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AEROSOLS RELEASED DURING LARGE-SCALE INTEGRAL MCCI TESTS IN THE ACE PROGRAM

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ABSTRACT

As part of the internationally sponsored Advanced Containment Experiments (ACE) program, seven large-scale experiments on molten core concrete interactions (MCCIs) have been performed at Argonne National Laboratory. One of the objectives of these experiments is to collect and characterize all the aerosols released from the MCCIs. Aerosols released from experiments using four types of concrete (siliceous, limestone/common sand, serpentine, and limestone/limestone) and a range of metal oxidation for both BWR and PWR reactor core material have been collected and characterized. Release fractions were determined for UO_2, Zr, the fission-products: BaO, SrO, La_2O_3, CeO_2, MoO_2, Te, Ru, and control materials: Ag, In, and B_4C. Release fractions of UO_2 and the fission products other than Te were small in all tests. However, release of control materials was significant.

INTRODUCTION

The test matrix for the ACE MCCI experiments is shown in Table 1. The ACE MCCI experiments and the thermal hydraulic results have been described in two other papers [1,2]. The focus of this paper is the aerosols released during these experiments. Therefore, only a short description of the experiments is given below followed by a section describing the aerosol system. The aerosol results are given in the third section. Release fractions of fission product elements and control materials are discussed in the last section.

The reactor core debris designated as corium contained UO_2, ZrO_2 and a small amount of the concrete constituents SiO_2 and CaO (MgO for serpentine concrete) to represent the concrete incorporated during the jet impingement phase. The

Table 1. ACE MCCI/Fission Product Release Test Conditions

Test	Concrete Type*	Net Heat Generation W/kg UO_2	Reactor Type	Initial Zr Oxidation, %	Control Rod Material
L5	L/S	300	BWR	100	
L2	S	450	PWR	70	
L1	L/S	350	PWR	70	
L6	S	350	PWR	30	Ag, In
L4	Soviet	250	BWR	50	B_4C
L7	L/S	250	BWR	70	B_4C
L8	L/L	350/150	PWR	70	Ag, In

*L/S = limestone/common sand concrete

S = siliceous concrete

Soviet = serpentine/ordinary structural (siliceous) concretes

L/L = limestone/limestone concrete

ratio of UO_2 to ZrO_2 was consistent with the reactor type and the extent of Zr oxidation. Oxides of fission-product elements Ba, Sr, La, Ce, and Mo were included in the corium at four times the amount present in the reactor inventory in tests L4, L7, and L8, and at twice the reactor inventory in all other tests. Tellurium was added as zirconium telluride in accordance with results of experiments at ORNL [3]. Ruthenium was alloyed with stainless steel. The Ag and In were alloyed. These metal alloys, Zr metal, B_4C, and stainless steel were included in the concrete/metal insert, which sat on top of the concrete, as shown in Figure 1.

The corium was heated by direct electrical heating. The molten pool started at the top of the corium and expanded horizontally and vertically until it reached the concrete/metal inserts. Because corium is an excellent insulator, the temperature of the concrete/metal inserts did not increase significantly above room temperature until shortly before insert ablation began. Significant aerosol release began when the concrete/metal inserts reached 373 K and free water was released from the concrete in the inserts.

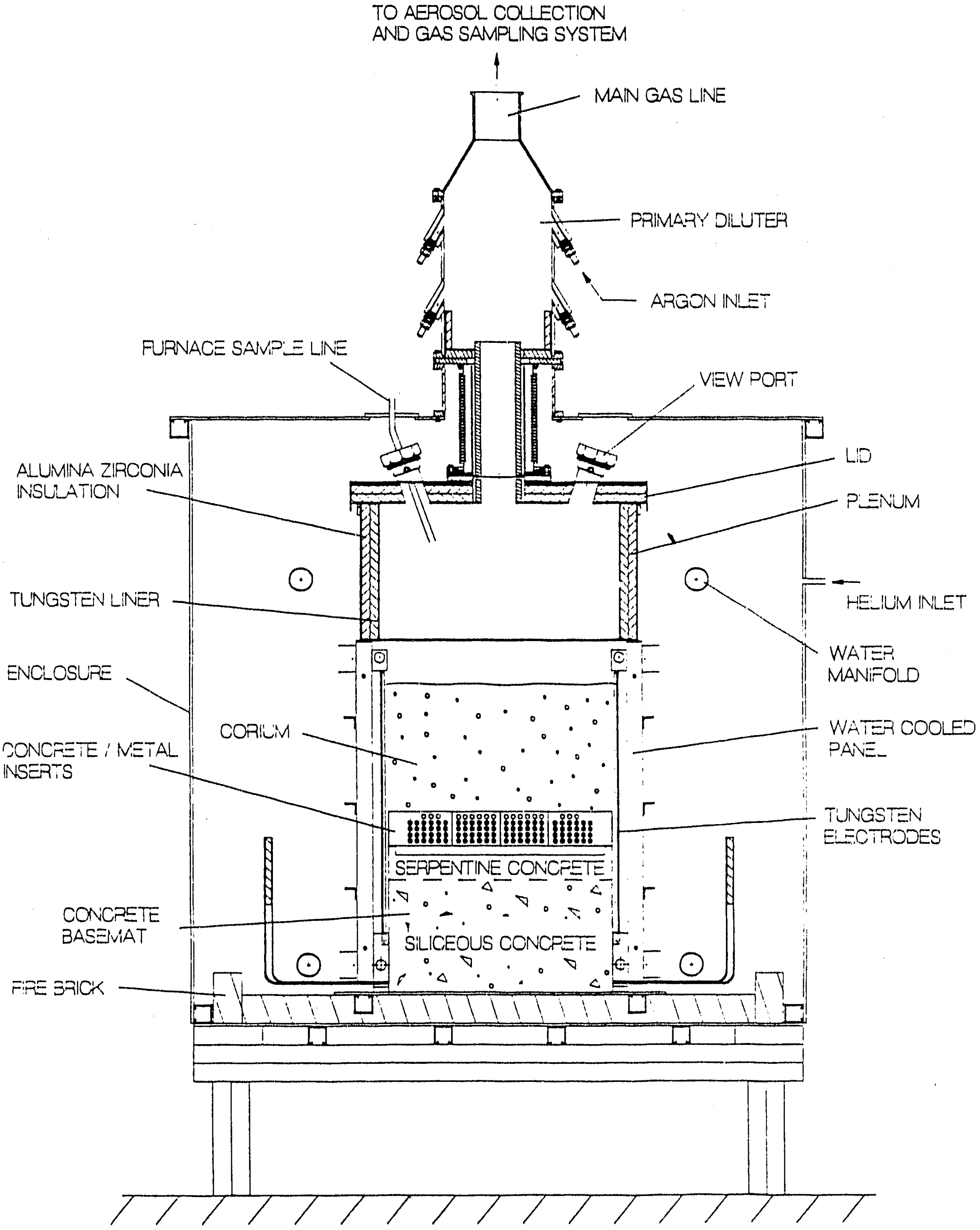

Figure 1. ACE MCCI Test Apparatus for Test L4

The aerosol compositions and release fractions have been determined for the seven experiments. Because Test L8 is an aerosol code comparison test, results from that test are being sequestered until calculations are complete. Thus, results from Test L8 are not included in this paper.

All the aerosols released from these integral tests were collected by the aerosol system so that the total aerosol mass released was measured. The aerosol compositions were determined from chemical analysis of representative samples of the aerosols released and the mass collected at each sampling location. A statistical analysis of the posttest sampling procedure was performed for Test L6 to ensure that reliable aerosol compositions and release data were obtained. The sampling error was less than the 10% uncertainty in the chemical analysis.

AEROSOL SYSTEM

The aerosol system was modified during the program to improve performance and alleviate plugging due to high silica release. A schematic of a typical aerosol system used in the ACE MCCI tests is shown in Figure 2. This system consisted of a main gas line, sample gas line, primary and secondary diluters, and three aerosol sampling modules (furnace sample line, elbow sample line, and cascade impactor sample line) plus vacuum pumps, filters, gas mass spectrometer, and pressure and temperature instrumentation. The furnace sample line rested on top of the enclosure; the remainder of the aerosol system was located on the aerosol deck above the test cell.

The released gas and aerosols were cooled and diluted in the primary diluter directly above the test apparatus. At the elbow above the primary diluter, two continuous isokinetic gas sample streams were drawn - one into the sample line, the other into the elbow sample line. The sample line aerosols were further diluted in a secondary diluter located ahead of the extinction system and sampling nozzle for the impactors. Variation in aerosol concentration with time was provided on line by the extinction system located on the sample line. Isokinetic samples by ten cascade impactors served to characterize the respirable fraction of the aerosols both in terms of particle size distribution and variation in composition with size.

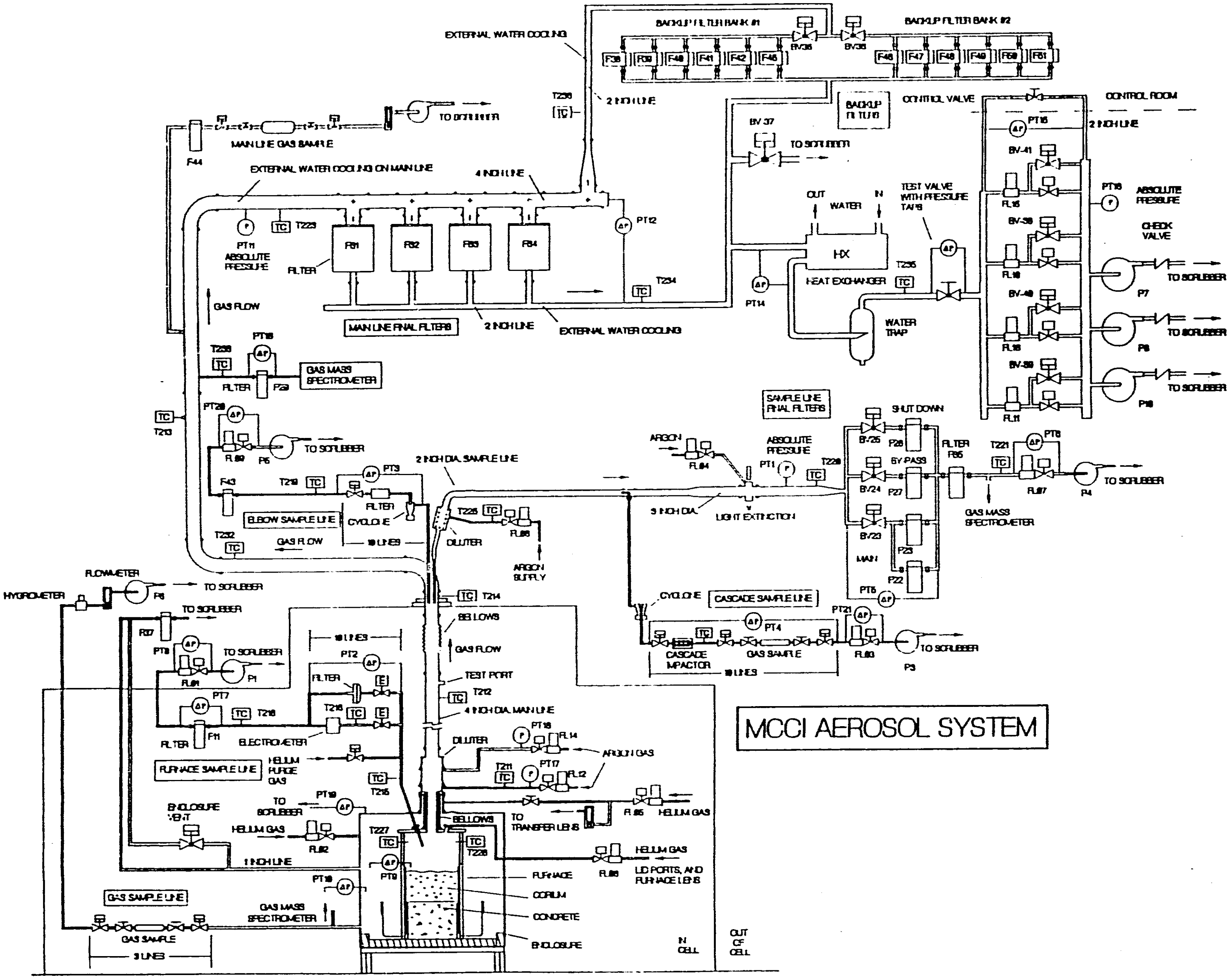

Figure 2. ACE MCCI Aerosol System for Test L7

The second isokinetic sample at the main line elbow went to the elbow sample line which contained ten cyclones followed by filters. Aerosol concentrations and compositions, and the ratio of large (mechanically generated aerosols) to small particles (vapor condensated aerosols) were determined as a function of time throughout the tests from the elbow line cyclone and filter data. Thus, variations in aerosol concentration and composition with changes during the tests such as change in concrete type, decrease in metal inventory, foaming, and change of input power were studied.

The first elbow in the main line changed the direction of flow from vertical to horizontal downstream of the sampling nozzles.. The remainder of the main line was horizontal. An on-line mass spectrometer off the horizontal main line provided gas compositions throughout each test. Flows were obtained from computer evaluation of the mass spectrometer data and the calibrated argon flow to the primary diluter. A gas sample was taken from the main line for posttest analysis using a different mass spectrometer to confirm the pre and posttest calibrations of the on-line mass spectrometer.

The furnace sample line module collected a small fraction of the gas and aerosols directly from the test enclosure without any dilution or cooling. The alumina sampling tube to this module was located low in the plenum, directly above the melt. The electrometer in the furnace sample line indicated the charge distribution on the aerosols.

The aerosol system was designed to collect the total aerosol mass released. Most of the aerosol mass was collected by large final filters and backup filters located at the end of the main line. Significant mass was also deposited in the main line. Samples for chemical analysis were selected from these filters, from deposits throughout the main line, and from sampling modules to completely characterize the aerosols. The aerosol mass distribution throughout the entire aerosol system was determined during aerosol system disassembly.

AEROSOLS RELEASED

Results of these experiments indicate that the aerosols are composed primarily of concrete decomposition products. Consequently, the aerosols released varied in concentration, composition, and form for tests with different concretes.

The major constituents in the five concretes used in these tests are given in Table 2.

Aerosol Mass Released and Concentration

A summary of the total aerosol mass and off gas concentrations for all tests is given in Table 3. Maximum aerosol concentrations in the noncondensable off gas from the MCCIs ranged from 30 g/m^3 to 4400 g/m^3. Average aerosol concentrations in the off gas ranged from 18 to 3300 g/m^3. The lowest aerosol concentrations were from Test L5 which was a fully oxidized test with limestone/common sand concrete. The highest concentrations were from MCCIs with siliceous concrete. Siliceous concrete had the lowest gas content of all the concretes. The highest mass released and highest aerosol concentrations in the off gas were from Test L6, the siliceous test with the highest metal inventory. Test L6 contained 34 kg of metal, 23 kg of which was Zr.

Aerosol Composition

The mass percent of the species of the major elements in the aerosols from each test are given in Figure 3. Most elements, other than Ag, were in oxide form. However, K and Na were also found as chlorides. Si species included SiO_2, SiC, and Si. Variations in aerosol composition and concentration with the presence of Zr in the melt were evident for all types of concrete. The effect of increased metal in the inventory on aerosol concentration and mass released is evident from comparison of Tests L2 and L6. Both tests used siliceous concrete but Test L6 had more metal. Variation in composition with the presence of Zr was most pronounced for limestone/common sand concrete. When Zr was present (L1 and L7), the aerosol compositions were high in Si; with no metal in the inventory (L5), the aerosol composition contained mainly Na and K species. In Test L5, K and Na species comprised 33 and 19%, respectively, of the aerosol mass. For Test L1, the aerosols were dominated by Si species, which comprised 81% of the aerosol mass. K and Na species each attributed only 3%.

In both limestone/common sand concrete and siliceous concrete tests with metal in the inventory, Si species dominated the aerosols. SiO_2 in the melt reacted with Zr to form SiO gas which condensed to form Si and SiO_2 or reacted with other off gases, vapors, and/or aerosols to form SiC and silicates. SiO_2, SiC, and Si have been identified in deposits from the ACE MCCI tests. In Test L4, which

Table 2. Major Constituents in Concrete

Constituent	Limestone/ Common Sand	Siliceous	Soviet		Limestone/ Limestone
			Serpentine	Ordinary	
	Mass %	Mass %	Mass %		Mass %
SiO_2	28.3	69.0	34.3	69.1	6.2
CaO	26.0	13.5	9.8	10.8	45.9
Al_2O_3	3.5	4.0	1.8	8.8	1.7
MgO	9.6	0.7	30.7	0.7	3.6
Fe_2O_3	1.6	1.0	6.4	1.7	0.9
K_2O	0.6	1.4	0.1	1.6	0.6
Na_2O	1.1	0.7	0.06	2.7	0.4
H_2O	6.1	3.68	14.4	1.9	6.1
CO_2	21.4	4.23	0.9	1.2	33.3

Table 3. Aerosol Mass Released in ACE MCCI Tests

Test	L5	L2	L1	L6	L4	L7
Aerosol Mass, g	153	455	1213	6546	4760	1077
Average Aerosol Concentration, g/m^3[a]	18	442	435	3300	395	324
Maximum Aerosol Concentration, g/m^3[a]	30	----	820	4400	1440	440
Maximum Off Gas Flow, sLpm	61	49	300	40	260	160
Metal, kg	0	13.6	13.6	34	32.2	19.6
Ablation Depth, cm[b]	10.2	3.8	4.0	8.8	12.7	----
Insert Depth, cm	----	----	----	7	7.9	5.7
Concrete Mass, kg[c]	61	23	24	72	97	16
Concrete Type	L/S	S	L/S	S	Sov	L/S

[a] In off gas

[b] Depth ablated for aerosol collection

[c] From insert & basemat ablation

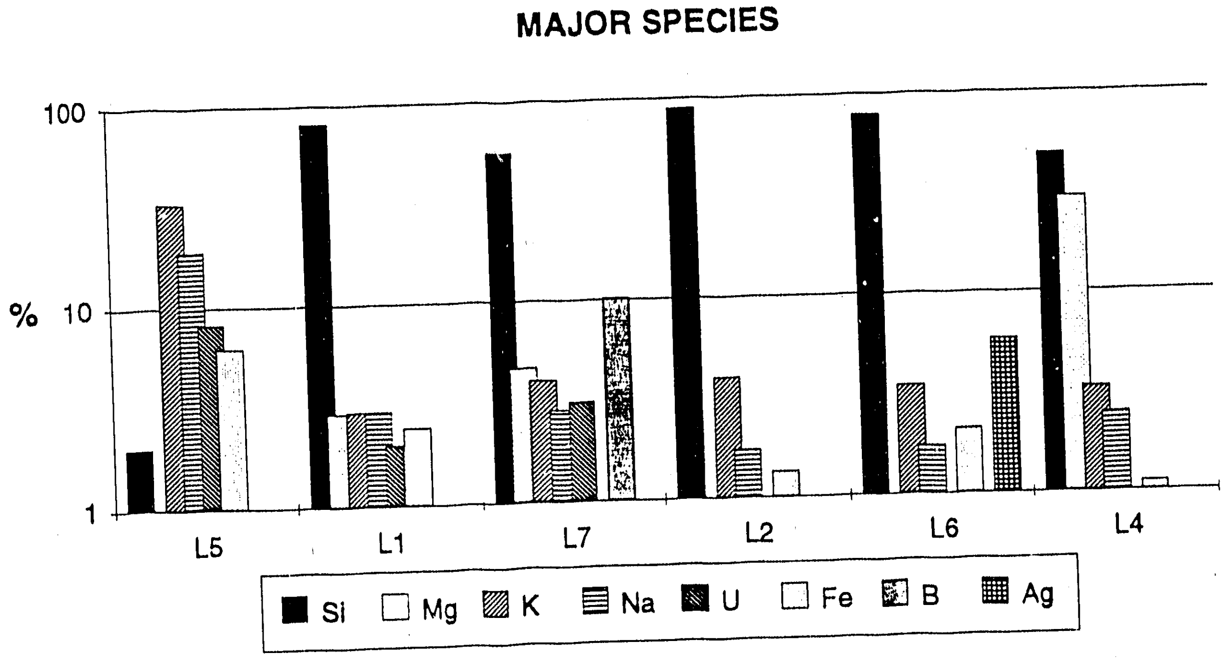

Figure 3. Mass Percent of Species of Major Elements in Aerosols in ACE MCCI Tests

contained serpentine concrete, $MgSiO_3$ and $MgSiO_4$ were identified in deposits. Silicon speciation for all ACE MCCI tests is in progress. In tests with siliceous concrete, the high concentration of SiO in the off gas and its rapid condensation in the aerosol system led to the formation of fibers or chains which then trapped other aerosols.

Effect of Concrete on Composition

The effects of concrete composition on the aerosols released is most clearly illustrated by the variation in aerosol composition as a function of time in Test L4. Test L4 employed two types of Soviet concrete in a layered basemat. A 5.1 cm layer of serpentine concrete rested on ordinary soviet concrete, which is a siliceous concrete. Serpentine concrete was used in the insert in this test. Figure 4 shows the main constituents of the aerosols released in Test L4 for 4 time periods: insert ablation, ablation of the serpentine concrete, beginning of ablation of the siliceous concrete, and end of ablation of the siliceous concrete. Although SiO_2 dominates the release at all times, SiO_2 release is highest during insert ablation, when the metal concentration is the highest. It decreases during

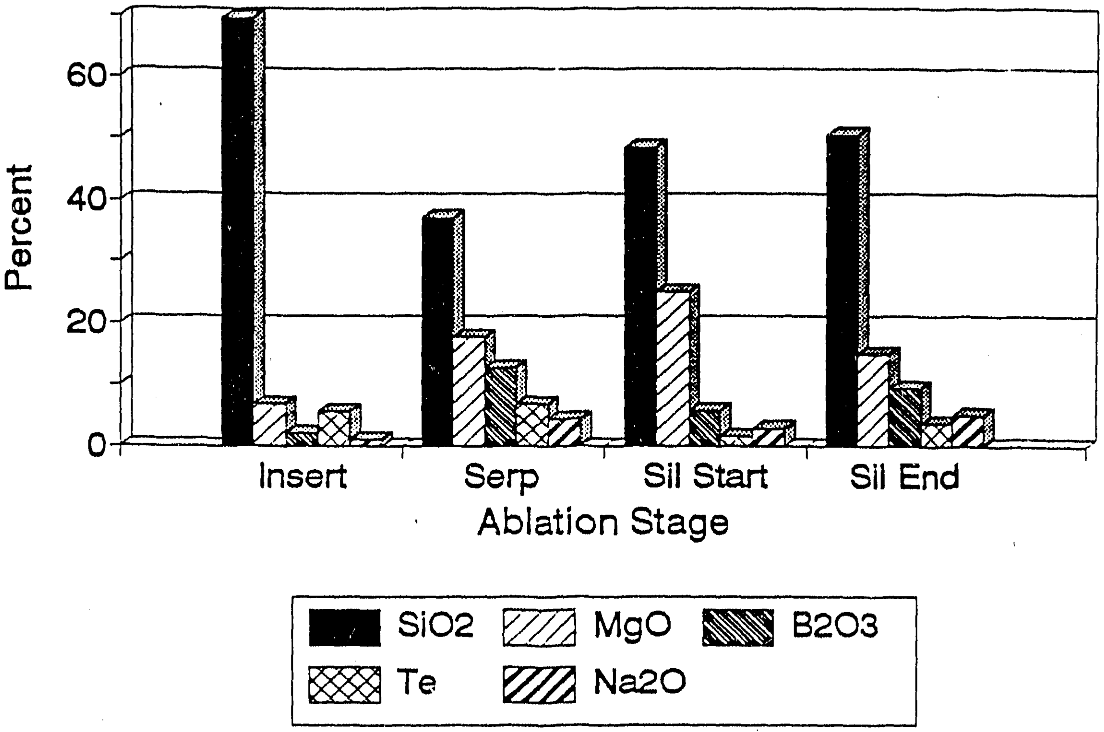

Figure 4. Mass Percent of Major Species in Aerosols Released at four Time Intervals in Test L4

ablation of the high magnesia serpentine concrete and then increases during ablation of siliceous concrete. The MgO fraction is the highest during ablation of the high magnesia serpentine concrete and at the start of ablation of the siliceous concrete.

Importance of Control Rod Materials

Inclusion of control rod materials Ag and In or B_4C also effected the aerosol composition because the release of Ag, In and B species was significant. Figure 3 shows that when Ag or B were present in the inventory, Ag or B species comprised significant fractions of the aerosol mass. Boron species (assumed to be B_2O_3) comprised 10% of the aerosol mass in Test L7; 6% in Test L4. The release fraction for boron was 0.045 in Test L7 and 0.12 in Test L4. Silver and indium were included in Tests L6 and L8. In both tests, Ag was a major constituent of the aerosols. Indium release was significant although it did not comprise as large a fraction of the aerosols. Release fractions for Ag and In in Test L6 were respectively 0.32 and 0.41.

RELEASE FRACTIONS

Except for Te, the release of fission products was small (less than 1% of the aerosol mass) in all tests. Tellurium release was close to 50% in most tests. Tellurium was detected in the aerosols primarily as a metal or metal telluride. The high Si release in all tests except L5 caused difficulty in detection of Ce and La. For tests L2, L1, L6, and L4, the Ce and La values were below the limit of detectability in most aerosol samples. Consequently, release fractions of Ce and La for these tests are maximum releases based on the limit of detectability in the chemical analysis. Neutron activation analysis for Ce and La is underway so that definitive releases may be determined. Because each test lasted a different length of time and a different mass of concrete entered the melt during aerosol sampling, comparison of total release fractions from one test to another provides little information. Consequently, in Figure 5, release fractions of the low volatility fission products and U for the first seven minutes of each test are shown. Seven minutes was chosen for comparison because that was the duration of aerosol collection in Test L1. Comparison of releases from the tests with limestone/common sand concrete (Tests L5, L1, and L7) shows that inclusion of metal in the inventory in Tests L1 and L7 increased the releases at the start of the tests. The release

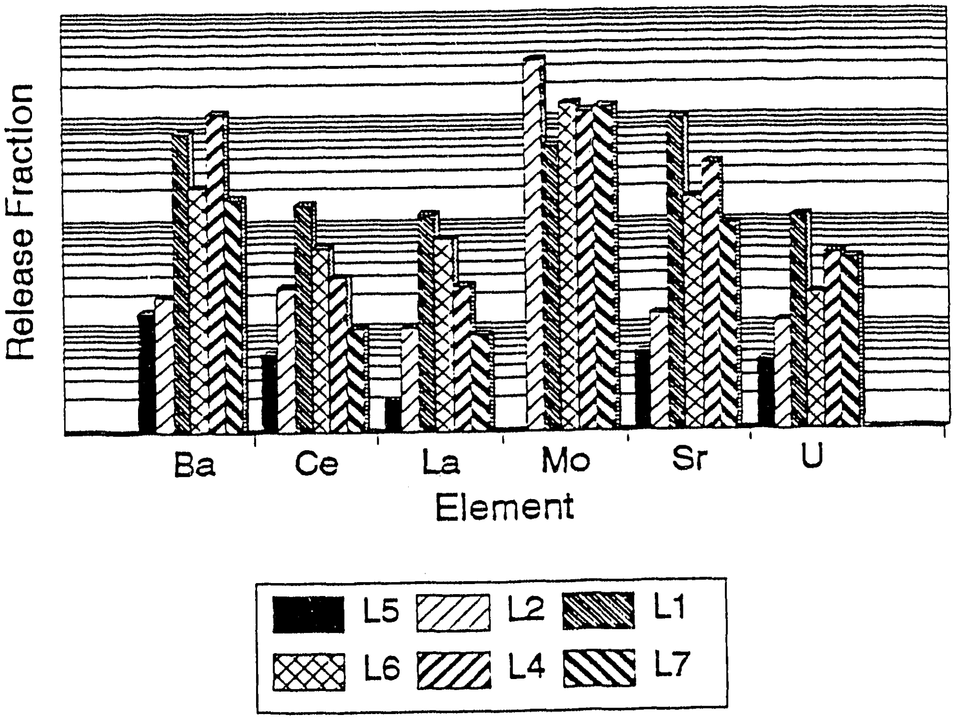

Figure 5. Relative Releases of Fission Product Elements and U for the First 7 Minutes of Each Test

fractions of Ba and Sr were significantly higher for Test L6 than Test L2. Test L6 had a higher metal content than Test L2. The release fractions of Ba, Ce, La, and Sr, were significantly below those predicted by the NRC code VANESA 1.01 [3,4].

ACKNOWLEDGMENTS

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