the basemat to obtain 4 sets of total releases for Test L6.

paper are included in the last column of Table 1.

Data from various sources were used in these calculations. In some calculations, off gas was determined from concrete ablation. Others used the upward off gas from Test L6 data. Melt temperatures were either held constant, obtained from the test data, or determined from thermal hydraulic code calculations. The sources of melt temperature, off gas released, and basemat ablation rate for each calculation are summarized in Table 2.

SOLGASMIX REACTOR is a modification by AEA Technology of the SOLGASMIX code developed by Eriksson [6]. In his calculation using the SOLGASMIX REACTOR code,

Table 2. Data Used in Code Calculations

Calculation	Temperature, K		Gas Available		Basemat Ablation
	Inserts	Basemat	H <sub>2</sub> O	CO <sub>2</sub>	
SOLGASMIX					
UK	2500	Test L6	Concrete	Concrete	Test L6
GRS	2400	2400	Concrete	Concrete	----- <sup>e</sup>
S-S	2400	2400	Test L6	Test L6	----- <sup>f</sup>
VANESA					
SV2	CORCON <sup>a</sup>	CORCON <sup>b</sup>	Test L6	Test L6	Test L6
JPN	2500	Test L6	Test L6	Concrete	Test L6
URC	2500	Test L6	Test L6	Test L6	Test L6
URE	2500	CORCON	CORCON	CORCON	CORCON
CORCON.UW					
WIS	CORCON.UW <sup>c</sup>	CORCON.UW <sup>d</sup>	CORCON.UW	CORCON.UW	CORCON.UW

<sup>a</sup>Temperatures ranged from 2500 to 2239 K.

<sup>b</sup>Temperatures were 100 to 200 K below Test L6 measured values; Temperatures ranged from 2226 to 2172 K.

<sup>c</sup>Temperature fell from 2500 to 2350 K in first 25 min then stayed around 2350 K for rest of time.

<sup>d</sup>Temperature started at 2350 K vs 2425 K (Test L6); good agreement with Test L6 data at 20 and 25 min.

<sup>e</sup>Only 2 time steps: insert, basemat.

<sup>f</sup>Only 1 time interval for entire test (insert and basemat ablation).

Mignanelli treated insert ablation in one time step and included 9 time steps for basemat ablation. SOLGASMIX-PV, the adaptation of SOLGASMIX by T. Besmann at ORNL [7], was used by Hontañón and Schrödl in their calculations. Only one time step at 2400 K was used in the calculation by Hontañón. Two calculations by E. Schrödl used single time steps at 2400 K. His third result, also at 2400 K, was comprised of two separate calculations. First the release from insert ablation was calculated. Then the residual mass from this calculation was included with the ablated basemat and rebar for the second calculation. The total release was the sum of the releases from the two calculations. This combined calculation

gave the best agreement with experimental releases.

The databases used by Mignanelli and Schrödl included zirconates and silicates. Mignanelli did calculations with and without lanthanum zirconate in the database. Agreement was best when lanthanum zirconate was included. The VANESA data base, which does not include silicates nor zirconates was used for the SOLGASMIX calculation by Hontañón.

VANESA 1.01 was used in all VANESA calculations. In response to the CSNI benchmark exercise which showed that failure to include condensed phase reactions of Zr with  $\text{SiO}_2$  significantly effected releases [1,2], modifications of the VANESA chemistry to include additional condensed phase reactions were made in some calculations. The multiple calculations with the VANESA code by Hontañón and Strizhov included both calculations with the standard version and with modifications to include the chemical reactions of  $\text{SiO}_2$  and Zr. Hontañón's calculation, SV2, included the reaction of  $\text{SiO}_2$  and Zr to form SiO gas. The modified VANESA calculations (URC and URE) by Strizhov included the chemical reaction of  $\text{SiO}_2$  and Zr to form Si. Results of calculations with either reaction gave better agreement than the unmodified VANESA 1.01 code. The four calculations by Strizhov included the four combinations of calculations with and without modifications to the VANESA chemistry and with either thermal hydraulic data from CORCON calculations or thermal hydraulic data from Test L6. Results of two calculations (URC and URE) were selected for comparison in order to compare completely blind posttest calculations and to evaluate the importance of test data versus CORCON output as input. The calculation by the JAERI analysts led by A. Hidaka used the standard VANESA code with a modification to limit the partial pressures of the releases. Details of the JAERI calculations are presented in a separate paper. [8]

CORCON.UW is a modified version of CORCON that includes complete melt chemistry. [9] It is a completely blind calculation in that both thermal hydraulic results and releases are calculated as a function of time in this code. The database for CORCON.UW does not include any silicates nor zirconates in the condensed phase.

## RESULTS

Release fractions were calculated from Test L6 for U, Zr, Si, Te, Ba, Sr, La, Ce, Mo, Ag, In, and Ru for comparison with the code calculations. Si was included in the aerosol code comparison because of its dominance in the aerosols released from Test L6. For Ce, La and Ru, only maximum and minimum release estimates are currently available from the test data because these elements were below the level of detectability in most aerosol samples. Minimum values were calculated by assuming zero when the element was not detected. Maximum values were calculated assuming the limit of detectability when the element was not detected. Calculated releases for Ce and La will be reassessed when more sensitive neutron activation analysis for Ce and La are complete.

Calculated release fractions ranged from within an order of magnitude of Test L6 values to differences of 5 orders of magnitude. In Table 3, the differences between the release fractions obtained from the calculations and Test L6 are summarized. Results for each element are discussed below.

### Ag, In, and Te

The best agreements between calculations and experimental data were obtained for the volatile elements: Ag, In, and Te. Results for Ag are shown in Figure 1. For these elements, releases from all calculations were within an order of magnitude of the Test L6 releases.

### Ce and La

Calculated release fractions of Ce and La ranged over 3 orders of magnitude as shown in Figures 2 and 3. Releases from all VANESA calculations except for calculation SV2 were high for both Ce and La. Results of the SOLGASMIX calculation S-S, which used the VANESA database, were also high. Mignanelli found that inclusion of lanthanum zirconates significantly reduced the calculated La release. In his vaporization studies, Roche[10] found inclusion of silicates and zirconates in the melt chemistry was necessary to obtain good agreement with his experimental results via a chemical equilibrium calculation. Although preliminary results indicate that silicates and zirconates of Ce may also be important, no conclusion should be drawn until final Ce analyses for Test L6 have

Table 3. Order of Magnitude Deviation of Calculated Releases from Test L6 Releases

	UK	GRS	S-S	SV2	JPN	URC	URE	WIS
Te	+0.5 <sup>a</sup>	+0.5	-0.5	+0.5	-0.5	+0.5	+0.5	+0.5
Ag	+0.5	+0.5	-1	----	-0.5	-1	-0.5	+0.5
In	+0.5	+0.5	----	----	----	+1	+0.5	----
Ce	+0.5	0 <sup>b</sup>	+0.5	0	+2	+2	+2	0
La	0	0	+0.5	0	+0.5	+1	+1	0
Ru	0	0	-0.5	-1	-1	-1	-1	0
Ba	-0.5	-1	+0.5	+0.5	+0.5	+2	+2	-0.5
Sr	-0.5	-0.5	+0.5	-1	+2	+2	+2	+1
U	-1	-1	-1	+0.5	-0.5	-3	-2	-2
Zr	-1	-2	-3	----	-2	----	----	-3
Mo	-3	-3	-5	+0.5	----	-2	-2	----
Si	+0.5	+0.5	+0.5	----	-2	-4	-5	----

<sup>a</sup>For orders of magnitude from 0.1 to 0.5, 0.5 is used.

<sup>b</sup>0 is used for calculated releases between maximum and minimum Test L6 releases.

been completed. The lack of silicates and zirconates in the VANESA database could be a major factor in the high releases from VANESA calculations and the SOLGASMIX calculation, S-S. The good agreement of CORCON.UW, which does not include silicates and zirconates in the database, is surprising if these species are important. Examination of the CORCON.UW calculation shows that the calculated melt temperatures during insert and early basemat ablation are far below Test L6 values (see Table 2). The good agreement of CORCON.UW results



could be fortuitous, a result of these low temperatures. The VANESA SV2 calculation includes the silica-zirconium reaction to produce SiO. Inclusion of this interaction has a significant effect on the oxygen potential and this modification may compensate for not including additional species in the condensed phase.

#### Ru

Calculated Ru releases were either between the Test L6 maximum and minimum or low. All calculations that used the VANESA database were low.

#### Ba and Sr

Calculated releases for Ba and Sr ranged from one to three orders of magnitude of Test L6 values. Figures 4 and 5 indicate that releases are over- and underestimated. Roche [10] showed the importance of barium and strontium silicates and zirconates in the solidified melt and in his chemical equilibrium calculations for his vaporization release studies. Ba and Sr release calculations without silicates and zirconates in the database were high except for the VANESA calculation SV2 for Sr and CORCON.UW for Ba. The UK and GRS SOLGASMIX calculations, which included silicates and zirconates in their databases, were low. The GRS calculation for Ba was significantly worse than for Sr indicating that the low temperature used in this calculation had a greater effect on Ba release than on Sr release. A SOLGASMIX calculation by Schrödl with 2550 K for the insert temperature and 2400 K for the basemat temperature gave Ba and Sr releases within a factor of 2 of experimental values [11]. The better agreement for Ba compared to Sr in calculations without silicates and zirconates may be due to the sensitivity of Ba release to temperature and the low temperatures used in the calculations.

#### U and Zr

Calculated releases for U and Zr were low except for calculation SV2. Results for U are shown in Figure 6. In Test L6, higher fractions of U and Zr were detected in large particles, that may have been mechanically generated, than in small particles from vaporization. This effect was particularly great for Zr. A possible explanation for the deviations for U and Zr is that the mechanical contribution to the release of these elements is significant and is either not calculated (SOLGASMIX and CORCON.UW) or is underestimated (VANESA). Other

effects such as additional vapor species should also be considered.

#### Mo

For Mo, calculated releases ranged over 6 orders of magnitude. All calculations give low releases for Mo except for SV2. The reason for the large disagreement between calculations and experiment for Mo is not understood.

#### Si

For Si, deviations between calculated and experimental releases were as large as 5 orders of magnitude. The SOLGASMIX calculations gave significantly better agreement with Test L6 releases than the VANESA calculations. This may be due to the importance of inclusion of all  $\text{SiO}_2$ -Zr reactions. In Test L6, the aerosol composition was dominated by aerosols formed from SiO gas.

#### Discussion of Results

The important differences in the calculations identified in the CSNI benchmark exercise were confirmed in the Test L6 code comparison. The five causes of differences in VANESA and other calculations identified in the CSNI study[1,2] are addressed with respect to the Test L6 code comparison below. The importance of inclusion of interactions between condensed phases is illustrated by the better agreement of results from the CORCON.UW code (WIS), which includes interaction between the condensed phases, compared with the standard VANESA calculations. Comparison of calculations with Test L6 releases confirm the importance of the oxygen potential and the assumptions used to set it. The oxygen potential from the JAERI VANESA calculation (JPN) was significantly lower than those from the UK and GRS SOLGASMIX calculations. In addition the JPN oxygen potential variation with time differed significantly from that obtained in the UK calculation. Failure to include all species in the gas composition and neglect of vaporization from the metal phase may contribute to the high releases for oxide species of Ce, La, Ba, and Sr and low releases of Ag and Zr in all VANESA calculations. The importance of inclusion of Zr-SiO<sub>2</sub> reactions was clearly illustrated by the differences in results of calculations: (1) SV2, which included the reaction to form SiO, (2) URC and URE, which included the reduction to Si, and (3) JPN, which did not include these reactions.

The CSNI study also found that the database used significantly effected release calculations. In his vaporization studies, Roche concluded that inclusion of silicates and zirconates was necessary for good agreement between chemical equilibrium calculations and his experimental results [10]. Silicates and zirconates were included in the condensed phases in the databases used in the UK and GRS calculations. Mignanelli did calculations with and without lanthanum zirconate in the database. The calculated La release was closer to Test L6 values when lanthanum zirconate was included. In general, agreements with Test L6 releases for Ba, Sr, and La were improved by inclusion of these species, as illustrated by comparison of the UK and GRS calculations with VANESA calculations JPN, URC, and URE. Although good agreement was obtained by the CORCON.UW calculation for some cases without inclusion of these species, the agreement is most likely fortuitous and due to the low temperature used in the calculations. Good agreement was not obtained for Sr which is not as temperature sensitive as Ba, Ce, and La. Inclusion of the reaction to form SiO by Hontañón in calculation SV2 seemed to compensate in some way for failure to include condensed phase species in the VANESA database.

#### CONCLUSIONS AND RECOMMENDATIONS

Results of this code comparison are useful in assessing progress on fission-product release calculations and in providing guidance with respect to areas for further research and model development. Conclusions and recommendations are given below.

1. Significant progress has been made by the development of various SOLGASMIX chemical equilibrium codes with extensive databases and the development of the CORCON.UW code which gives better agreement with Test L6 than the CORCON mod2.04/VANESA 1.01 codes. The SOLGASMIX calculations on Test L6 and other ACE MCCI tests have provided valuable contributions on the importance of various species in the melt chemistry and the effects of various test parameters on release.
2. Although some possible causes for discrepancies between calculated and measured releases have been proposed in this paper, the combined efforts

of specialists are needed to identify the causes of discrepancies between the calculated and measured releases for each element.

3. International agreement on an assessed database that includes silicates and zirconates of Ba, Sr, La, and perhaps Ce is needed. Experiments required to provide the experimentally sound data required for this database should be identified.
4. The melt chemistry and physics such as representation of simple or complex phases for oxide and metal solutions, choice of solution model (ionic vs molecular), treatment of melt as a mixture of immiscible liquids or separate layers and effect on oxygen potential, gas release and migration, and mechanical aerosol release should be examined to determine if more sophisticated models of thermochemical interactions and/or of physical processes are necessary to obtain good agreement with experimental releases.

#### ACKNOWLEDGEMENTS

Work sponsored by the Electric Power Research Institute under Contract RP 2802-12.

#### REFERENCES

1. M. A. Mignanelli and P. N. Smith, "CSNI/PWG4/TGFPC Benchmark Exercise on the Chemical Modelling of the Release of Radionuclides due to Core-Concrete Interactions: Results of Part A," CSNI Report 164, December 1989.
2. M. A. Mignanelli, "Benchmark Exercise on the Chemical Modelling of the Release of Radionuclides due to Core-Concrete Interactions: Results of Part B," October 1990.
3. B. R. Sehgal and B. W. Spencer, "ACE Program Phase C: Fission Product Release From Molten Corium Concrete Interactions (MCCI)," paper presented at the OECD CSNI Specialist Meeting on Core Debris Concrete Interactions, Karlsruhe, Germany, April 1-3, 1992.
4. D. H. Thompson, J. K. Fink, D. R. Armstrong, B. W. Spencer, and B. R. Sehgal, "Thermal-Hydraulic Aspects of the Large-Scale Integral MCCI Tests in the ACE Program," paper presented at the OECD CSNI Specialist Meeting on Core Debris Concrete Interactions, Karlsruhe, Germany, April 1-3, 1992.

5. J. K. Fink, D. H. Thompson, B. W. Spencer, and B. R. Sehgal, "Aerosols Released During Large-Scale Integral MCCI Tests in the ACE Program," paper presented at the OECD CSNI Specialist Meeting on Core Debris Concrete Interactions, Karlsruhe, Germany, April 1-3, 1992.
6. G. Eriksson, "SOLGASMIX, A Computer Program for Calculation of Equilibrium Compositions in Multiphase Systems," *Chemica Scripta* 8, 100-103, 1975.
7. T. M. Besmann, "SOLGASMIX-PV, A Computer Program to Calculate Equilibrium Relationships in Complex Chemical Systems," ORNL/TM-5775, April 1977.
8. A. Hidaka, K. Soda, J. Sugimoto, and N. Yamano, "Analyses of ACE MCCI Test L6 with the CORCON/VANESA Code," paper presented at the OECD CSNI Specialists Meeting on Core Debris Concrete Interactions, Karlsruhe, Germany, April 1-3, 1992.
9. J. K. Norkus and M. L. Corradini, "Modeling of Molten Core Concrete Interactions: Fission Product Release," Proceedings of the ANS/ENS Conference on Thermal Reactor Safety, Avignon, France, October 2-7 1988.
10. M. F. Roche, L. Leibowitz, J. L. Settle, C. E. Johnson, R. C. Vogel, and R. L. Ritzman, "Vaporization of the Strontium, Barium, Lanthanum, and Uranium from Mixtures of Uranium, Zirconia, Steel, and Concretes at 2150 K and 2400 K," *Nucl. Technol.* 96, 96-116, 1991.
11. E. Schrödl, private communication, 1991.

### DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Figure 1. Ag Release

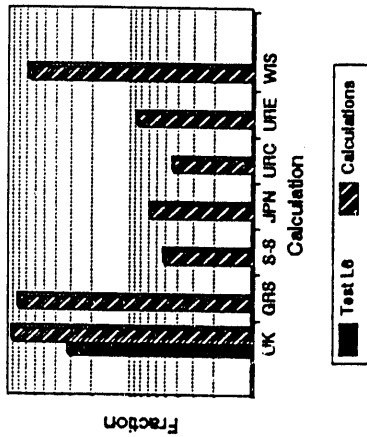


Figure 2. Co Release

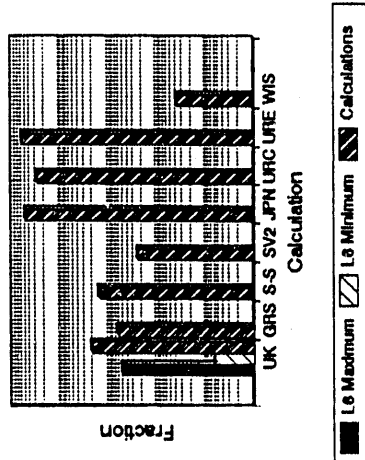


Figure 3. La Release

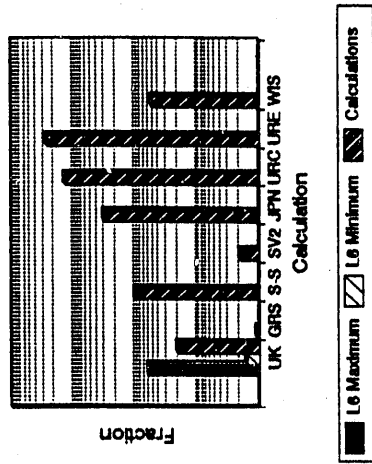


Figure 4. Ba Release

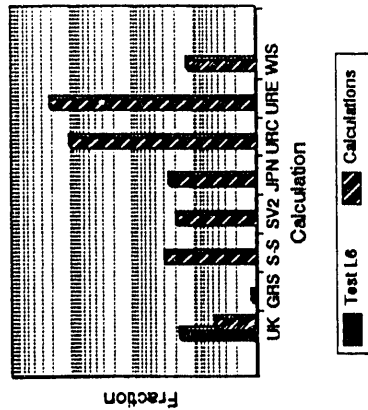


Figure 5. Sr Release

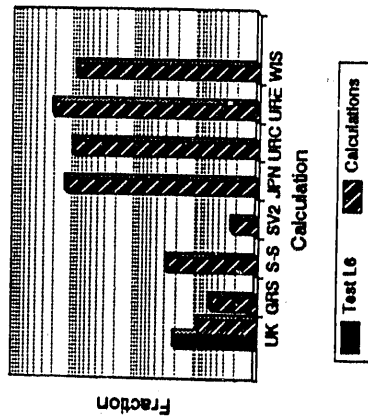
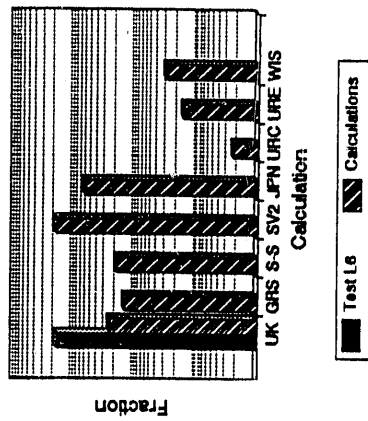


Figure 6. U Release



**END**

**DATE  
FILMED**

**5 / 19 / 92**

