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H. W. Randolph[1], J. E. Payne[2], and J. L. Margrave[3]

[1]E. I. du Pont de Nemours and Company
Savannah River Laboratory
Aiken, SC 29808-0001

[2]Department of Physics
South Carolina State University
Orangeburg, SC 29115

[3]Materials Science Research Center
Houston Area Research Center
The Woodlands, TX 77380

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INTRODUCTION

The production reactors at the Savannah River Plant are powered by uranium-aluminum alloy fuel cooled by flowing heavy water. As part of current severe accident consequence analysis , hypothetical cases of total loss of cooling are being evaluated. If there is such a loss of cooling in the main reactor tank, heat from radioactive fission product decay could cause the fuel to melt and penetrate the bottom of the reactor. Molten metal, called "corium" (Slide 2) consisting of fuel, other core material, and part of the tank itself would then contact the concrete floor below the reactor and react chemically and physically to produce aerosols and other products. There is a serious need for basic experimental data on these reactions that can be incorportated into compuer codes being developed to describe accident scenarios. It is especially important to identify any unpredicted chemical compounds that might affect aerosol transport of radioactive fission products. Though studies have been done on molten metal reactions with concrete, (References 1-5) these have not used materials similar to those at Savannah River.

PROGRAM DESCRIPTION

A study of the interactions of molten uranium-aluminum corium with granitic concrete has been established at Savannah River (slide 3) and experiments are being performed at the Savannah River Laboratory , the Houston Area Reasearch Center and Rice University , and Sandia National Laboratories.The objective of the program is to gain an understanding of reactions to be expected when molten core material contacts granite aggregate concrete of the type under the reactors at the Savannah River Plant. Small scale (one to ten grams of metal) studies were designed to determine reaction chemistry when samples are heated with and without ionizing radiation. Larger scale studies with fifty to one hundred kilograms of metal were designed to gain an understanding of the physics, aerosol production and scale-up of processes.

The Savannah River Laboratory is responsible for overall program coordination and for electron beam heating of metal-concrete samples on a small scale.The houston Area Research Center and Rice University are doing research on induction and laser-heated small samples. Sandia National Laboratories work is with induction heated killogram quantities of metal and is similar to work they have done in their SWISS interaction studies using other materials.

RESEARCH EFFORTS AT THE SAVANNAH RIVER LABORATORY

In the experimental research program at the Savannah River Laboratory, samples are heated as diagramed in Slide 4 using an Airco Temescal 8kw electron beam system. The electron gun produces electrons of 10 KEV energy and deflects them magnetically through a 270 degree arc to strike the top of the sample. Beam power is typically 200 to 300 watts. Electron beam heating provides an important simulation of corium heating by providing high-energy ionization that may produce chemical reactions not caused by high temperature alone. Careful comparisons are being made between reaction products obtained by electron beam heating and heating by other methods. Slide 5 shows the laboratory setup that has been used. The apparatus allows for visual monitoring of the experiments and video recording of sample behavior and equipment displays during a run. Temperature of the sample is monitored using an optical pyrometer whose output is also recorded on the video tape.

Experiments were carried out on granite aggeregate (Slide 6) and concrete (Slide 7) samples as well as on various combinations of concrete aggregate and clay with aluminum and stainless steel. (From Slide 1 it can be seen that once the corium melts through the stainless steel vessel, aluminum and stainless steel will be two of the major components interacting with the concrete and possibly the underlying clay.) A typicial experiment would consist of drying a 15-gram sample of concrete and then placing the concrete along with a 2-gram sample of metal on top of the concrete in the vacuum chamber. The chamber was then evacuated below 10^{-4} torr and the electron beam turned on to heat the sample. Once the temperature of the sample exceeded $1500^{o}C$, there was an obvious reaction present which became more vigorous as the temperature exceeded $1800^{o}C$. There was usually a bright flare during reactions at the higher temperatures which extended several centimeters above the sample and contained sparks of hot material ejected from the surface. The shape and behavior of this flare appeared to fluctuate and probably depended on the material (aggregate or cement) on which the beam was impinging. Also, as material was removed from the spot, new material with different characteristics might have been exposed. Once the samples had cooled they were removed from the system and examined by visual and electron microscopy.

The residue of a typical experiment with stainless steel on concrete is shown in (Slide 8). The usual sample characteristics are presented here; the area where the beam impinges on the sample shows radial cracking, there is an area in the center where material has reacted to form a glasssy green mineral, and some unreacted stainless steel has melted and remains on the sample. A reaction of aluminum would produce a similar sample structure with a different clear glassy material. Some samples have finely divided metal mixed in the glass. The materials that are formed as the temperature rises above $1800^{o}C$ are similar to sapphire or topaz and are the result of metal oxides combining with SiO_2 and other minerals in the concrete. A scanning electron microscope study of concrete before and after heating with stainless steel (Slide 9) showed that metal components are present in the glass. It seems probable that the exothermic reactions are similar to the reaction (Slide 10) of aluminum with SiO_2 and CaO to form anorthite, a glassy mineral. Studies of these reactions are being continued. (Slide 11)

Initial experiments have been made with a larger 14 kW electron beam system. A residual gas analyzer (RGA) monitoring gases emitted during an experiment has shown evolution of hydrogen and acetylene during sample heating. Further experiments will be conducted to determine the source and amounts of these gases. Hydrogen could be produced by dissociation of water by the electron beam as well as by hot metal reduction of water. It is possible that the acetylene is generated by reaction between water and carbon in the concrete induced by energetic electrons. No noticeable differences in sample behavior have been observed during heating with the larger power system.

RESEARCH AT THE HOUSTON AREA RESEARCH CENTER AND RICE UNIVBERSITY

The objective of this research is to study the thermophysical and chemical properties of concrete under conditions of high temperature in a chemically harsh environment. These studies involve the interactions between liquid metals (Al, Ni) and clean, untreated concrete surfaces. The experiment performed to date have included: high-flux CO_2 laser heating of the concrete matrix material, electromagnetic levitation and melting of nickel followed by dropping of the liquid on the concrete surface, RF heating of the aluminum in contact with the concrete surface, RF heating of the aluminum in contact with an alumina surface, and electromagnetic levitaton and melting of aluminum to determine the maximum possible temperature attainable in the liquid. Two of the above experiments will be discussed in detail, the laser heating of concrete samples with the associated mass spectrometry and the RF heating of aluminum in contact with the concrete.

The laser heating experiments used a 100-Watt, cw CO_2 laser (10.6um) built by the high-temperature chemistry group at Rice University. The laser beam passed through the ionization region of the mass spectrometer and impinged on the concrete sample. Vapor species emitted from the heated sample surface were collimated by a baffle assembly and the resultant neutral beam entered the ionization chamber of the mass spectrometer. The ionization energy of the electron beam was set to 70 eV and the experiments were conducted under a vacuum of approximately 10^{-6} torr. The experiment allowed measurements on five samples with monitoring of temperature of the front surface of the sample using a Pyrometer Instrument Co., micro-optical pyrometer. As indicated by the mass spectra, (Slide 11) it is clear that the dominant vapor species was water. Two important features at $m/Z = 28$ and 44 were assigned to Si plus CO and to SiO plus CO_2, respectively.

The resolution of the mass spectrometer was not sufficent to distinguish between these pairs of vapor species but it is probable they were all present in the vapor. Oxides , hydroxides, and carbonates of the metals K, Na, Mg, Ca, Fe, and Al were observed, but were not important components of the vapor. Related species observed were $NaCO_2$ and $CaCO_2$. Elemental species were C, K, K_2, and Fe with the carbon atom being more abundant than the other three. The consistent presence of the carbon atom in all mass spectra gives additional evidence that carbonates as well as metal complexes of the ions CO_2^- and $CO_2^=$, were present in the vapor. The carbon atom would be a fragmentation product of these species.

Some samples were heated and visual and photographic observations were made of the resulting effects on the concrete. While the laser heated the sample, a blue-white flame projected perpendicular to the sample surface to a distance of about 1 cm. Sparks were associated with the flame much like those described in the electron beam experiments. It was observed that the shape and structure of the flame depended on the material in the concrete on which the laser was impinging. Slide 13 shows a typicial concrete sample after laser heating.

In experiments using induction heating of aluminum on concrete, a cylindrical sample of high-purity aluminum was placed on the concrete sample and positioned with the metal centered inside an RF induction coil. Argon purge gas was turned on to shield the sample from air. RF power was turned on to melt the metal which then heated the concrete by conduction. Sample temperature was monitored using a pyrometer and was recorded using a high-speed camera provided by Du Pont. After the experiments were complete, the concrete sample was removed and photographed at 10 - 20 X in the vicinity of the metal. (Slide 14) These experiments gave visual results similar to those from the electron beam experiments. Several points of interest are to be noted. First, and most obvious, is the black crater resulting from the liquid aluminum reaction and the bubbles resulting from the passage of volatile gases through the liquid material. The peripheral region of the crater shows glassy material, seen in the electron beam experiments, interspaced with a white chalky material that formed if the experiments were not purged carefully. Observations indicate liquid aluminum will not universally attack concrete. Rather, certain constituents react more strongly than others. Interfaces between aggregate and matrix materials as well as thermal stress cracks provide channels through which liquid metal can flow and react with material in the concrete. Experiments with liquid Ni on concrete indicate that it is relatively unreactive toward concrete. The nickel appeared to wet the granite aggregate but little reaction occurred.

RESEARCH EFFORTS AT SANDIA NATIONAL LABORATORIES

The research effort at Sandia National Laboratories involves inductively heating a large cylinder of metal to high temperature in contact with concrete of the same type used in the reactors at the Savannah River Plant. Heating is continous to simulate fission product heat in reactor corium. A preliminary scoping test has been completed in which a 30 kg aluminum cylinder was heated in contact with basaltic concrete. At 1400^{o} C the temperature began to increase and continued to increase after RF heating was turned off. A considerable amount of flame was generated from burning hydrogen. A relatively small amount of smoke and aerosol were generated. The reaction residue was an earthy gray material similar to lava and contains pockets of aluminum and finely divided aluminum. Though the basaltic concrete is not exactly the same as granitic concrete it is sufficiently similar to expect much the same reactions in both concretes.

The SRP type concrete has been cast for the test and the aluminum ingot is fabricated. The material to simulate corium is established (Table1). In Slide 15 the basic design of the experiment is detailed. As shown, the metal is heated to the desired temperature and sustained at this temperature for a length of time to qualify the melt. The material is then poured onto the concrete surface and the resulting reactions are monitored. These experiments will allow for large-scale measurements that are not possible in the work being conducted at the other two locations.

FUTURE EFFORTS

Savannah River Laboratory

The experiments conducted to date have given excellent qualitative results. The continuing program will involve more electron beam experiments using the 14kW system with monitoring of off-gases during the experiments. Efforts will be made to identify mineral species in the reacted samples and to measure the molar amounts of metal reacted and of gases generated during reaction.

Houston Area Research Center and Rice University

The basic experiments on metal-concrete interactions will be continued. Studies of the thermal conductivity of concrete will be undertaken. In addition, a study of the direct interaction of various liquid metals with liquid water and water vapor will be done since there will certainly be water and water vapor present in the case of a meltdown.

Sandia National Laboratories

The experiments at Sandia will continue with aluminum and simulated fission products on concrete, simulated corium on dry concrete and simulated corium on concrete covered with water.

CONCLUSION (Slide 8)

Electron beam heated samples of aluminum and other metals on Savannah River concrete produced exothermic reactions in which the metal became incorporated in a glassy mineral residue and moderate amounts of gas were evolved. Laser heating of concrete and induction heating of metals on concrete produced no unexpected species in gases evolved and left reaction residues similar to electron beam heated samples. Induction heating of 30 kg of aluminum on basaltic concrete produced a highly exothermic reaction with hydrogen evolution and moderate aerosol production. The dominant chemical reactions involved with the granitic concrete are thought to be similar to the reaction in which aluminum oxidizes exothermically and the oxide reacts again exothermically with silica and calcium oxide to produce the mineral anorthite. Studies will continue to determine reactions of simulated corium with granitic concrete, quantify hydrogen generation, and measure high-temperature properties needed for severe accident computer codes.

ACKNOWLEDGMENT

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E. I. DU PONT SAVANNAH RIVER PLANT
HENRY W. RANDOLPH

SOUTH CAROLINA STATE UNIVERSITY
J. E. PAYNE

HOUSTON AREA RESEARCH CENTER
J. L. MARGRAVE

SLIDE #2

MATERIAL COMPOSITION OF SIMULATED SRP CORIUM

MATERIAL	MASS GRAMS	PERCENTAGE WT %
Al	75000.00	92.17
U	4065.86	5.00
S.STEEL	991.26	1.22
Cd	235.22	0.29
Zr	206.65	0.25
Ce	176.75	0.22
Nd	162.97	0.20
Mo	142.47	0.18
Ru	91.40	0.11
Ba	71.24	0.09
Sr	69.89	0.09
La	64.18	0.08
Pr	50.07	0.06
Tc	42.34	0.05

SLIDE#3

MOLTEN CORE-CONCRETE PROGRAMS

OBJECTIVE:

- o GAIN AN UNDERSTANDING OF THE CHEMISTRY AND PHYSICS OF REACTIONS OF MOLTEN CORE MATERIAL WITH SRP-TYPE CONCRETE THROUGH EXPERIMENTAL STUDIES

PROGRAM:

o SRL The experiments were carried out on concrete samples and various other

- PROGRAM COORDINATION, SMALL-SCALE ELECTRON BEAM SAMPLE HEATING IN VACUUM

o RICE UNIVERSITY

- SMALL-SCALE INDUCTION AND LASER HEATING OF SAMPLES IN AIR

- STEAM EXPLOSION STUDIES

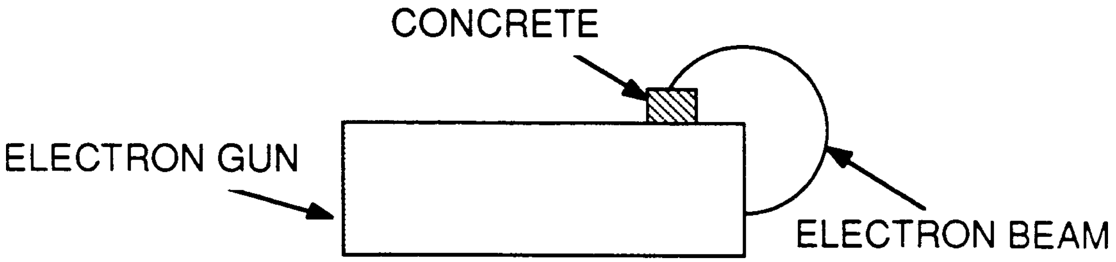
o SANDIA NATIONAL LABORATORIES

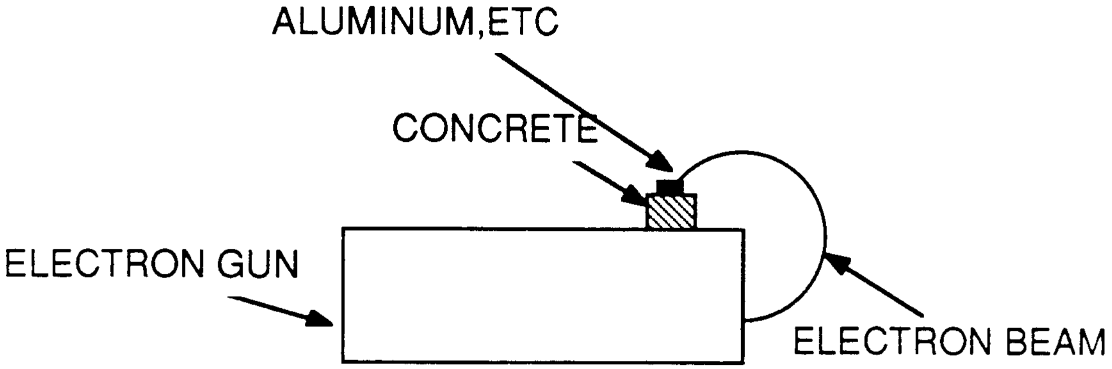
- LARGE-SCALE MOLTEN METAL-CONCRETE REACTION STUDIES

- COMPUTER CODE MODIFICATIONS AND PREDICTIONS

SLIDE#4

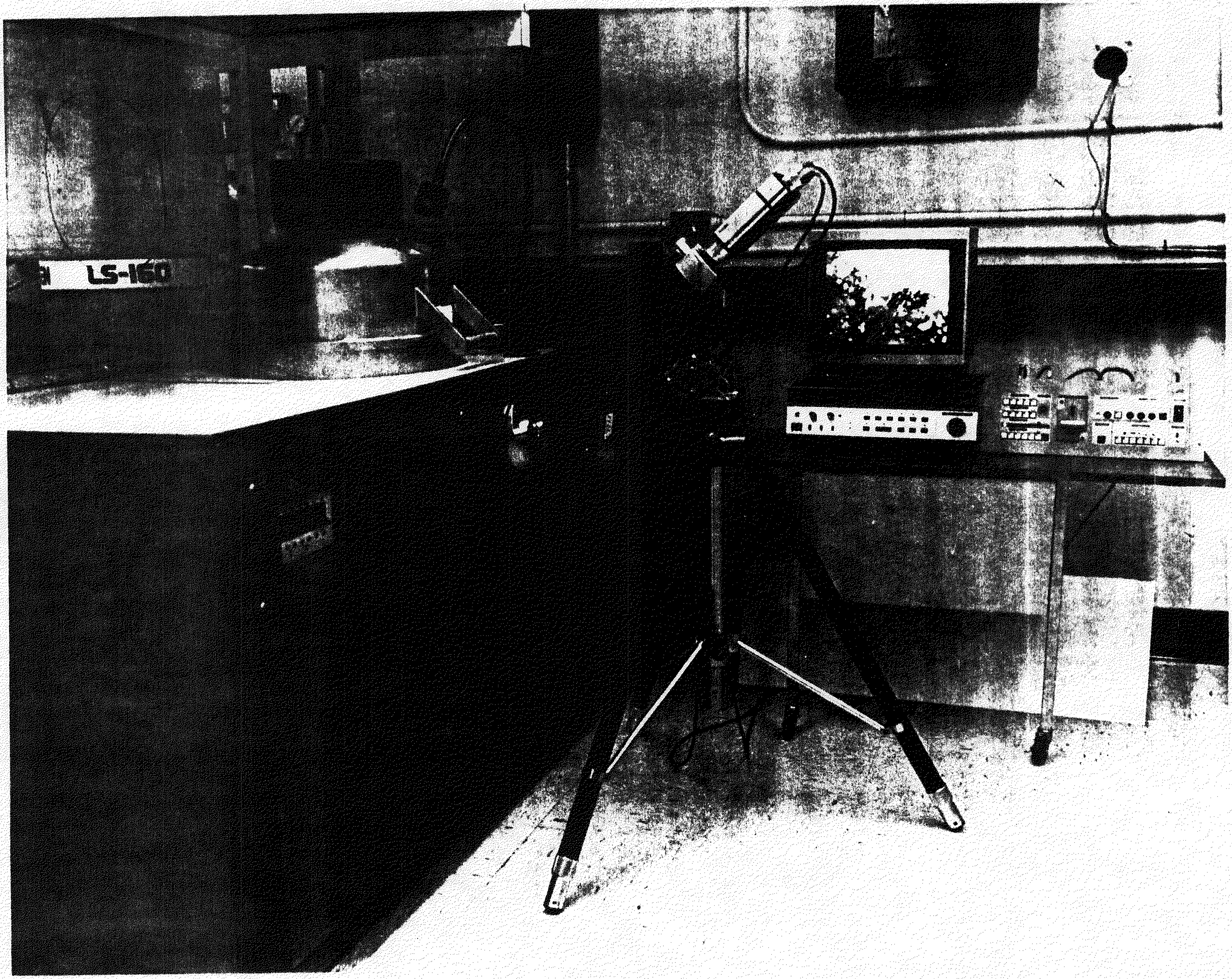
ELECTRON BEAM STUDIES





SLIDE #5

8 KW ELECTRON BEAM EXPERIMENTAL SYSTEM


LS-160

SLIDE #6

PETROGRAPHIC ANALYSIS OF 1961 AGGREGATE

MINERAL	PERCENT (WT)	COMPOSITION
QUARTZ	65%	SiO_2
SODIC ANDESINE	10%	$Na(Al,Si)(AlSi_2O_8)$
MICROCLINE	10%	$K(AlSi_3O_8)$
CHLORITE	6%	$(Mg,Fe,Al)_6(OH)_8[(Al,Si)_4O_{10}]$
SPHENE	2%	$CaTi\ SiO_5$
PYRITE	5%	FeS_2,Ni,FeS_2
EPIDOTE	2%	$Ca_2(Al,Fe)Al_2O(OH)(SiO_4)(SiO7)$
OTHERS	TRACE	(SiO2)

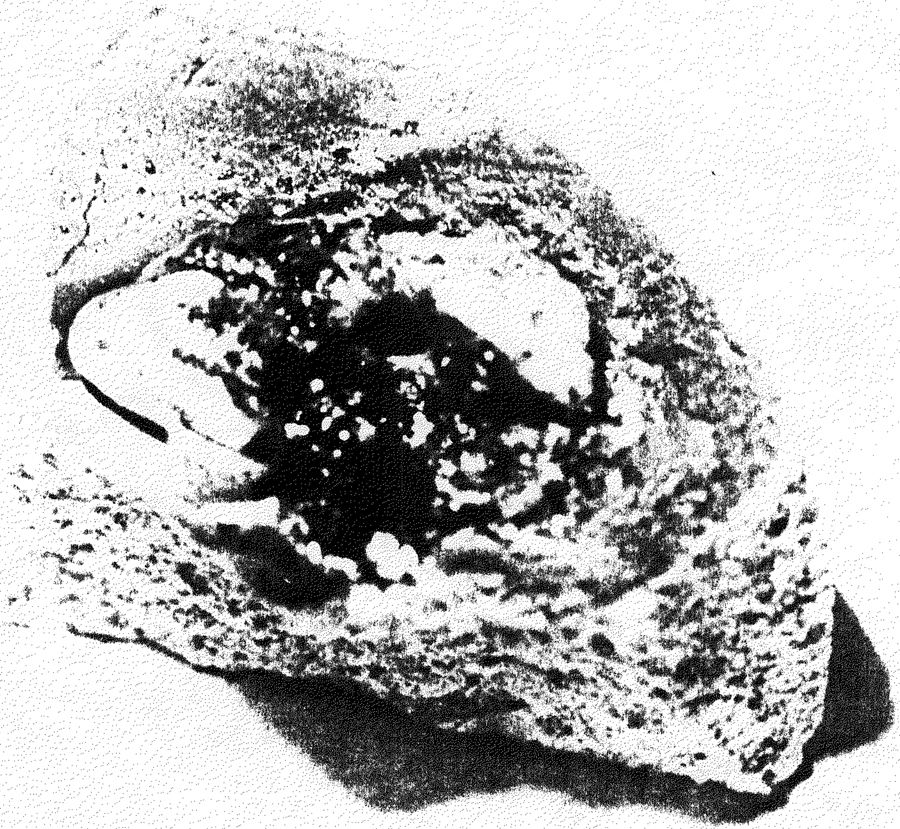
SLIDE #7

PORTLAND TYPE II CEMENT

COMPOUND	PERCENTAGE
CaO	63.6
SiO_2	21.7
Al_2O_3	4.7
Fe_2O_3	3.6
MgO	2.9
SO_3	2.4

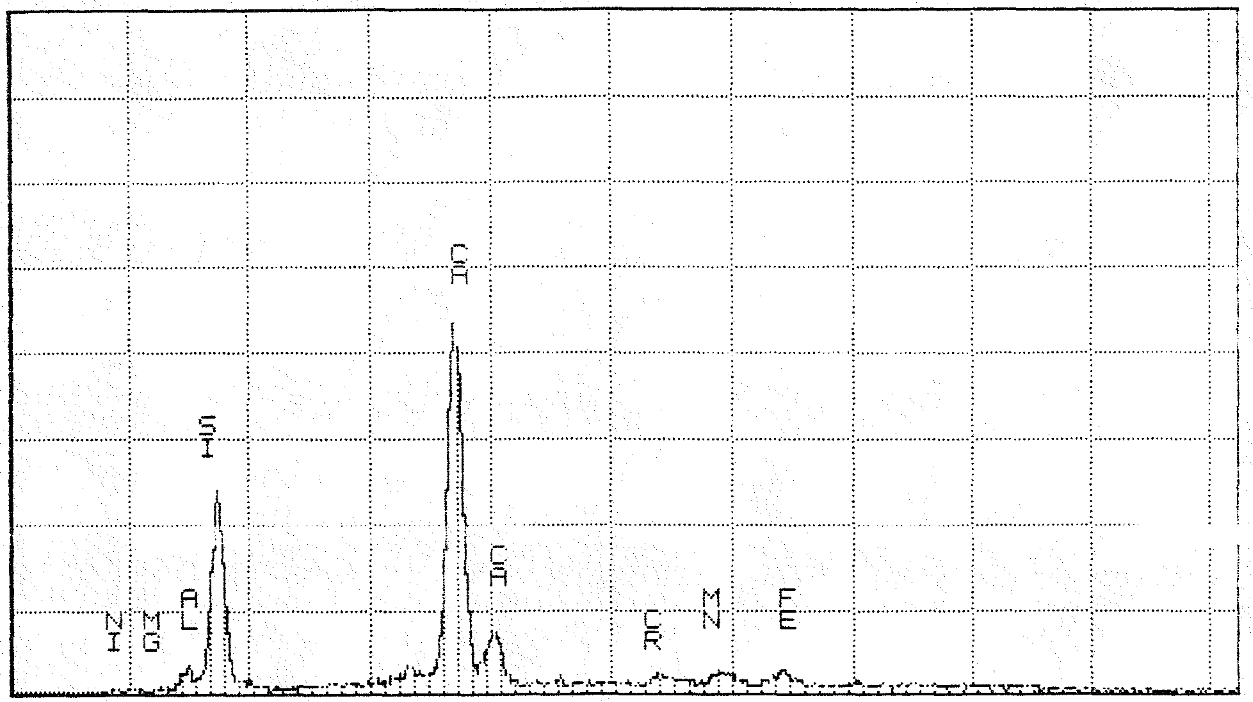
SLIDE #8

CONCRETE WITH STAINLESS STEEL HEATED BY ELECTRON BEAM



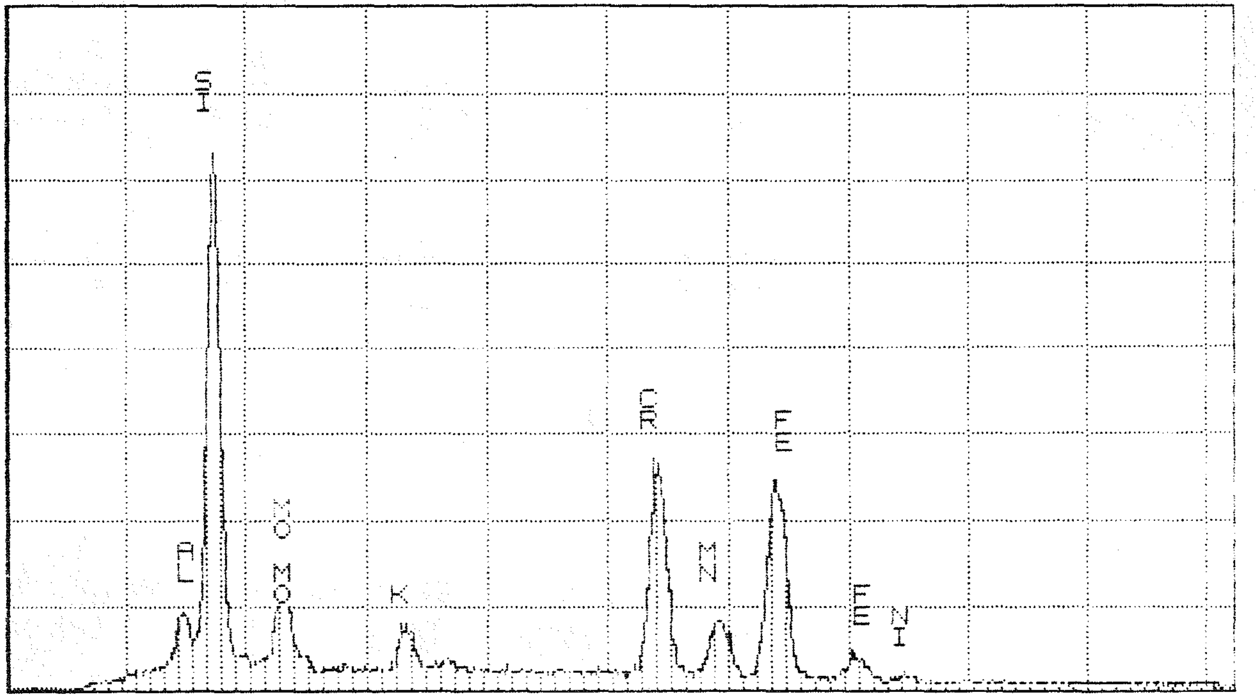
SLIDE #9

SRL Scanning Electron Microscope , C-112   FRI 15-JUL-88   12:50
Cursor: 0.000KeV = 0



0.000   VFS = 1024   10.240
30   SAMPLE #5 CONCRETE ZONE

SRL Scanning Electron Microscope , C-112   FRI 15-JUL-88   12:57
Cursor: 0.000KeV = 0



0.000   VFS = 1024   10.240
30   SAMPLE #5 SS

SLIDE #10

REPRESENTATIVE REACTION

$$4Al + 3O_2 \text{ ----> } 2Al_2O_3 + 800{,}000 \text{ CAL}$$

$$CAO + AL_2O_3 + 2SiO_2 \text{ ---> } \underset{\text{ANORTHITE}}{CaAl_2Si_2O_8} + 16{,}900 \text{ CAL}$$

SLIDE #11

SOME SILICATE MINERALS

TYPE	MINERAL	COMPOUND
NEOSILICATES	FAYALITE	$Fe_2(SiO_4)$
	MONTICELLITE	$CaMg(SiO_4)$
	ANORTHITE, KYANITE	$Al_2O(SiO_4)$
	TOPAZ	$Al_2(OH)(SiO_4)$
	STAUROLITE	$Fe_2Al_9O_7(OH)(SiO_4)$
TEKTOSILICATES	QUARTZ	SiO_2
	ORTHOCLASE	$K(AlSi_3O_8)$
	PLAGIOCLASE/AND RTHITE	$Ca(Al,Si)(AlSi_2O_8)$
	SODALITE	$Na_4Cl(AlSiO_4)_3$
PHYLLOSICATES	MUSCOVITE	$(K,Na)Al_2(OH)_2(AlSi_3O_{10})$
	BIOTITE	$K(Mg,Fe)_3(OH)_2(AlSi_3O_{10})$
	CHLORITE	$(Mg,Fe,Al)_6(OH)_8[(Al,Si)_4O_{10}]$
INOSILICATES	AUGITE	$Ca(Mg,Fe,Al)[(Al,Si)_2O_6]$
	HORNBLENOE	$(Na,Ca)_2(Mg,Fe,Al)_5(OH)_2(Si,Al)_8O_{22}$
	DIOPSIDE	$CaMg(Si_2O_6)$

SLIDE #12

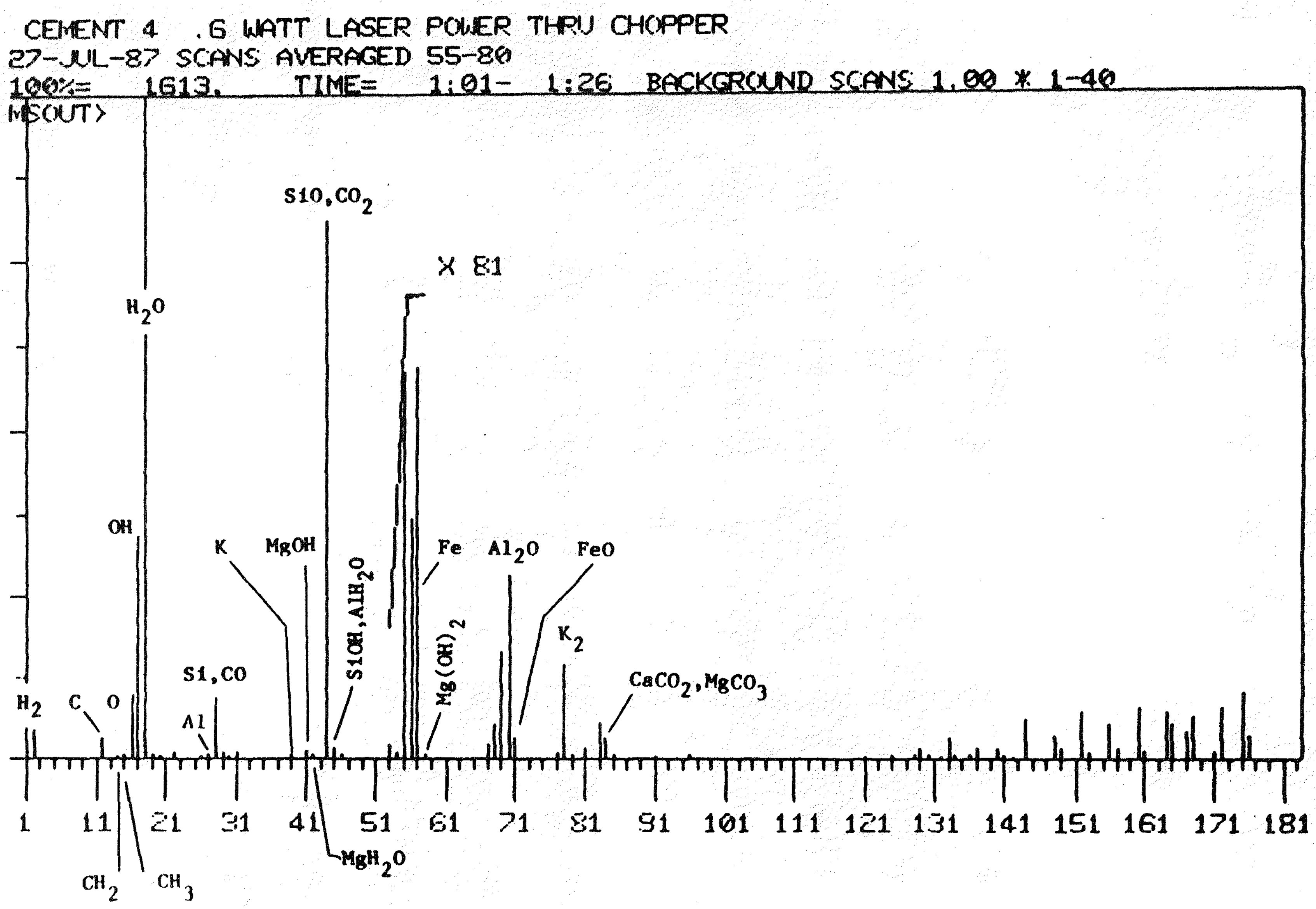

CEMENT 4 .6 WATT LASER POWER THRU CHOPPER
27-JUL-87 SCANS AVERAGED 55-80
100%= 1613. TIME= 1:01- 1:26 BACKGROUND SCANS 1.00 * 1-40
MS(OUT)>
X 81
H_2
C
O
CH_2
CH_3
OH
H_2O
Al
Si,CO
K
MgOH
SiO,CO_2
MgH_2O
$SiOH,AlH_2O$
Fe
$Mg(OH)_2$
Al_2O
FeO
K_2
$CaCO_2,MgCO_3$
1
11
21
31
41
51
61
71
81
91
101
111
121
131
141
151
161
171
181

FIGURE 12

SLIDE #13
CONCRETE HEATED BY LASER

Figure 5 (Magnification: 11.3 X)

SLIDE #14

CONCRETE WITH ALUMINUM HEATED BY INDUCTION

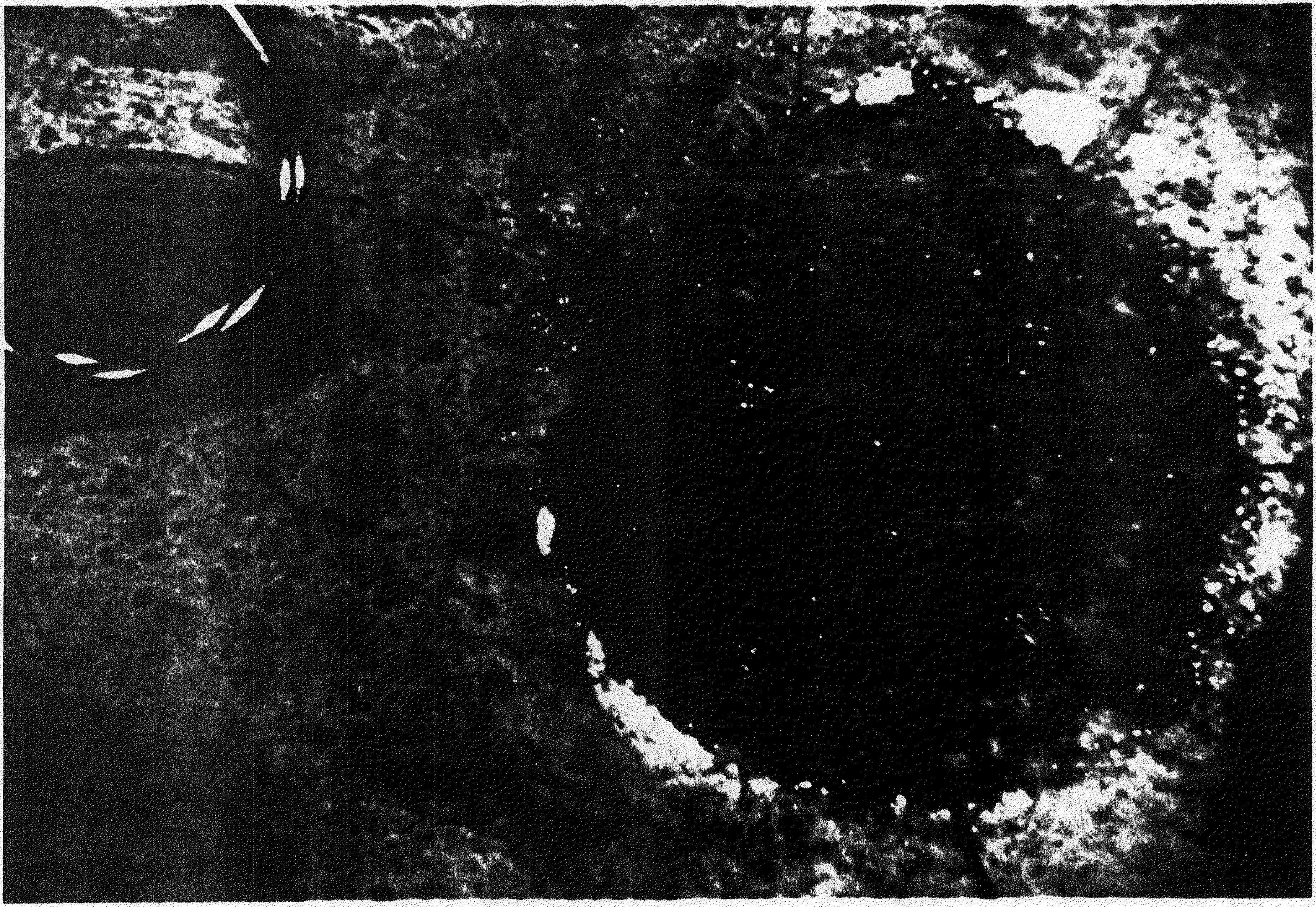
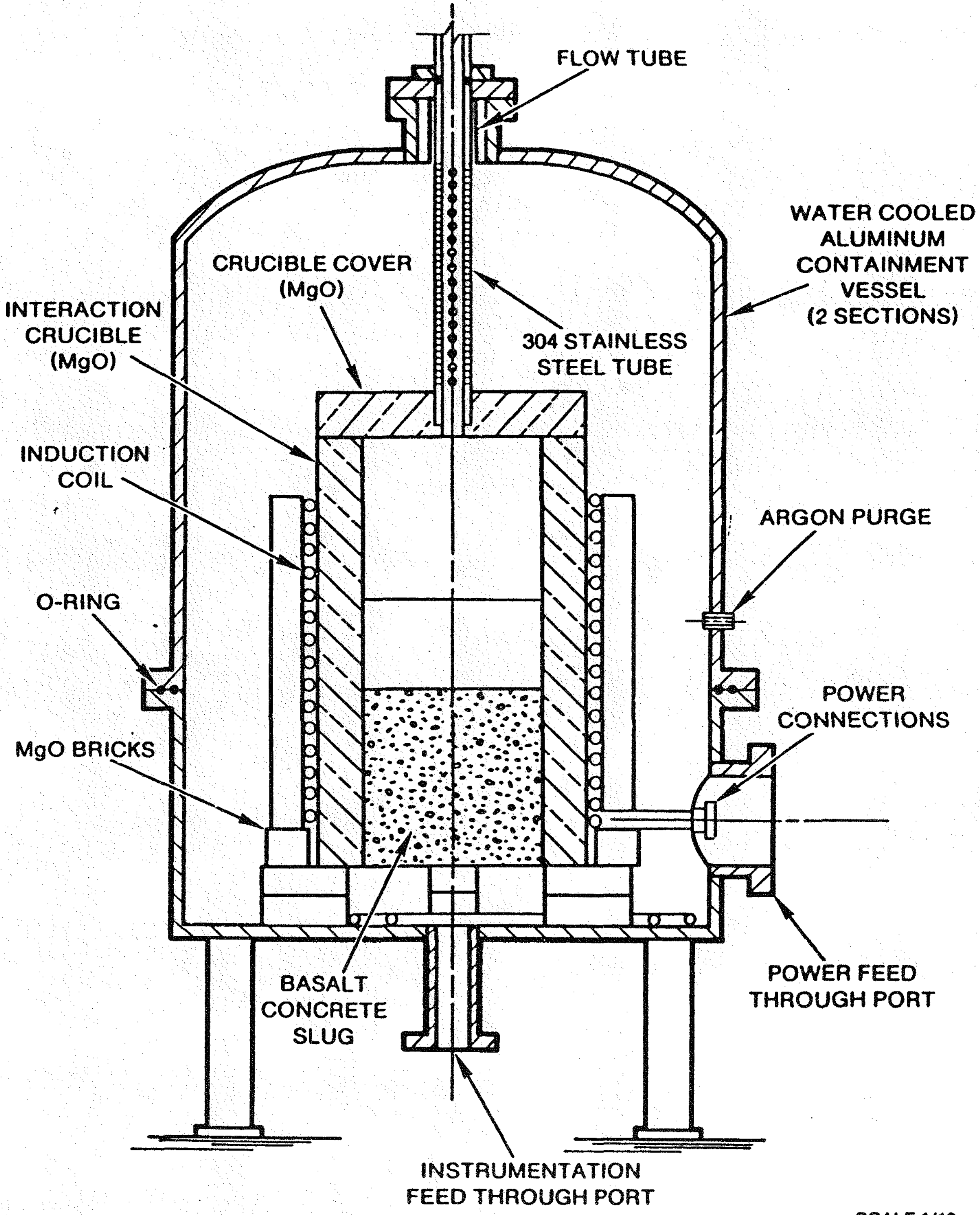


Figure 8 (Magnification: 5.2 X)

SLIDE #15

SANDIA INDUCTION HEATING EXPERIMENT


FLOW TUBE
WATER COOLED ALUMINUM CONTAINMENT VESSEL (2 SECTIONS)
CRUCIBLE COVER (MgO)
INTERACTION CRUCIBLE (MgO)
304 STAINLESS STEEL TUBE
INDUCTION COIL
ARGON PURGE
O-RING
POWER CONNECTIONS
MgO BRICKS
BASALT CONCRETE SLUG
POWER FEED THROUGH PORT
INSTRUMENTATION FEED THROUGH PORT

SCALE 1/10