



IN9800167

BARC/1997/E/021

BARC/1997/E/021



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BEARING HIGH LEVEL WASTE

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Process Engineering and Systems Development Division

1997

29-11

GOVERNMENT OF INDIA
ATOMIC ENERGY COMMISSION

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BIBLIOGRAPHIC DESCRIPTION SHEET FOR TECHNICAL REPORT

(as per IS : 9400 - 1980)

01	Security classification :	Unclassified
02	Distribution :	External
03	Report status :	New
04	Series :	BARC External
05	Report type :	Technical Report
06	Report No. :	BARC/1997/E/021
07	Part No. or Volume No. :	
08	Contract No. :	
10	Title and subtitle :	Development of melt compositions for sulphate bearing high level waste
11	Collation :	30 p., 11 figs., 6 tabs.
13	Project No. :	
20	Personal author (s) :	P.B. Jahagirdar; P.K. Wattal
21	Affiliation of author (s) :	Process Engineering and Systems Development Division, Bhabha Atomic Research Centre, Mumbai
22	Corporate author (s) :	Bhabha Atomic Research Centre, Mumbai-400 085
23	Originating unit :	Process Engineering and Systems Development Division, BARC, Mumbai
24	Sponsor(s) Name :	Department of Atomic Energy
	Type :	Government
30	Date of submission :	August 1997
31	Publication/Issue date :	September 1997

contd...2

40 Publisher/Distributor : Head, Library and Information Division,
Bhabha Atomic Research Centre, Mumbai

42 Form of distribution : Hard Copy

50 Language of text : English

51 Language of summary : English

52 No. of references : 5 refs.

53 Given data on :

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70 Keywords/Descriptors : HIGH-LEVEL RADIOACTIVE WASTES; VITRIFICATION; BOROSILICATE GLASS; CHEMICAL COMPOSITION; LEAD OXIDES; DIFFERENTIAL THERMAL ANALYSIS; LEACHING; SPECIFIC HEAT; THERMAL CONDUCTIVITY; SODIUM SULFATES; URANIUM; URANIUM 238; RADIOACTIVE WASTE PROCESSING

71 INIS Subject Category : E5100

99 Supplementary elements :

DEVELOPMENT OF MELT COMPOSITIONS FOR SULPHATE BEARING HIGH LEVEL WASTE

ABSTRACT

The report deals with the development and characterization of vitreous matrices for sulphate bearing high level waste. Studies were conducted in sodium borosilicate and lead borosilicate system with the introduction of CaO, BaO, MgO etc. Lead borosilicate system was found to be compatible with sulphate bearing high level wastes. Detailed product evaluation carried on selected formulations is also described.

BACKGROUND

Management of high level wastes from the reprocessing of spent fuel has been a matter of great concern due to the hazards posed to the man and his environment on account of long lived radionuclides present in these wastes. Many matrices are under development and evaluation, but there is a growing consensus in favour of vitreous matrices for the immobilisation of high level wastes. Apart from good compatibility and favourable long term behaviour of these vitreous waste forms, techno-economic considerations also favour their choice as an immobilisation matrix for high level wastes.

Glass as a matrix has received attention because it has a number of desirable chemical and physical properties such as good chemical durability, good mechanical strength, good radiation stability etc. Besides technology of producing massive glass blocks is available. Glasses do not have regular lattice structure in the arrangement of constituent atoms like Silicon, Boron, Oxygen etc. These atoms are however, strongly bonded to one another as a single continuous, three dimensional network. It is this property of glass that is responsible for its ability to accommodate a wide variety of different elements in the structure. One important consideration guiding the development of glass matrix formulation is its ability to accommodate and retain varying amounts of radio nuclides present in the waste. Extensive studies carried out on various vitreous systems reveal that Sodium borosilicate system is the best in this regard. However, it was also found that there are certain constituents in the waste that cannot be accommodated with ease in Sodium borosilicate glass formulations.

High level waste is the first cycle raffinate from spent fuel reprocessing and it consists of intensely radio-active fission products, corrosion products and certain amount of actinides and process chemicals. The presently stored high level waste at Trombay is characterized by higher levels of Uranium, Sodium and Sulphate in addition to the above constituents Table I gives details of the waste characteristics.

The Sulphate in the waste is derived from ferrous sulfamate which was used earlier as a reducing agent for conversion of Pu^{+4} to Pu^{+3} during partitioning stage of reprocessing. Sulphur in the form of Na_2SO_4 can be accommodated only to a limited extent (1wt %, max.) in the sodium borosilicate melts.(1) At higher sulphate concentration, a separate phase of alkali sulphate known as "gall" is formed. Its presence in the glass is not desirable as this phase is enriched with respect to cesium and has high solubility in water. Besides, sulphate in the melt tends to volatilize during vitrification process resulting in its carry over into the off-gas system. It is therefore, necessary to modify the glass forming system to ensure homogeneous distribution of sulphate in the matrix with minimum volatility during vitrification process.

Uranium retention in the matrix assumes significance in view of very long half life of U-238. It is desirable that the matrix should accommodate all the uranium present in the waste and strongly resist leaching of uranium under a wide variety of conditions.

Presence of sodium, to a certain extent in glass, is helpful in lowering the formation temperature and to reduce the viscosity of the melt. This is on account of loosening of the glass structure wherein non-bridging oxygens are formed. However, presence of sodium beyond certain limits contributes to significant lowering of chemical durability of matrix. Hence sodium content in the glass forming system is kept within limits to ensure that desired properties of glass remain unaffected.

It can be seen from the literature that stability of sulphate compounds is enhanced if the associated cation has a large radius and a low charge. Thus most alkali and alkaline earth sulfates exhibit good thermal stability up to 950 deg C.(Fig I) (2&3) However, alkali sulfates suffer the drawback of high solubility in water whereas alkaline earth sulphates are found to combine high thermal stability and extremely low solubility in water. Melting temperature of Na_2SO_4 is 884 deg C with a solubility of 19.4 g/100 ml water. CaSO_4 melts at 1450 deg and has a solubility of 0.298 gm/100 ml of water. BaSO_4 melts at 1580 deg C and has a solubility of 0.000285 g/100ml of water.(4)

APPROACH

One of the approaches to vitrify these wastes with high concentrations of sodium, uranium and sulphate in sodium borosilicate system is to use alkaline earth oxides as additives. Al_2O_3 , MnO , TiO_2 etc are known to enhance chemical durability and hence these oxides are added in conjunction with alkaline earth oxides to combat the presence of sulphate and high sodium content. However, studies in our laboratory showed that these formulations were found to lack homogeneity and gave products of high leach rates. Sulphates react with PbO to form PbSO_4 which has a melting temperature of 1170 degC, Its solubility in water is very low (0.0001 gm/100 ml of water). Compatibility of PbSO_4 with silicate matrix is also good. Considering these facts,, lead borosilicate system was chosen for investigation for containment of sulphate bearing waste with high

sodium content. Preliminary studies on lead borosilicate system showed that it has definite advantage over sodium borosilicate system with alkaline earth oxides as additives. Accordingly, detailed studies were taken up in lead borosilicate system to arrive at a suitable composition. Details of these studies are presented below.

EXPERIMENTAL STUDIES

All compositional development studies were conducted using inactive simulated waste. Table I gives the compositional details of the waste which was simulated using nitrate salts of the constituents. Sulphate was introduced in the form of sodium sulphate and balance of sodium was made up by the addition of NaNO_3 .

Experiments were carried with the aim of identifying compositions having desired pour temperature with good homogeneity and high resistance to aqueous attack. A known volume of waste was mixed with required amount of additives comprising of glass formers such as SiO_2 , B_2O_3 etc. and network modifiers like CaO , BaO , TiO_2 etc. and were mixed intimately and dried under IR lamp. Dried mass was crushed to a fine powder and transferred to a fire clay crucible. The powdered mass was first heated at 800°C for two hours and further heated at the interval of 50°C and kept for two hours at each temp. till the glass became pourable. State of fusion and pourability, presence of air bubbles and distinct separation of phase if any, were recorded. After cooling, the glasses were tested for their homogeneity by visual observation and subsequently by optical microscopy.

Preliminary investigations on leaching were carried using static leach tests in a conventional boiling water unit where in glass grains of -16+25 BSS mesh size were exposed to the attack of boiling distilled water under total reflux conditions. Water was replaced after first four hours, next 20 hours and subsequently after every 24 hours during the week..

Wt. loss suffered during the given time interval was determined. Leached out sodium out during the above intervals were determined by analysis of the corresponding leachate samples by using flame photometry. Leach rates were calculated both on wt. loss and sodium loss basis. Only those glasses that showed relatively less leaching were taken up for further evaluation.

During these studies, waste oxide loading was kept in the range of 25 to 35 wt % and other glass forming additives were accordingly adjusted. Silica being a glass former was not reduced below 25 wt. % as its further lowering would adversely affect the leaching behaviour of the glass. Incorporation of silica above 40% would result in higher pour temperature beyond 1100°C which is not desirable from process considerations.

All the melts developed with the objective of incorporating sulphate in various borosilicate systems as given in Table II can be grouped as under:

Group I:

Incorporation of CaO, BaO, MgO etc as one of the alkaline earth oxide up to maximum of 15 wt % in sodium borosilicate system.

Group II A:

Same as group I and further addition of chemical durability enhancers like Al_2O_3 , TiO_2 , MnO etc. up to a maximum of 10 wt %.

Group II B:

Same as group IIA with the addition of PbO up to a maximum of 15 wt.% in lieu of alkaline earth oxide to ensure sulphate compatibility.

Group III:

Incorporation in lead borosilicate system with the total elimination of external addition of Na_2O and alkaline earth oxides and addition of Fe_2O_3 in lieu of them.

RESULTS & DISCUSSIONS

All compositions in group I (Sodium borosilicate system with alkaline earth oxide & waste oxide) were pourable at or below $1000^{\circ}C$. Most of the glasses on visual examination gave the evidence of phase separation. Short term static leach tests by conventional boiling water method showed that these glasses were characterised by poor chemical durability with cumulative fraction leached (CFL) on wt. loss basis ranging from 0.065 to 0.2722.

In an effort to improve the chemical durability of the above glasses, minor additives like TiO_2 , Al_2O_3 , MnO etc. were added and are termed under Group IIA. This exercise was not found successful since these were found to give phase separation and low chemical durability. High leachability of the above sodium borosilicate compositions could be attributed due to the formation of soluble phase formed on account of sulphate present in the waste.

Lead oxide is known for its homogenising effect for sulphate into the glass matrix. Besides, glasses incorporating PbO are known to have relatively lower pour temperature. Compositions in group IIB contained PbO up to 10wt%. They also contained durability enhancers like BaO, Al_2O_3 , MnO etc. in varying proportions. The pour temperatures of these compositions were in acceptable limits and the products exhibited fair homogeneity in most cases. However, leach test results were not encouraging. CFR (Cumulative fraction released) values were not low enough and further improvisation of the compositions in this system was not considered worthwhile. Sodium borosilicate system with the incorporation of alkaline earth oxides and durability enhancers, like

PbO etc. did not result in an acceptable composition to accommodate sulfate bearing high level waste. It can be seen that with 18 g/l of sodium oxide in the waste, the contribution of sodium oxide to the waste oxide works out to 28.6%. In addition to this, the formulated compositions also contain 10-15 wt% of Na₂O which is added externally. Extremely high total sodium content in the matrix could be the reason for not achieving lower CFR values despite the addition of durability enhancers and homogenizing components.

An alternative route followed was to formulate the compositions by replacing externally added Na₂O by Fe₂O₃ in lead borosilicate system. The choice of Fe₂O₃ is on account of its ability to lower the pour temperature and viscosity without impairing the leach resistance when incorporated up to a certain limit. A series of such compositions were formulated and studied. These are listed under group III in Table II. Summary of observations during melting of these compositions is as follows:

- a) Higher SiO₂ leads to higher pour temp, which is in keeping with the general trend.
- b) Higher PbO content leads to lowering of pour temperature. This is on account of the fact that PbO acts as a fluxing agent.
- c) When PbO is gradually replaced with Fe₂O₃, pour temperature of the melt increases.

Fig II is a graphical representation of above studies in the form of a triangular diagram for PbO-SiO₂-Fe₂O₃ system with other components viz, waste oxide and B₂O₃ being kept at constant levels. Shaded portion of the diagram represents the region of homogeneous vitreous mass formation in the pour temp range of 950-1000 deg C. In all the compositions studied, the basic network formers are SiO₂ & B₂O₃. Other components can be considered to act as net work modifiers. In other words, tendency to devitrification is contained essentially by SiO₂ & B₂O₃. From the region of homogeneous vitreous mass formation, two compositions namely WTR-62 & WTR-63 were identified for detailed investigations. The compositions of these two formulations are given in Table III. Detailed product evaluation was carried w.r.t homogeneity, melting behaviour, chemical durability, sulphate retention, thermal properties etc. Following are the salient observations.

PRODUCT EVALUATION :

A) MELTING BEHAVIOUR & HOMOGENEITY

Table IV lists the melting behaviour and homogeneity of the selected compositions viz, WTR-62 & WTR-63. It can be seen that

- I. In the case of both the compositions, pour temps are in the acceptable range of 950-1000 deg C.
- II. Visual observation of these two glasses did not reveal any phase separation or inhomogeneity.

- III. Optical microscopic examination at 100X has confirmed the vitreous nature of these two glasses as given in Table V.
- IV. X-ray diffraction pattern of these two glasses does not show any detectable crystallinity. Both the glasses were heat treated at 450 deg C for seven days to see if there was any onset of crystallisation owing to prolonged heating. WTR-63 showed the evidence of crystal formation, while WTR-62 was found to remain unaffected as can be seen from Fig III.
- V. SEM investigations carried on these two glasses practically do not show the presence of any crystallinity as shown in Fig IV.

B) CHEMICAL DURABILITY

- I. Leach behaviour of the glass grains of these two glasses based on weight loss basis using static leach test method over a period of 70 days is presented in Fig V. It is seen that WTR-62 is a superior composition compared to WTR-63.
- II. Leachate analysis by ICP method for both the glasses showed that uranium release was below detection levels after seven days of leaching.
- III. Both the glasses have average density of 3.6 gm/cc.
- IV. As homogeneity and leach behaviour tests have clearly shown the superiority of WTR-62 composition, long term leaching studies were carried using the pellet form of WTR-62 glass. In presence of boiling distilled water as the leachant, studies have been carried for the period of over 900 days. The results are given in Figs VI, VII & VIII. It is seen that the leach rate values are quite low even after the completion of leaching for 925 days

C) SULPHATE VOLATILIZATION

Both the glasses were analyzed for the sulphate retention during the entire cycle of vitrification. Glass dissolution was done using HF & HClO₄ treatment. Sulphate present in the solution was precipitated by adding BaCl₂ in presence of glycerol and alcohol and to ensure uniform distribution of turbidity caused by the formation of BaSO₄. The sulphate content was estimated by spectrophotometric method by determining the absorbance of the turbidity at 260nm. As can be seen from Table IV, sulphate retention was found to be 85% for WTR-62 as against 65% for WTR-63.

D) THERMAL STABILITY

- I. Glass transition temps. determined by DTA method are in the range of 505-510 deg C which is acceptable from long term storage consideration of the vitrified high level wastes. (Table IV)
- II. Thermal conductivity, specific heat & thermal diffusivity of WTR-62 was evaluated by using Laser flash method. The experiments were carried from room temperature up

to 400 deg C at 100 deg C intervals. It is seen from Fig IX that thermal diffusivity falls off with rise in temp., which is keeping with the general trend. Both the specific heat & thermal conductivity values at various temps reveal that the WTR-62 has the acceptable thermal properties as shown in Fig X & XI

E) SENSITIVITY TESTS

For plant scale operation, it is necessary that chosen glass should withstand minor variations in waste loading. Keeping this in mind, melting behaviour of WTR-62 was critically studied by varying the waste oxide content. The results of these studies are presented in Table VII. It can be seen from these studies that increase in waste content by a factor of 10% of the total loading is well accommodated in WTR-62 without undue adverse effect.

In the light of the above, it can be concluded that WTR-62 is the candidate composition for high level waste having high uranium, sodium and sulphate content. IR-110, a Sodium borosilicate glass composition developed for sulphate free high level waste from Tarapur has a leach rate of 1.8×10^{-5} gm/cm²/day at 70 deg C on cumulative sodium loss basis after 900 days of leaching. And this composition is currently being adopted for vitrification on plant scale. On the other hand, WTR-62 glass developed for sulphate bearing high level waste of Trombay has a leach rate value of 2.5×10^{-5} gm/cm²/d in boiling distilled water at the end of 925 days of leaching on cumulative sodium release basis. This means that the leach rate of WTR-62 at 100 deg C is comparable to that of IR-110 at 70 deg C. It has been found experimentally that leach rate increases by a factor of 1.5 to 2 for every 20 deg. C rise in the temperature of leaching (5). Thus WTR-62 has better chemical durability and hence is considered acceptable.

CONCLUSIONS

Based on studies with respect to the above important product characteristics, it can be concluded that WTR-62 can be taken as a promising composition for the incorporation of Trombay high level waste which is characterized by high U, Na & SO₄ content and can be considered for adoption on plant scale vitrification..

ACKNOWLEDGMENTS

The authors express their sincere thanks to Waste Management Division for their help in carrying out thermal analysis and X-ray diffraction studies. Thanks are also due to Shri A. B. Patwardhan of Fuel Reprocessing Division for the analysis of leachate samples using ICP. Authors are also thankful to Shri A. R. Biswas of Atomic Fuels Division for carrying out SEM studies. We also thank Shri A.K.Govindan of Waste

Management Project Division and Smt Smita Manohar for their valuable help and suggestions. Our sincere thanks are also to Shri V.P.Kansra Head PE&SDD and Shri K.Balu Director FR&NWM for the encouragement.

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Table I

High Level Waste characteristics

1. Average burn-up	: 1000 MWD/te
2. Cooling time	: 2 years
3. Generation	: 250 L/day
4. Acidity	: 3-3.5 M HNO ₃ (max.)
5. Density	: 1.25 g/cc (approx.)
6. Uranium	: 15-30 g/l
7. Sulfate	: 3 g/l (max)
8. Sodium	: 5-13 g/l
9. Plutonium	: 5-10 mg/l
10. Fission products	: 1.25 g/l (max)
11. Corrosion products	
Fe	: 2.77 g/l (max)
Cr	: 0.56 g/l
Ni	: 0.30 g/l
12. Gross beta/ gamma activity	: 150 Ci/L (max)
13 Noble metals	
Total	: 0.130 g/l (max)
Ru	: 0.100 g/l "
Rh	: 0.020 g/l "
Pd	: 0.010 g/l "

Table II

Detailed Characteristic of Various Vitrified Waste Product in Borosilicate System

Glass code	Composition in weight percent					Pour Temp Deg C	Physical appearance	Leach Data	
	WO	SiO ₂	B ₂ O ₃	Na ₂ O	Others			Days	CFR
GROUP-I									
WTR-12	30	30	15	10	15 (CaO)	950	Cream, liq. phase separation	3	0.061
WTR-13	30	30	15	10	15 (MgO)	1000	separation	3	0.271
WTR-14	30	30	15	10	15(MnO)	950	Dark, spots of syp present	3	0.193
WTR-18	35	30	15	10	10(BaO)	1000	Grey, liquid phase separation	3	0.200
WTR-19	35	30	15	10	10(MnO)	1000	Dark brown, homogeneous	3	0.138
WTR-20	35	30	15	10	10(MgO)	1000	Pale yellow, liquid phase separation	3	0.264
GROUP II-A									
WTR-22	30	35	10	10	10(MnO), 5(TiO ₂)		Dark brown, homogeneous	7	0.048
WTR-24	30	35	10	10	10(BaO), 5(TiO ₂)	950	Pale yellow, liquid phase separation	3	0.070
WTR-25	30	35	10	10	10(CaO), 5(TiO ₂)	1000	Grey, massive phase separation	7	0.011
WTR-27	27	35.4	6.6	14.6	9.7(MnO), 6.5(TiO ₂)	950	Dark, homogeneous	3	0.220
WTR-28	25	35	10	15	10(CaO), 5(TiO ₂)	950	Cream, liquid phase separation	3	0.075
WTR-29	30	35	10	10	5(MnO), 2.5(CaO), 5(TiO ₂), 2.5(Al ₂ O ₃)	1000	Spots of syp present	7	0.038
WTR-30	30	35	10	10	5(MnO), 5(CaO)	950	Light brown, homogeneous	7	0.049
WTR-34	30	35	10	10	5(CaO), 5(TiO ₂), 5(Al ₂ O ₃)	1050	Grey, spots of syp present	7	0.030
WTR-36	30	35	10	10	5(MnO), 5(Al ₂ O ₃), 5(TiO ₂)	1100	Dark, homogeneous	7	0.029
WTR-37	30	35	10	10	5(BaO), 5(Al ₂ O ₃), 5(TiO ₂)	1100	Cream, liquid phase separation	7	0.015
WTR-38	30	35	10	10	5MgO), 5(Al ₂ O ₃)	1050	Cream, liquid phase separation	7	0.025
WTR-39	25	35	10	10	10(MnO), 5(Al ₂ O ₃), 5(TiO ₂)	1000	Dark brown, homogeneous	7	0.025
WTR-42	25	40	10	10	5(MgO), 5(TiO ₂), 5(Al ₂ O ₃)	1000	Grey, liquid phase separation	7	0.025

WO - Waste oxide

SYP- Soluble yellow phase

CFR- Cumulative fraction released on wt. loss basis

NA - Not available

Table II (contd.)

Detailed Characteristic of Various Vitrified Waste Product in Borosilicate System

Glass code	Composition in weight percent					Pour Temp Deg C	Physical appearance	Leach Data	
	WO	SiO ₂	B ₂ O ₃	Na ₂ O	others			Days	CFR
WTR-43	25	40	10	10	5(MnO), 5(TiO ₂), 5(Al ₂ O ₃)	1000	Dark, homogeneous	7	0.019
WTR-44	25	40	10	10	5(CaO), 5(TiO ₂), 5(Al ₂ O ₃)	1000	Light brown, liquid phase separation	7	0.016
WTR-45	30	35	10	10	5(BaO), 5(TiO ₂), 5(Al ₂ O ₃)	950	Grey, syp & liquid phase separation	3	0.455
WTR-46	25	40	10	10	5(BaO), 5(TiO ₂), 5(Al ₂ O ₃)	1000	Grey, spots of syp present	7	0.017
WTR-50	25	40	10	10	5(ZnO), 5(TiO ₂), 5(Al ₂ O ₃)	1000	Grey, homogeneous	7	0.188
WTR-51	25	40	10	10	5(CaO), 5(TiO ₂), 5(MgO)	1050	Grey, liquid phase separation	3	0.098
WTR-52	25	40	10	10	10(CaO), 5(TiO ₂)	1050	Grey, massive phase separation	na	na
WTR-53	25	40	10	10	10(MgO), 10(TiO ₂)	1000	Grey, massive phase separation	na	na
WTR-54	25	40	10	10	10(BaO), 10(TiO ₂)	1050	Grey, syp & liquid phase separation	na	na
WTR-55	25	40	10	10	5(Fe ₂ O ₃)	900	Dark, syp & liquid phase separation	na	na
WTR-56	25	40	10	10	0(Fe ₂ O ₃) 5(TiO ₂)	950	Dark, liquid phase separation	na	na
WTR-57	25	40	10	10	5(Fe ₂ O ₃) 10(TiO ₂)	900	Yellowish green, thick layer syp	na	na
GROUP IIB									
WTR-23	30	35	10	10	10(PbO), 5(TiO ₂)	950	Yellow, homogeneous	3	0.053
WTR-26	25	35	10	15	10(PbO), 5(TiO ₂)	950	Pale yellow, homogeneous	3	0.439
WTR-31	30	35	10	10	2.5(MnO), 2.5(PbO) 2.5(CaO), 2.5(MgO)	950	Light brown, liquid phase separation	7	0.075
WTR-32	30	35	10	10	5(MnO), 5(TiO ₂), 5(PbO)	950	Brown, homogeneous	7	0.063
WTR-33	30	35	10	10	5(CaO), 5(TiO ₂) 2.5(PbO), 2.5(Al ₂ O ₃)	950	Cream, liquid phase separation	na	na
WTR-35	30	35	10	10	5(PbO), 5(Al ₂ O ₃), 5(TiO ₂)	1050	Dark, thick spots of syp present	7	0.027

WO - Waste oxide

SYP- Soluble yellow phase

CFR- Cumulative fraction released on wt. loss basis

NA - Not available

Table II (contd.)

Detailed Characteristic of Various Vitrified Waste Product in Borosilicate System

Glass code	Composition in weight percent					Pour Temp Deg C	Physical appearance	Leach Data	
	WO	SiO ₂	B ₂ O ₃	Na ₂ O	others			Days	CFR
WTR-58	25	40	10	10	5(PbO), 5(TiO ₂), 5(Fe ₂ O ₃)	950	Dark, syp&liquid phase separation	na	na
WTR-60	25	40	10	10	5(PbO), 10(Fe ₂ O ₃)	950	Yellowish green, phase separation	na	na
WTR-61	25	40	10	10	10(PbO), 5(Fe ₂ O ₃)	950	Dark green, layer of liquid phase	na	na
GROUP III									
WTR-62	25	30	10	nil	25(PbO), 10(Fe ₂ O ₃)	950	Dark brown, homogeneous	7	0.004
WTR-63	25	30	10	nil	20(PbO), 15(Fe ₂ O ₃)	1000	Dark brown, homogeneous	7	0.005
WTR-64	25	30	10	nil	15(PbO), 20(Fe ₂ O ₃)	1050	Dark, homogeneous	7	0.008
WTR-67	25	30	10	nil	30(PbO), 5(Fe ₂ O ₃)	900	Pale yellow, not homogeneous	na	na
WTR-68	25	25	10	nil	25(PbO), 15(Fe ₂ O ₃)	950	Pale yellow, thick layer of syp	na	na
WTR-69	25	25	10	nil	20(PbO), 20(Fe ₂ O ₃)	950	Dark, phase separation	na	na
WTR-72	25	35	10	nil	25(PbO), 5(Fe ₂ O ₃)	1050	Pale yellow, spots of syp	7	0.006
WTR-73	25	35	10	nil	20(PbO), 10(Fe ₂ O ₃)	1050	Yellow, spots of syp	7	0.008
WTR-74	25	35	10	nil	15(PbO), 15(Fe ₂ O ₃)	1100	Dark, not homogeneous	na	na
WTR-75	25	40	10	nil	20(PbO), 5(Fe ₂ O ₃)	1100DP	Dark, not homogeneous		
WTR-76	25	40	10	nil	10(PbO), 15(Fe ₂ O ₃)	NP	Semifused mass	na	na
WTR-77	25	25	10	nil	15(PbO), 25(Fe ₂ O ₃)	1000	Dark brown, not homogeneous	na	na
WTR-78	25	25	10	nil	30(PbO), 10(Fe ₂ O ₃)	900	Dark, hygroscopic	na	na
WTR-79	25	25	10	nil	35(PbO), 5(Fe ₂ O ₃)	900	Dark, hygroscopic	na	na

WO - Waste oxide

SYP- Soluble yellow phase

CFR- Cumulative fraction released on wt. loss basis

NA - Not available

Table III

**Selected melt compositions
(Lead Borosilicate System)**

S.No.	Constituents	Composition (wt.%)	
		WTR-62	WTR-63
1.	Waste oxide	25	25
2.	SiO ₂	30	30
3.	B ₂ O ₃	10	10
4.	PbO	25	20
5.	Fe ₂ O ₃	10	15

Table IV**Product properties of selected formulations
(Lead Borosilicate System)**

Property	Glass code	
	WTR-62	WTR-63
1. Pour Temp(degC)	950	1000
2. Homogeneity	Good, Practically no phase separation.	Good, practically no phase separation.
3. L.R.on wt.loss basis, 70 days (g/cm ² /d)	6.8×10^{-6}	1.01×10^{-5}
4. Density (g/cc)	3.58	3.62
5. CFL on wt.loss basis, 70days (g/cm ² /d)	0.0100	0.0147
6. Glass Transition Temp. by DTA (deg C)	505	510
7. Percent Sulphate retention	85	65
8. Uranium release in the leachate (7days)	Not detected	Not detected

L.R. = Leach Rate

CFL = Cumulative fraction leached

Table V**Product evaluation studies on selected formulations
(Lead Borosilicate System)**

Characterization	Glass code	
	WTR-62	WTR-63
1. Optical microscopic examination	Vitreous mass, few inclusions & voids	Vitreous mass, few inclusions & voids
2. X-ray diffraction studies	No Crystallinity observed	No crystallinity observed
3. SEM examination	-do-	-do-
4. X-ray diffraction studies after heat treatment at 450 deg C 7 days	No crystallinity observed	Few crystals
5. Thermal conductivity by Laser flash method (W/M/K) at 27 deg C	0.41	---
400 deg C	1.01	---

Table VI

Variation in waste loading Vs. melt behaviour for WTR-62

S.No	Waste oxide incorporated (wt%)	Pour temp (deg C)	Soaking time (hours)	Physical appearance
1	25	950	2	homogeneous, no phase separation
2	27.5	1000	4	molten mass, slightly viscous, practically no phase separation
3	30	1050	4	in homogeneous, phase separation
4	32.5	1050	4	mass not completely fused, inhomogeneous

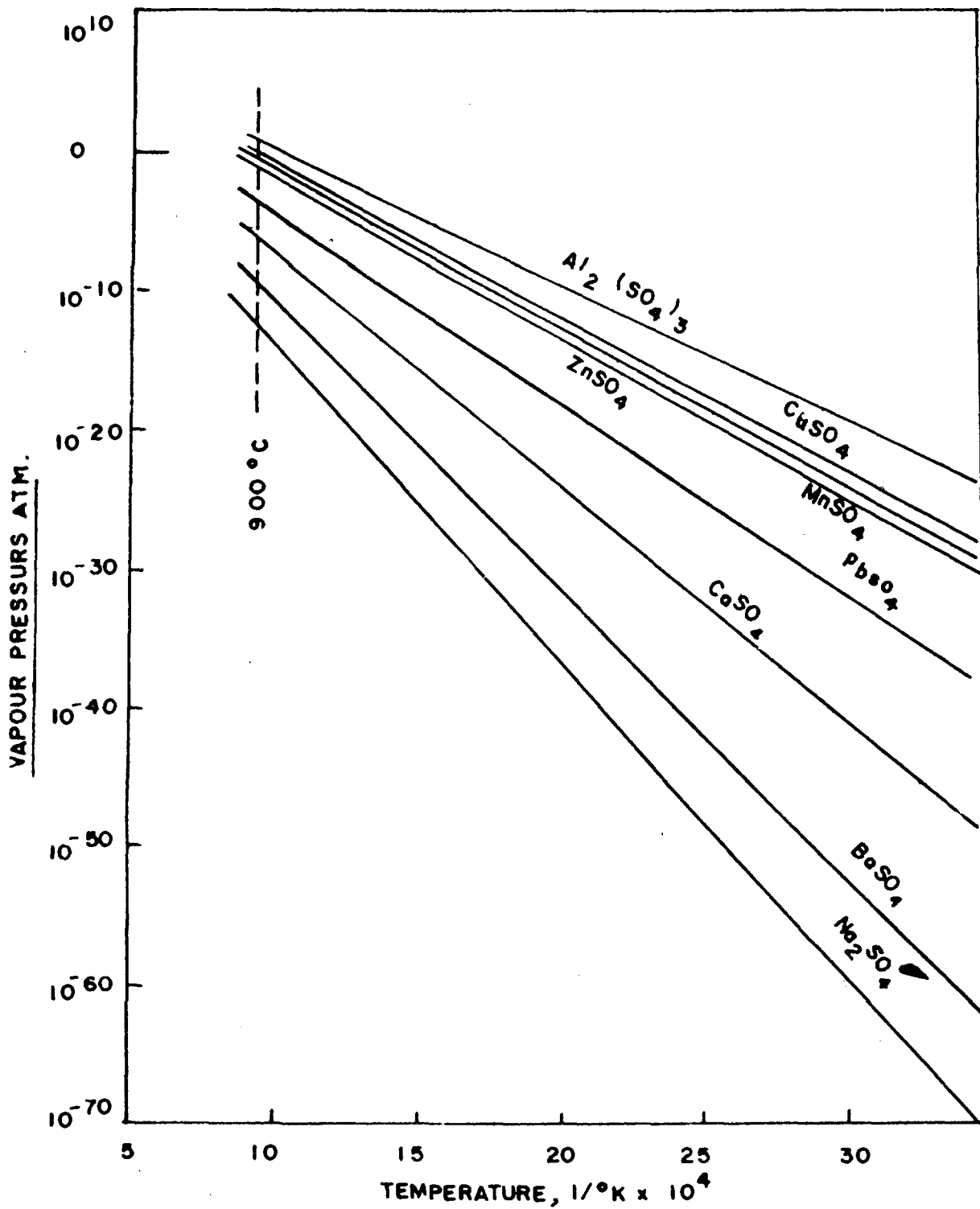


FIGURE: I. DECOMPOSITION PRESSURE OF SULFATES (Ref 1)

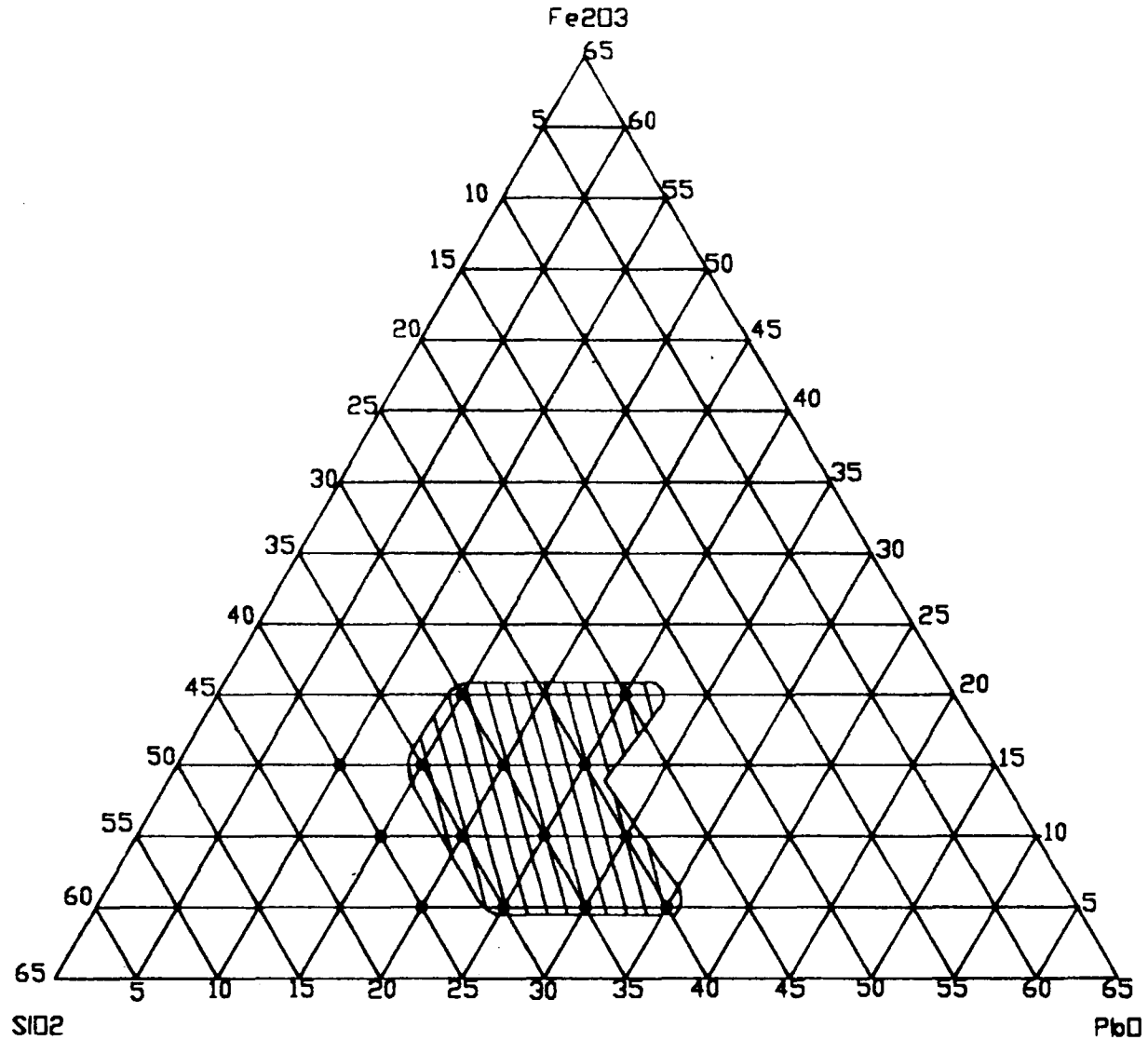
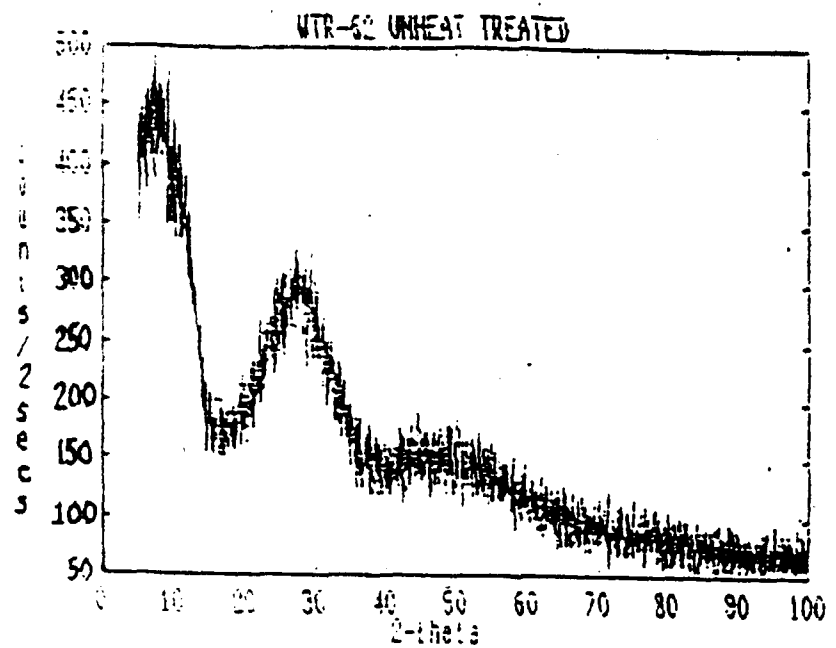
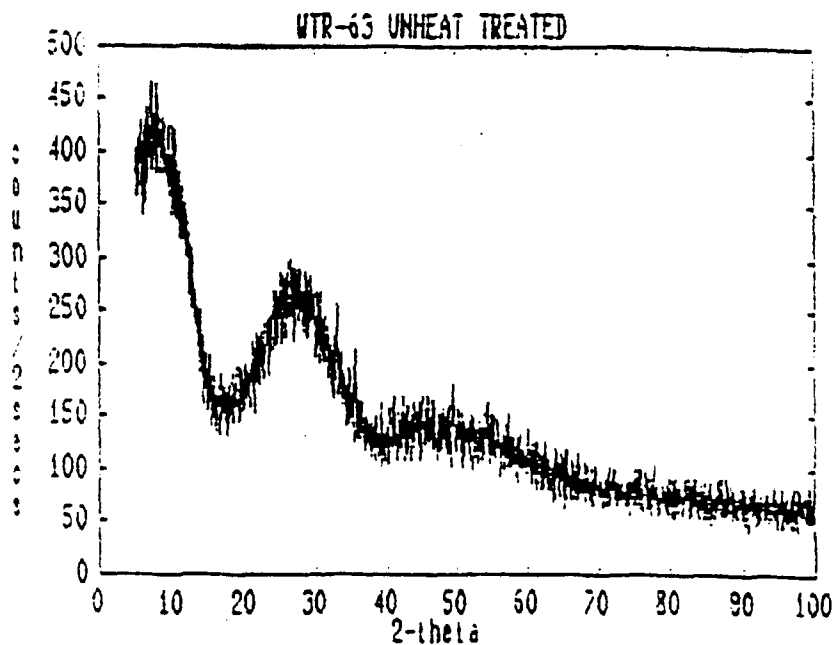


FIG II : REGION OF HOMOGENEOUS VITREOUS MASS FORMATION IN
 THE RANGE OF 900-1000 DEG C IN PbO-B₂O₃-SiO₂-WO-Fe₂O₃
 SYSTEM WITH CONSTANT LEVELS OF 25% WO & 10% B₂O₃



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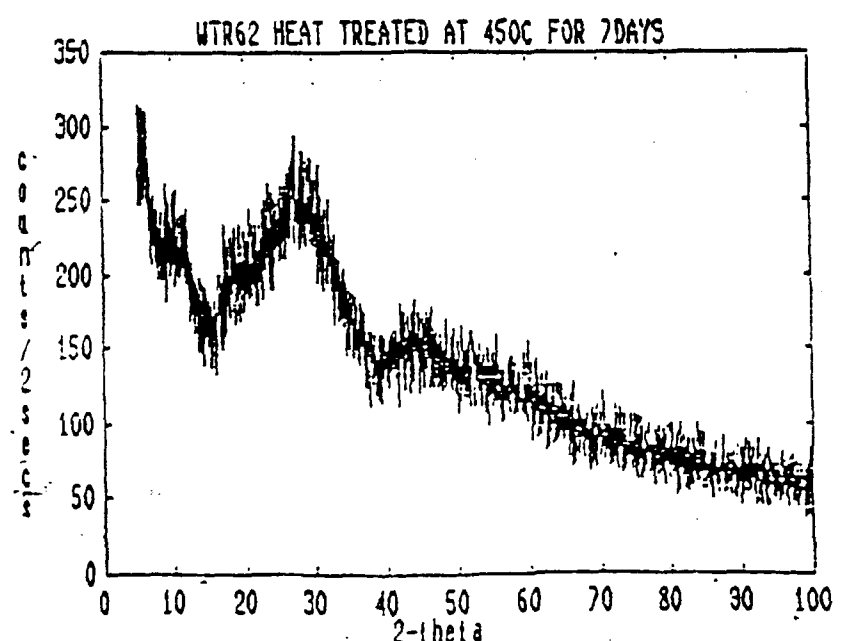
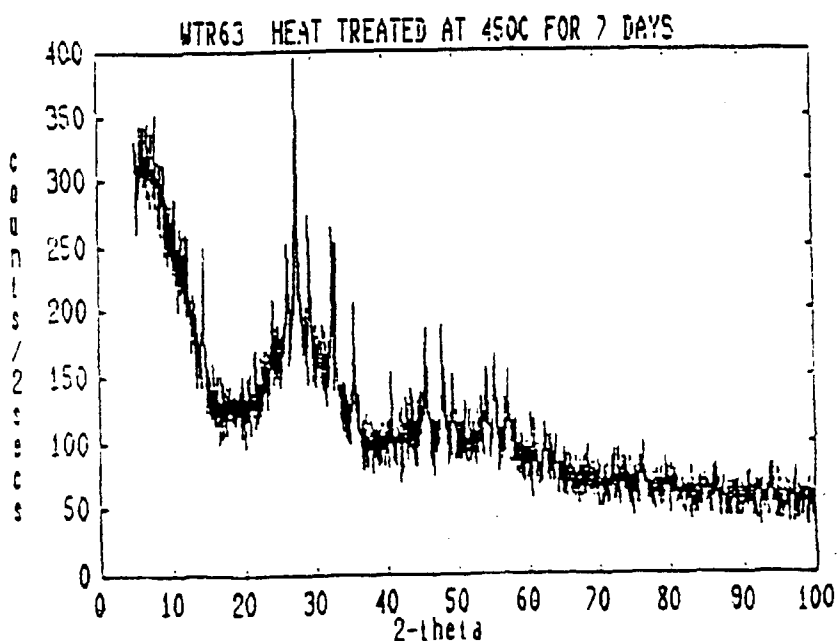


FIG III - DTA PATTERN OF SELECTED VITRIFIED WASTE PRODUCT

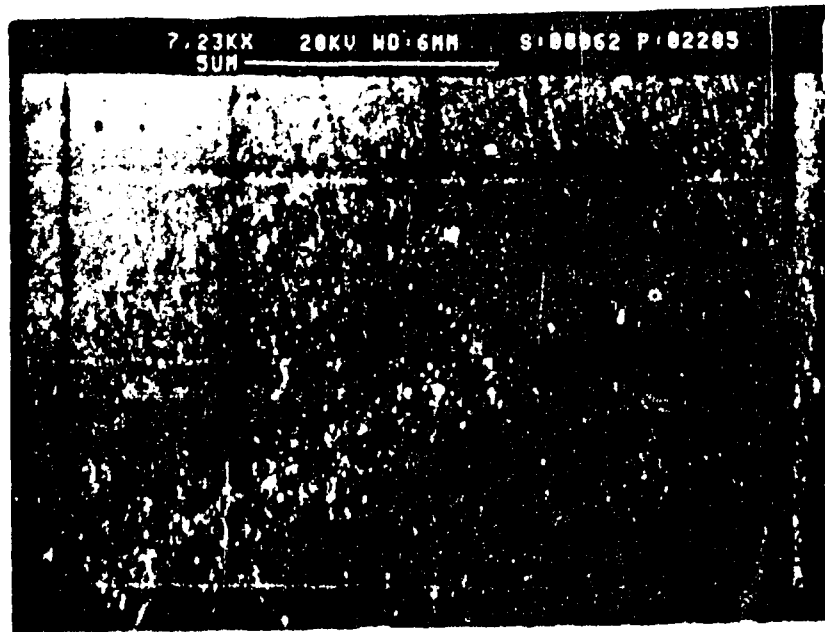


FIG. IV A : SEM PHOTOGRAPH OF VITRIFIED
WASTE PRODUCT WTR-62

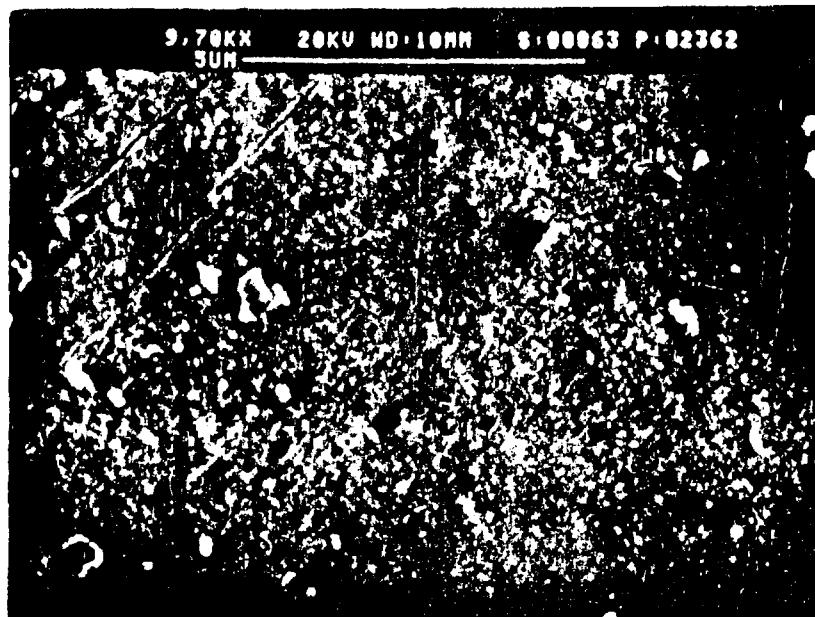
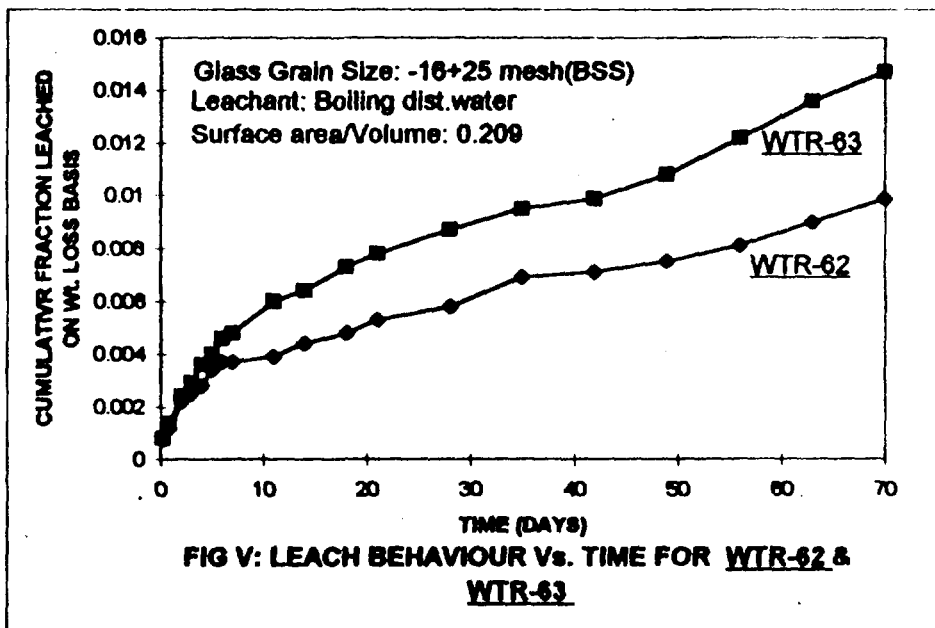
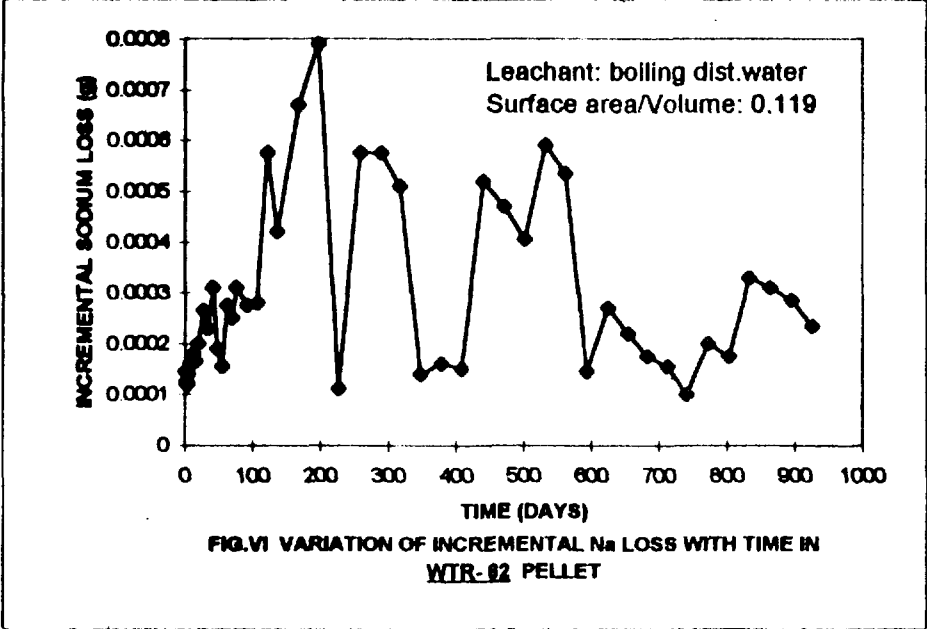
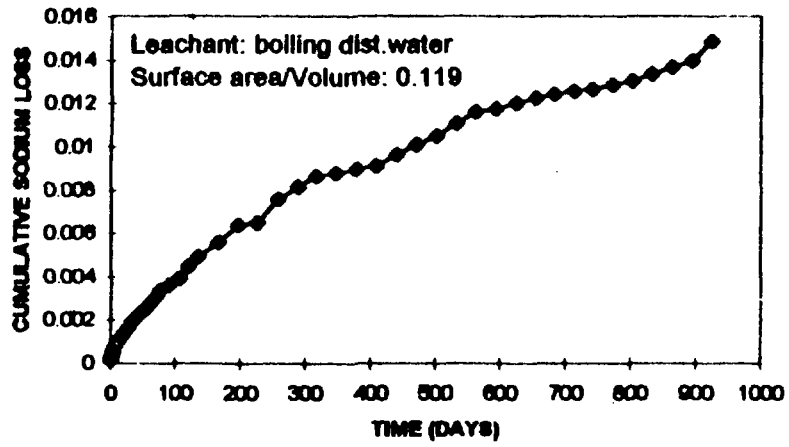


FIG. IV B : SEM PHOTOGRAPH OF VITRIFIED
WASTE PRODUCT WTR-63







**FIG VII : CUMULATIVE Na LOSS Vs. TIME IN
WTR-92 PELLETT**

