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MEMORANDUM

January 19, 1981

TO: L. M. PAPOUCHADO

FROM: B. M. ROBNETT/G. G. WICKS

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DURABILITY OF SRP WASTE GLASS - EFFECT OF DEVITRIFICATION

INTRODUCTION AND SUMMARY

An important program at SRP involves assessment of the durability of SRP waste glass as a function of fabrication, transportation and storage conditions. One phase of this study deals with devitrification of waste glass and the potential effect of crystalline phases on leachability.[1] Devitrification, or crystallization of the glass, can occur if the glass is exposed to high temperatures for long periods of time. This may occur during cooling of the glass canister, during annealing treatments to reduce cracking, or during potential accidents such as high temperature fires.

In this study, crystallization of three simulated SRP waste glass compositions was studied as a function of time and temperature. This includes waste glasses high in iron content (W-Fe), those high in aluminum content (W-Al) and an average (TDS-3A) composition. In order to permite the distribution of Cs and Sr to be measured, the glasses were doped with 1 wt % of each element or about 100 times the amounts of Cs and Sr expected in SRP waste glasses. The crystalline phases formed for each system were identified, the degree of crystallinity determined, and the distribution of Cs and Sr on glass surfaces defined for selected samples. This information was then correlated with leachability data and pH changes in the leachate.

Based on this work, we conclude that:

- Extensive devitrification is unlikely to occur during normal cooling of waste-glass canisters after manufacture or during storage.
- Devitrification could occur during an accidental fire or an annealing treatment.

Major experimental results were:

- Devitrification was observed for all three compositions for heat treatments between 550 to 900°C. Devitrification generally increased the leachability by no more than 10X.
- The largest increase in leachability due to devitrification was about a factor of 50. This occurred for the high iron waste glass heat treated at 600°C and 650°C for one day. This corresponded to the presence of a cesium-rich phase, whose formation was probably enhanced by the high cesium content of the glass.
- The composition with the best leachability, W-Al, also demonstrated the smallest increase in leachability (<5X) due to heat treatments.
- In W-Al and TDS-3A glasses Cs and Sr are generally uniformly distributed throughout the devitrified material, although more concentrated in the glassy phase than in crystalline regions. With the exception of the cesium-rich phase, the same distribution is present in the W-Fe glass.

EXPERIMENTAL PROCEDURE

Simulated waste glasses made with Frit 131 and W-Al, TDS-3A, and W-Fe waste compositions were doped with 1 wt % each of nonradioactive cesium and strontium (Table 1). This doping represents about 100 times the actual amounts of Cs and Sr expected in SRP waste-glasses. Because of the increased content of these constituents, the distribution of each could be assessed. Samples were heat treated at various times and temperatures to simulate potential fabrication, processing and storage conditions (Table 2). Also, samples were program cooled from the melt using a Lindberg furnace equipped with a microprocessor programmer to simulate ICM cooling, continuous melter cooling, and possible annealing treatments.

Crystalline phases were identified by use of optical and scanning electron microscopy, complementary x-ray energy spectroscopy, wide angle x-ray diffraction and microprobe analysis. In addition, the distribution of the simulated radionuclides within the devitrified material was determined through microprobe mapping.

The degree of crystallinity was estimated by comparing diffraction pattern peak heights of known crystalline weight percent standards to peak heights of samples containing that phase. This was used in conjunction with the grid technique on selected micrographs.

For leachability tests, samples were ground to 40-60 mesh size and heated in a 90°C oil bath in a buffered solution of either pH 3, 7 or 11 (Table 3) for three days. Surface area to leachant volume was about 7 to 1. The pH of each leachate was measured approximately 16 hours after the samples were removed from the constant temperature bath. Silica content within the leachate was measured using colorimetry, and sodium, cesium and strontium content was measured by atomic absorption.

RESULTS

A. Leachability of Program Cooled SRP Waste Glass

Effects of the two leading vitrification processes, continuous ceramic melting (CM) and in-can melting (ICM) on formation of crystalline phases in the product, were assessed by programmed cooling of SRP waste glasses from 1100°C. In addition, the effect of slow cooling the molten glass through the annealing range was also investigated.

- Continuous Melter Cooling

A 21-hour cooling schedule shown in Table 4 was used to simulate cooling of a 2 ft. diameter CM canister. This was the cooling profile measured at Battelle Pacific Northwest Laboratory for selected continuous ceramic melter experiments.[2]

After cooling, the solidified laboratory scale product was primarily glassy which corresponded to a leachability very similar to the standard. Therefore, the thermal history of glass cooled from a joule-heated ceramic melter would not be expected to significantly effect the leachability of the solidified product.

- In-Can Melter Cooling

A 14 hour cooling schedule was used between 700°C and 500°C to simulate the cooling rates observed at TNX for full-scale ICM glasses over this temperature interval (Table 4). This temperature range represents a region of maximum formation of the important acmite phase. No significant changes in leachability due to this cooling procedure were observed.

Samples from a large scale ICM test at PNL were then compared with laboratory-scale furnace-treated glass at SRL. Both were TDS compositions, but the PNL glass contained Frit 211 while the SRL glass contained Frit 131. The effects of different frit compositions, cooling schedule and waste form sizes on leachability of the ICM glasses were observed to be small. Changes in leachability measured at three different pH values differed by about a factor of two (Figure 1).

- Annealing Procedure During Solidification

  Thermal cracking of full scale SRP waste glasses can be minimized by slowly cooling the product through the annealing range during fabrication. However, for this type of operation, crystalline phases can be formed within the glass which may effect the durability of the product.

  An extreme cooling schedule includes holding the glass between 500 and 600°C for 83 hours (Table 4). This results in a large degree of crystallinity. The greatest effect on subsequent leachability was seen in W-Fe glass leached in neutral solution where the leachability based on silica increased by about a factor of 50.

B. Leachability of Heat Treated SRP Waste Glass

The effect of crystallinity on leachability was examined for SRP 131/W-Al, 131/TDS-3A, and 131/W-Fe simulated waste glasses. Glass samples were held at various temperatures for 1 day and 1 week. These heat treatments were used to simulate temperature increases the final glass product might experience, such as during annealing treatments, fires, or loss of convection cooling in the storage facility. The leachability of these heat treated samples increased by no more than a factor of 10 in most cases. The crystalline products formed in each of the three compositions are summarized in Figure 2. The subsequent leachabilities of these compositions were determined for each heat treated sample, based on extraction of Si, Cs, Sr, and Na and leached in three different buffered solutions. These data are summarized in Tables 5A, 5B, and 5C.

- Time, Temperature Dependence

  - W-Al Glass

    The highest leachability occurred after being held at 800°C for one day. Leachabilities were also relatively high at 650°C. Spinel ($NiFe_2O_4$) was the only crystalline phase formed in W-Al glass after one day, with glasses heated at 650°C having the highest degree of crystallinity. After one week at 600°C, acmite ($NaFeSi_2O_6$) and nepheline ($NaAlSiO_4$) were formed. Leachability based on sodium for this treatment was greater than the standard by a factor of 15 and greater than the 550°C treatment by a factor of 10 in basic leachant. The 550°C treatment contained only spinel.

  - TDS-3A Glass

    Highest leachabilities for TDS-3A glass occurred for heat treatments of 600-650°C and 800°C. In the 600-650°C range, the amount of acmite was at its maximum and at 800°C spinel was introduced along with the acmite resulting in the maximum total crystallinity. Samples heated at 600°C and leached in basic solution had a leachability based on silica ∿30X greater than the standard. Leachability based on sodium was highest at 650°C which corresponds with maximum degree of acmite. At 800°C, leachability based on sodium was on the decline as well as the degree of acmite. At 900°C, no acmite was formed and the leachability based on sodium was only 50% greater than the standard. This may suggest that in the TDS-3A waste glass composition, the presence of acmite accelerates sodium leachability. After one week at 550°C and 600°C, lithium metasilicate (Li_2SiO_3) was formed. The degree of acmite was higher at 550°C after one week than after one day. With this increase in acmite, leachability based on sodium increased by about a factor of 10. At 600°C, where the change in crystallinity was only the introduction of Li_2SiO_3, leachability based on sodium did not change, but leachability based on silica increased for neutral and acidic leachants.

- W-Fe Glass

Leachabilities of W-Fe glass were highest for samples heated between 600 and 650°C which corresponds to the range of maximum crystallinity (600-700°C). These 600°C and 650°C heat treatments resulted in a leachability based on silica of ∿50X greater than the standard in neutral solutions. The crystalline phase in these samples was acmite. Leachability based on sodium follows the same general trend as in the TDS-3A glass. As the degree of acmite increases, so does leachability based on sodium. At 900°C, spinel replaced acmite as the only crystalline phase, and the leachability based on sodium was drastically reduced. Leachability based on cesium was much higher in W-Fe glass than in the other types. It generally had a maximum around 600°C. Glass heated for one week at 600°C was found to contain a cesium-rich phase not found in W-Al or TDS-3A glasses. This sample's leachability based on cesium was greater than the standard by about a factor of 70. X-ray energy spectroscopy showed that there was no Cl^- in the phase eliminating the possibility that the phase was residual CsCl used to dope the glass. Samples heated for one day also showed these high leachabilities. The formation of this phase was emphasized since these glasses were highly doped. After one week, the degree of acmite formed at 550°C was greatly increased as well as the leachability based on sodium.

- pH Dependence

Basic and acid solutions were generally more corrosive to amorphous and devitrified glasses than neutral leachants. In general, the larger the degree of crystallinity, the more the glass leached at each of the pH values examined. Leachability based on strontium was extremely low in basic solutions for all glass compositions. A possible explanation for this is that the strontium may be absorbed into a gel layer that forms on the surface of glass leached in basic solutions and hence would not be measured in solution.

- Fire Scenario

A standard Department of Transportation (DOT) fire would result in a temperature rise to 790°C. At this temperature, devitrification would occur in less than one day. For W-Al glass, spinel would form; for TDS-3A glass, spinel and acmite; and for W-Fe glass, acmite alone would be the primary phase produced. The leachability of all simulated compositions generally will increase due to this heat treatment to a maximum of about 45 times the initial leachability. Leachability increases due to a 800°C heat treatment as a function of waste composition and solution pH are summarized in Table 5.

- Loss of Convection Cooling

  Loss of convection cooling during interim storage could result in a temperature rise to about 325°C. The effect of this temperature for tests one month in duration were assessed. These samples showed no significant devitrification or changes in leachability.

- Long Term Storage Effects

  As a result of the radionuclide content of the waste, the storage temperature will be about 100°C. Samples were heated at 150°C (maximum storage temperature considered) for three months and then examined for crystallinity and leachability. Again, there was no devitrification and no significant changes in leachability for each of the three compositions studied. Because of the very low storage temperatures considered, nucleation and growth of crystalline phases would not be expected to occur in the absence of liquid water, even for very long storage times.

C. Cooling from the Melt vs. Heat Treatment of Solidified Forms

Samples heated at 700°C for one day using both methods show that in general significantly less crystallinity will result on cooling from the melt than during heat treatment of solidified forms. This is a result of the nucleation and growth curve encountered during heat treatments. This difference is also reflected in the leachability data.

D. Full Scale SRP Waste Glass

The practical application of data from the present study to production of full-scale SRP waste glass forms can be summarized as follows:

(1) Appreciable amounts of crystallization would not be expected to occur due to cooling rates associated with CM or even ICM glasses. Hence, the leachability of glasses from these two fabrication methods would not be expected to be significantly different.

(2) Loss of convection cooling in an interim storage facility would not result in significant changes in devitrification or leachability characteristics.

(3) A standard DOT fire could cause appreciable amounts of devitrification and a maximum leachability increase of about 45X.

(4) Potential annealing treatments to minimize thermal cracking or heal cracks already produced could result in a significant increase in crystalline content of the product. This could result in a maximum leachability increase of about 50X. Hence, a balance between glass cracking and devitrification will have to be made.

(5) Due to the relatively low storage temperatures being considered, devitrification and changes in leachability due only to thermal effects will not be expected, even for long term storage.

E. Microstructures

• Structural Determination of Phases

Crystalline phases were identified using wide angle x-ray scattering (WAXS). Patterns of devitrified samples were compared to those of crystalline standards supplied by W. C. Mosely. Phases were also examined using the scanning electron microscope (SEM) and quantitative analyses were carried out on the microprobe. Phases formed as a function of temperature are presented in Figure 2.

• Degree of Crystallinity

A relative measure of crystallinity was obtained by comparing peak heights of unknown samples to those of standards. Pure crystals were mixed with the fines (<60 mesh) of nonheat treated glass so that the mixture was 25% crystalline. Percent crystallinity in samples were calculated using the following formula:

$$\frac{\%\ \text{crystallinity in standard}}{\text{peak height standard}} = \frac{\%\ \text{crystallinity in sample}}{\text{corresponding peak height sample}}$$

These numbers correspond with those obtained using the grid technique on microstructures and are summarized in Table 6.

• Distribution of Cs and Sr

The distribution of elements throughout the devitrified material was obtained by microprobe mapping with the help of W. C. Mosely. This aided in phase identification and showed where the cesium and strontium were in the devitrified material (Figure 3). Both cesium and strontium were uniformly distributed in W-Al and TDS-3A glasses. However, a phase was identified in W-Fe glass as being $\sim$50 weight percent Cs. This corresponds to large increases in leachability based on cesium. The chemical analyses for the various phases in each glass are shown in Tables 7A, 7B and 7C.

• Correlation of Phases with Leachability

The presence of acmite appears to increase leachability based on silica in neutral and basic solutions. It also increases leachabilities based on sodium at all pH's.

Spinel slightly increases leachabilities based on silica as shown in W-Al and by the increase in leachability when it is introduced into TDS-3A glass at 800°C.

Li_2SiO_3 and nepheline are not present in sufficient quantities to detect any significant effects.

F. pH Considerations

Earlier studies have suggested that there is a correlation between leachant pH and glass corrosion.[3] Glasses leached below pH 4 or above pH 9 seem to leach at extremely high rates. This general trend was observed in this study (Table 8). When there was an initial pH of 3, the highest leachabilities corresponded to leachates with a final pH of below 3 and those that increased in pH above 9. Neutral leachates hold their initial pH value except for W-Fe glass where the pH increased significantly to above the critical pH of 9. Basic leachates (pH 11) maintain their pH above 9, and the final pH values follow the correlation that the higher the pH, the higher the leachability of the glass. The changes observed in the pH values of the buffered leachants represent the inability of the buffer to maintain a constant pH. The experimental data suggests that monitoring the ground water pH in the repository could be a nondestructive means of predicting waste glass durability as previously suggested. Also, the pH of the ground water could possibility be controlled if engineered barriers were designed to provide buffering potential.

REFERENCES

1. G. G. Wicks and M. D. Dukes, "Durability of SRP Waste Glasses - Program Document," DPST-80-542, September 1980.
2. G. G. Wicks, "Trip Report to PNL - Canister Instrumentation," DPST-78-327, March 1978.
3. G. G. Wicks, "Durability of SRP Waste Glass - Effect of Ground Water pH," DPST-80-675, November 1980.

Table 1

A. SIMULATED WASTE GLASS COMPOSITIONS

Component	W-Al	TDS-3A	W-Fe
Frit 131	69.3	68.2	68.2
CsCl	1.0	1.0	1.0
$SrCl_2$	1.0	1.0	1.0
Fe_2O_3	3.4	14.1	17.6
MnO_2	3.2	4.1	1.2
Aw-500	2.9	3.0	2.9
Al_2O_3	14.1	2.8	0.4
NiO	0.6	1.7	3.0
SiO_2	1.3	1.2	0.9
CaO	0.3	1.1	1.2
Na_2O	1.4	0.9	1.8
Coal	0.7	0.7	0.6
Na_2SO_4	0.2	0.2	0.2

B. FRIT 131 COMPOSITION

SiO_2	57.9
Na_2O	17.7
B_2O_3	14.7
TiO_2	1.0
Li_2O	5.7
MgO	2.0
ZrO_2	0.5
La_2O_3	0.5

Table 2

TEMPERATURE	TIME				COMMENTS
°C	1 Day	1 Wk.	1 Mo.	3 Mos.	
R.T.	X	X	X	X	Standard
150°C				X	Storage (Maximum)
325°C			X		Loss of Cooling in Storage
350°C	X	X			Annealing Range
400°C	X	X			Annealing Range
450°C	X	X			Annealing Range
500°C	X	X			Annealing Range
550°C	X	X			Annealing Range
600°C	X	X			Annealing Range
700°C	X				
800°C	X				DOT Fire
900°C	X				

Table 3

BUFFERED SOLUTIONS

Desired pH	Buffered Solution
3	0.5M Glycine, 14 drops HCl
7	50 ml of 0.1M Tris* + 46ml 0.1M HCl
11	LiOH + H_2O

Table 4

TEMPERATURE SEQUENCES FOR PROGRAM COOLED SAMPLES

CM	ICM	Annealing
1100°C	1100°C	1050°C or 1150°C
↓ 1 hr	↓ 1 hr	↓ Quench (∿1 hr)
900°C	700°C	600°C
↓ 1 hr	↓14 hr	↓ 1.2°/hr
800°C	500°C	350°C
↓ 3 hr		↓ 2.4°/hr
700°C		R.T.
↓ 6 hr		
600°C		
↓10 hr		
500°C		

Table 5A

HEAT TREATMENTS

A. *Leachabilities of 131/W-Al*
(Based on extraction of Si, Na, Cs, Sr)

		Temperature (°C)									
		(One Day)							(One Week)		
pH	*	R.T. Std.	550	600	650	700	800	900	500	550	600
3	L_{Si}	112	105	103	119	110	156	97	117	144	93
	L_{Na}	236	30	537	658	277	828	218	266	335	1400
	L_{Cs}	83	173	206	463	118	1965	190	96	250	951
	L_{Sr}	163	179	224	319	199	745	241	122	498	1800
7	L_{Si}	9	14	31	49	25	69	2	9	24	1
	L_{Na}	81	109	192	485	141	376	79	86	132	-
	L_{Cs}	15	37	93	293	46	826	87	17	118	-
	L_{Sr}	25	36	94	243	69	253	39	25	194	-
11	L_{Si}	23	47	58	123	61	169	27	34	48	230
	L_{Na}	110	153	210	411	169	388	210	120	166	1740
	L_{Cs}	19	195	37	90	41	363	37	16	78	558
	L_{Sr}	2	3	2	1	9	14	12	8	14	26

*Leachabilities are shown as actual leachabilities x 10^7 (g/cm^2-day)

Table 5B

B. Leachabilities of 131/TDS-3A
(Based on extraction of Si, Na, Cs, Sr)

Temperature (°C)

pH	*	R.T. Std.	(One Day)						(One Week)		
			550	600	650	700	800	900	500	550	600
3	L_{Si}	129	147	51	64	78	134	100	137	83	146
	L_{Na}	227	495	3899	4459	2716	1510	304	255	4020	3710
	L_{Cs}	107	216	596	849	266	2990	150	104	856	1030
	L_{Sr}	215	240	520	754	1571	1550	341	220	955	1160
7	L_{Si}	18	20	38	61	70	58	11	16	283	621
	L_{Na}	156	338	2295	2977	1884	987	282	140	2640	2720
	L_{Cs}	35	110	293	330	183	719	43	34	256	312
	L_{Sr}	69	117	139	77	310	302	194	63	36	39
11	L_{Si}	92	96	2950	780	463	302	70	89	576	584
	L_{Na}	290	279	3714	4190	3049	1040	239	229	2520	3470
	L_{Cs}	34	53	542	622	230	437	57	36	1470	429
	L_{Sr}	2	3	6		8	13	12	5	13	21

*Leachabilities are shown as actual leachabilities x 10^7 (g/cm^2-day)

Table 5C

C. Leachability of 131/Fe
(Based on extraction of Si, Na, Cs, Sr)

Temperature (°C)

		(One Day)							(One Week)		
pH	*	R.T. Std.	550	600	650	700	800	900	500	550	600
3	L_{Si}	150	88	250	162	122	130	149	184	362	391
	L_{Na}	414	3817	18547	11378	4403	3620	1229	503	16200	22100
	L_{Cs}	153	871	2510	2120	416	2900	12	160	4110	4140
	L_{Sr}	296	728	1060	1131	1260	1370	953	382	2010	2110
7	L_{Si}	45	218	2860	2420	180	190	48	59	757	704
	L_{Na}	211	3520	18781	16477	3377	4710	797	246	16900	17800
	L_{Cs}	75	426	5150	3655	1912	1320	18	71	5190	5470
	L_{Sr}	101	34	103	18	63	67	429	146	713	765
11	L_{Si}	339	652	2660	3640	1435	1800	590	379	810	956
	L_{Na}	1278	9265	20297	15686	8760	11100	2446	1475	15500	17600
	L_{Cs}	121	2080	5920	5612	2927	2160	15	158	5690	5790
	L_{Sr}	3	15	93	17	16	24	21	9	342	540

*Leachabilities shown are actual leachabilities x 10^7 (g/cm^2-day)

Table 6

DEGREE OF CRYSTALLINITY

		Phase (%)							
		(One Day)				(One Week)			
Composition	Temp. (°C)	Acmite	Li_2SiO_3	Spinel	Nepheline	Acmite	Li_2SiO_3	Spinel	Nepheline
W-Al	550	0	0	0	0	0	0	6	0
	600	0	0	2	0	6	0	<1	7
	650	0	0	8	0				
	700	0	0	6	0				
	800	0	0	6	0				
	900	0	0	6	0				
TDS-3A	550	19	0	0	0	54	8	0	0
	600	46	0	0	0	44	6	0	0
	650	48	0	0	0				
	700	35	0	0	0				
	800	38	0	14	0				
	900	0	0	27	0				
W-Fe	550	3	0	0	0	39	5	0	0
	600	28	0	0	0	37	6	0	0
	650	37	0	0	0				
	700	31	0	0	0				
	800	17	0	0	0				
	900	0	0	27	0				

Table 7A

MICROPROBE STUDY

A. 131/W-Al 600°C/One Week

Phases Present:

Multitone Gray Matrix--------------------------Nepheline and Residual Glass

Black Phase-----------------------------------Li_2SiO_3

White Particles--------------------------------Spinel

Light Gray Phase Around Particles---------------Acmite

Chemical Analysis:

	Nepheline + Glass*	Li_2SiO_3**	Spinel	Acmite
Cs	.927	.056	.055	.592
Sr	.702	.059	.172	.625
Al	20.717	2.303	2.118	3.501
Na	17.918	1.156	2.162	12.381
Ti	.445	.665	2.800	3.991
Ni	.055	.329	11.386	1.672
Fe	3.006	.694	22.045	16.693
Mn	2.123	.806	8.070	4.427
Ca	.446	.014	.251	2.483
Si	51.227	68.916	55.806***	49.063***

*The nepheline and glassy phases of the matrix could not be accurately distinguished for separate analyses.

**Li cannot be measured by this analytical technique.

***Si analysis results from adjacent phase.

NOTE: Cs and Sr are concentrated in the acmite and matrix.

Table 7B

MICROPROBE STUDY

B. 131/TDS-3A 600^{o}C/One Week

Phases Present:

Multitone Gray Matrix-------------------------------Acmite, Glass

Black Phase---Li_2SiO_3

White Particles-------------------------------------Mn Rich Phase, Spinel

Chemical Analysis:

	Acmite + Glass	Li_2SiO_3	Mn/Spinel
Cs	.527	.757	.485
Sr	.699	.442	.400
Al	3.291	3.477	3.386
Na	12.880	7.667	9.479
Ti	.870	.542	.432
Ni	1.104	.968	9.753
Fe	21.328	9.796	6.149
Mn	2.628	1.692	36.683
Ca	1.770	.773	.290
Si	54.071	54.572	24.436

NOTE: In general, Cs and Sr have a tendency to concentrate in the matrix which is predominantly acmite. However, important quantities of these elements were also seen in Li_2SiO_3 and the Mn-rich phase.

Table 7C

MICROPROBE STUDY

C. 131/W-Fe 600^{o}C/One Week

Phases Present:

Multitone Gray Matrix---------------------------------------Acmite, Glass

Black Phase---Li_2SiO_3

White Particles---Cs-rich Phase

Chemical Analysis:

	Acmite + Glass	Cs-rich Phase
Cs	.293	42.558
Sr	.438	.206
Al	1.070	12.156
Na	14.548	4.951
Ti	.986	.199
Ni	4.232	.532
Fe	20.298	6.366
Mn	1.060	.394
Ca	1.823	.293
Si	50.787	47.879

NOTE: Particles formed were very rich in Cs. This is the first time that a Cs-rich phase has been found in SRP waste glass for these highly doped glasses. Subsequent analysis found no chlorine present in this phase indicating that it was not the CsCl starting material.

Table 8

FINAL pH AFTER LEACHING

		Temperature (°C)								
		(One Day)						(One Week)		
Initial pH	Composition	550	600	650	700	800	900	500	550	600
3	W-Al	3.27	3.27	3.25	3.13	2.97	3.04	3.12	2.91	7.63
	TDS-3A	3.25	8.26	8.41	6.52	3.27	3.08	3.13	8.53	9.05
	W-Fe	8.55	9.61	9.21	8.35	8.21	3.21	3.20	9.56	9.89
7	W-Al	7.35	7.41	7.66	7.01	7.48	7.11	7.08	7.91	8.93
	TDS-3A	7.60	8.68	9.01	8.31	7.76	7.39	7.08	9.82	10.48
	W-Fe	9.48	11.89	11.52	9.45	9.34	7.85	7.25	10.52	10.80
11	W-Al	10.07	10.03	10.22	9.96	10.49	9.90	9.62	9.61	10.77
	TDS-3A	10.03	11.60	11.74	11.37	10.69	9.88	10.04	11.48	11.65
	W-Fe	11.93	12.25	12.22	11.91	11.97	11.21	11.03	10.84	11.33

Figure 1
IN-CAN MELTING

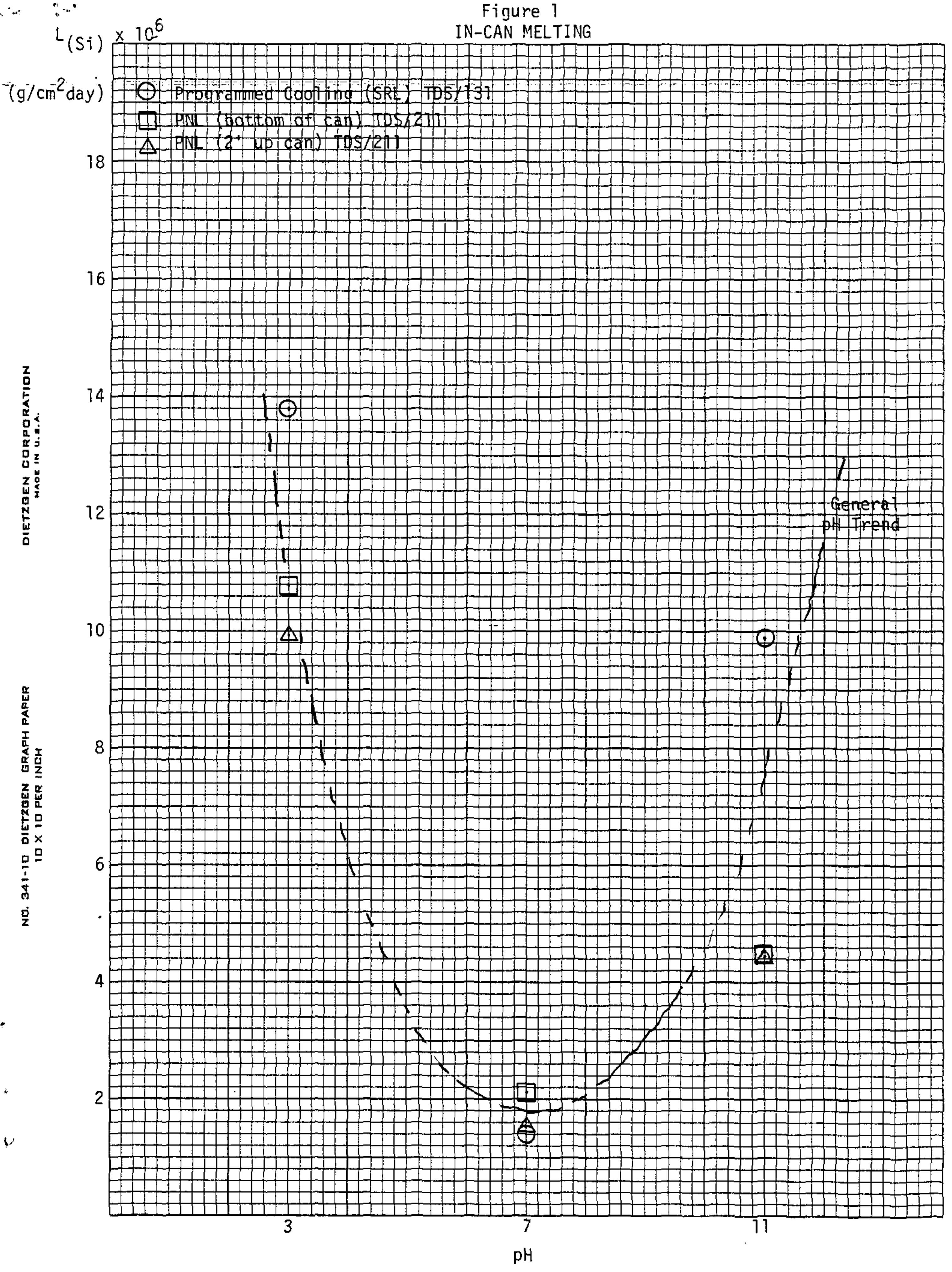


Figure 2

PHASES

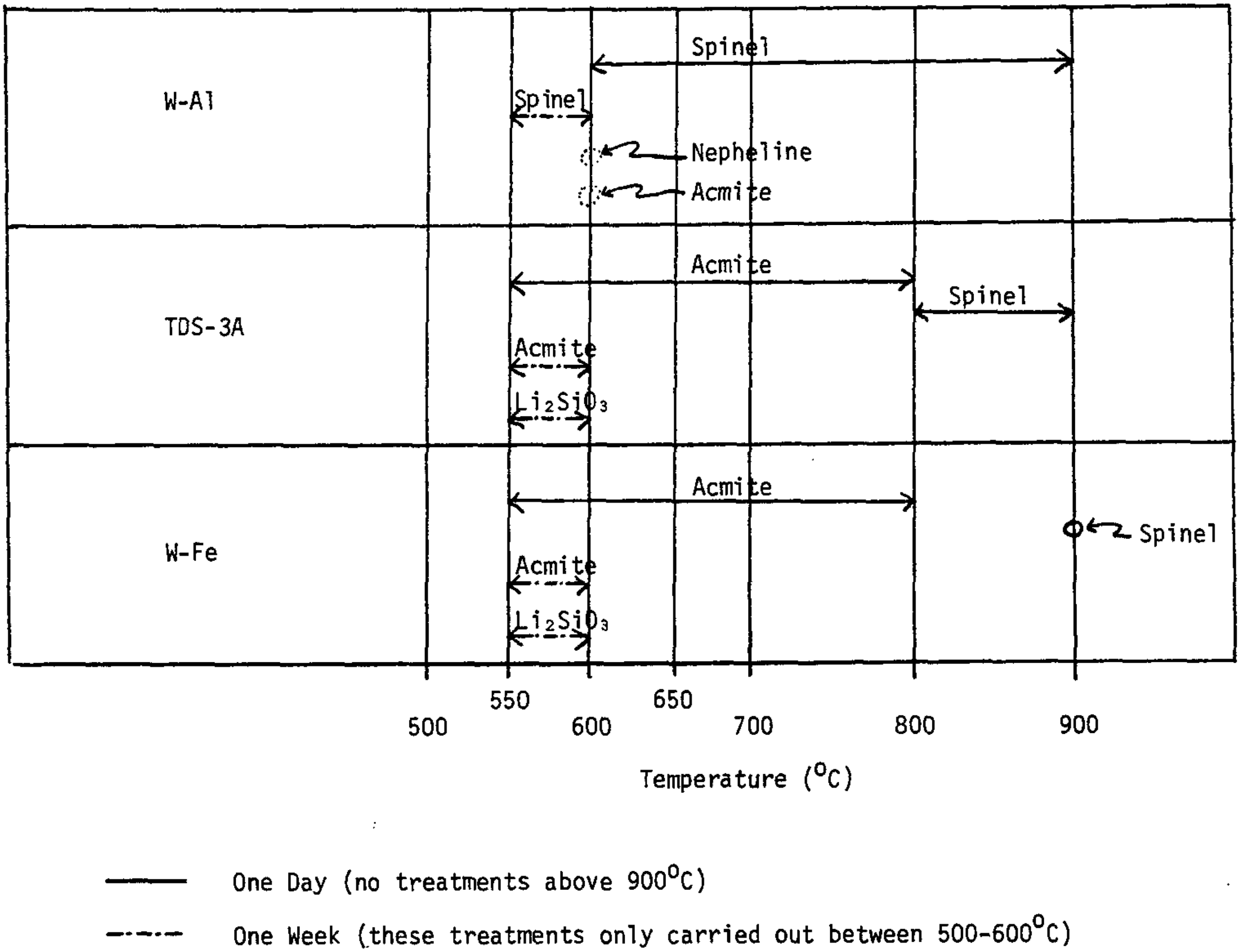
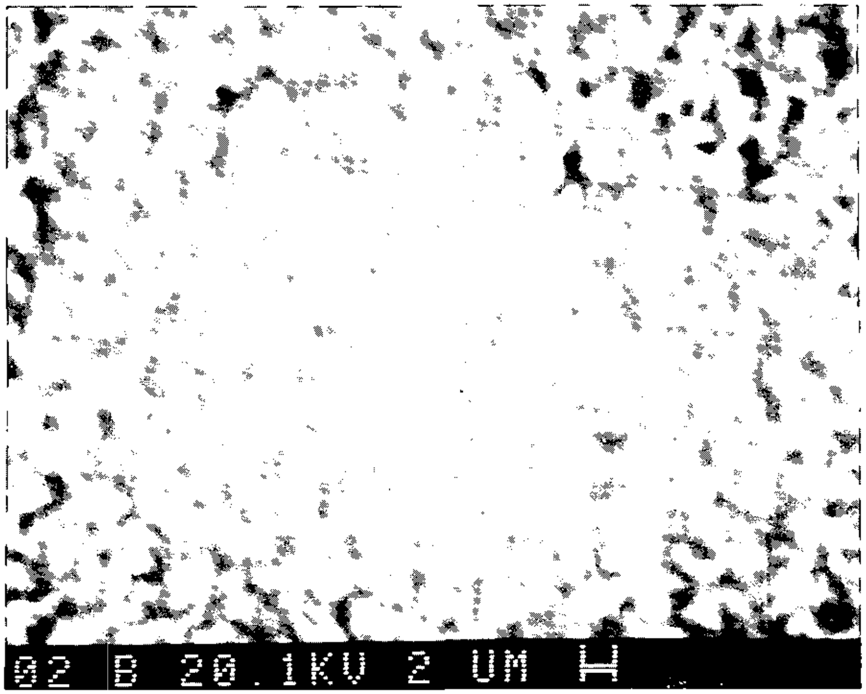


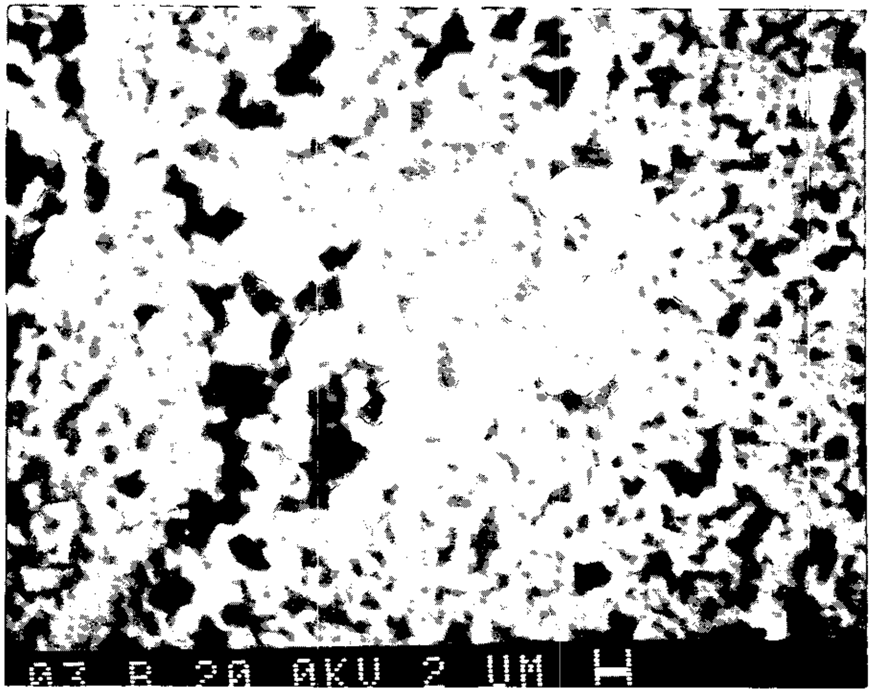
Figure 3

MICROSTRUCTURES USED
FOR MICROPROBE ANALYSES

131/W-Aℓ 1000x



131/TDS-3A 2000x



131/W-Fe 2000x