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DEVELOPMENT OF CERAMIC COATINGS FOR FISSION PRODUCT RETENTION IN CERAMIC FUELS

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ABSTRACT

A general discussion on glaze–type coatings for fission product retention within BeO spheres fuelled with $PuO_2 - ThO_2$ particles is presented. From these considerations, a glaze thickness of 0.004 inch was chosen as a basis for laboratory studies.

Glaze development commenced with a conventional high temperature glaze; this was modified firstly by increasing the SiO_2 content and then by progressively replacing some of the SiO_2 by TiO_2, ZrO_2, $TiO_2 - ZrO_2$, BeO, and BeO plus TiO_2. Glaze structures varied from amorphous to predominantly crystalline. When applied to fuelled BeO cubes, some glazes failed to cover fuel particles and others tended to react with them.

Two of the most promising glazes behaved poorly in fission product release experiments, probably because glaze–fuel interaction had allowed fuel migration to the glaze surface.

It is concluded that glaze type coatings show little promise for the present applications.

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CONTENTS (continued)

1. INTRODUCTION

In the current High Temperature Gas Cooled Reactor Project at Lucas Heights, a pebble–bed type reactor fuelled by all–ceramic beryllium oxide–based spheres is being considered. The proposed fuel element consists basically of a sphere with a fuelled core and an unfuelled fission product retentive coating. In the core, $PuO_2 - ThO_2$ fuel particles are dispersed in BeO, which acts as both fuel dispersant and moderator. For the coating, two possibilities have been considered: dense BeO coatings and modified ceramic glazes.

This report describes the development and testing of glaze–type coatings. The development of BeO coatings, which proceeded to some extent in parallel, will be reported separately. All work described here has been carried out on an analogue system in which particles of $UO_2 - ThO_2$, rather than $PuO_2 - ThO_2$, are dispersed in BeO.

2. DISCUSSION OF GLAZE–TYPE COATINGS

2.1 Purpose of Coating

The purpose of the unfuelled coating on the proposed fuel element sphere is to reduce the release of gaseous (and solid) fission products from the core of the fuel element to the coolant to an absolute minimum, and so minimise difficulties that would be associated with a highly active primary coolant circuit. Release mechanisms from uncoated fuel would include direct recoil from exposed fuel particles, diffusion from exposed fuel particles, diffusion from fuel particles to open pores in the BeO and rapid escape therefrom by gaseous permeation, recoil into the BeO matrix and diffusion therefrom, and corrosion, erosion and wear of exposed particles. Direct recoil, corrosion, erosion and wear can be prevented by practically any coating which itself remains chemically and mechanically stable during its lifetime. On the other hand prevention of direct escape of gaseous fission products diffusing from fuel particles or BeO matrix requires a completely non–porous coating. Although lattice diffusion of fission product atoms through the structure of even a dense coating is still theoretically possible, this is a slow process and there should be no difficulty in choosing a coating thickness such that escape by diffusion is negligible over the fuel element lifetime.

2.2 Coating Design Parameters

It will be assumed that the fuel element must operate for long periods (up to 3 years) at temperatures up to 1200 °C and for short periods, during transients only, in the region of 1350 °C. It will be further assumed that the fuel particle size is in the range 150 – 200μ. The BeO sintering temperature is taken to be 1500 °C and a recoil distance of 15μ is assumed for fission product atoms recoiling into either BeO or the coating.

2.3 Suitability of Glaze–Type Coatings

Ceramic glaze compositions readily fuse to form impermeable glasses, and since glaze application is a simple, well–established process, this class of materials presents an attractive possibility for fission–product retentive coatings.

2.3.1 Glaze thickness

For complete prevention of recoil escape from fuel particles at the surface of the core the glaze need be only 20μ thick. However, the additional requirement that diffusional escape of fission product atoms recoiled into the coating must be acceptably low over the fuel element lifetime must also be considered. The required glaze thickness will depend on the diffusion coefficient of fission product atoms in the glaze and on the release level considered acceptable. The appropriate diffusion coefficients are not known, but if a 100μ (0.004 inch) thickness is considered, and if a release of 20 p.p.m. of all fission products formed in the fuel element over a 3–year lifetime is allowed, it can be calculated that the appropriate diffusion coefficient should be no higher than 2×10^{-13} cm^2/sec. This is much higher than D for xenon in dense UO_2 at 1200 °C, but slightly lower than that for Be in BeO. However, on this basis 100μ is considered to be a suitable working thickness for glaze development.

2.3.2 Maturing temperature

During operation of the proposed reactor, 1 inch diameter glazed spheres would be in contact under a load of up to 60 pounds at temperatures up to 1200 °C. Under these conditions a conventional high temperature glaze would soften and adhere at points of contact. A suitable glaze should thus have a maturing temperature above 1200 °C but not so high as to cause over-sintering in the BeO matrix of the fuelled spheres (sintered at 1500 °C), and the glaze should not react appreciably with the BeO matrix. Ideally the glaze coating should mature over a narrow temperature range near 1450 °C and the softening range must be sufficiently high to withstand compressive deformation and adherence under operating conditions for temperatures up to 1200 °C.

2.3.3 Fabrication problems

A uniform coating around a sphere can only result if during fusion the glaze melt remains viscous, as otherwise flow towards the lowest point will occur. This requirement should cause no insuperable problem.

However, an inherent difficulty in the glazing process is the support of the glazed sphere during firing. The conventional method of placing a sphere on a suitable stilt would introduce several small pinholes in the glazed surface. Provided that the glaze remains viscous during fusion it may be possible to support the sphere on a bed of refractory grain without damage to the coating. The most undesirable case is that where spheres are supported upon ceramic stilts causing the formation of three pinholes in the fired coating of each sphere. The importance of these can be gauged by comparing the total area exposed to the coolant from this cause to that from diametral cracking of 1 inch diameter spheres in service. If a pinhole is assumed to be circular with a diameter of 0.020 inch, the area exposed is equivalent to the contribution from one sphere in 1700 cracked in service. Since actual cracking rates in a real system could be higher than this, it can be seen that the contribution from 0.02 inch diameter pinholes is unlikely to be serious, and glazing by supporting on stilts would be allowable.

2.3.4 Other requirements

It can be expected that with some development glazes can be prepared to satisfy the above thickness, maturing temperature, and fabrication requirements.

Carefully chosen glazes are potentially capable of satisfying the following additional requirements of a coating:

(i) The thermal expansion coefficients of BeO and glaze material should be closely matched to prevent crazing or peeling during thermal cycling.

(ii) The glaze should not react with or dissolve fuel either during fabrication or in service as this would accelerate the escape of fission products from the fuel element. While some reaction and possibly inter-solution can be expected at 1450 °C between UO_2 or $UO_2 - ThO_2$ and many glaze components e.g. K_2O, SiO_2, Al_2O_3, MgO, TiO_2, and ZrO_2, it may be possible to keep such reactions within acceptable limits by the use of short maturing times. Reaction rates at 1200 °C should be much slower again, and this should allow satisfactorily low reaction rates during service.

(iii) The glaze composition should be free from materials of high neutron absorption cross section such as B_2O_3 and Li_2O.

(iv) The glaze should be structurally stable when maintained for long periods at the maximum operating or lower temperatures, in the presence of high neutron fluxes and high local fission product fluxes.

3. GLAZE DEVELOPMENT

3.1 Philosophy

Since many of the properties discussed above were not known with certainty for a wide range of glaze compositions and in many cases could only be determined by direct measurement, it was decided to undertake glaze development in order first to obtain an adherent non-porous glaze of acceptable composition, maturing over the required range and able to withstand repeated thermal cycling without cracking or crazing, and then to assess its fission product retentive properties and possibly its longer-term irradiation behaviour. The glazes were to be applied initially to planar surfaces of dense BeO-$(UTh)O_2$ dispersions rather than to the surfaces of spheres.

To initiate the programme, a conventional porcelain type glaze maturing at 1290°C was applied to high density BeO cubes to ascertain whether a standard glaze would adhere, cover the surface, and remain free of defects. The promising results led to the development of glazes maturing at higher temperatures and this work is described in the following sections.

3.2 Preparation and Examination of Glazes

Glazes were prepared from high quality English china and ball clays, feldspar, and high purity ceramic oxides. Glaze compositions were calculated assuming the theoretical compositions for these materials, and in Tables 1 – 3 are always quoted as mole percentages of constituent oxides in the fired glaze.

The glaze batches were wet ground for four hours in a vibratory mill using dense alumina cylinders in a porcelain jar. The water-ground suspension was sieved through a 200 mesh screen and its specific gravity adjusted between 1.4 and 1.6.

A thickness of 0.004 inch was the aim in all glazing experiments. Glazes containing no BeO were applied by spray, brush, or dip techniques in a vented Perspex booth. Those containing BeO were applied in a similar way in a totally enclosed glove-box. The glazes were first applied to ½ inch cubes of hot pressed Pechiney BeO (99% theoretical density) which had been annealed in air at 800°C to remove traces of carbon introduced during hot pressing. Cubes were chosen to provide flat surfaces and edges for observing differences in thermal expansion between the glaze and matrix which could either result in a "crazed" surface or "peeling" at the edges. An indication of "fluidity" of the glaze could be ascertained from the thickness of the glaze at the edges. Glazes were fired in a Super Kanthal Furnace (in air) heated at approximately 250°C per hour and were maintained at the maximum temperature for 2 hours. After firing, the average glaze thickness was approximately 0.004 inch, determined by measurement.

From each of the various glaze types, compositions were selected having the best surfaces (free from pinholing and crawling) and these were applied to BeO cubes containing a 10 v/o dispersion of 200μ $(UTh)O_2$ particles, the U:Th ratio being 1:50. A glaze which will cover BeO satisfactorily may, when applied to fuelled BeO, (i) fail to wet and cover the fuel particles, (ii) react with fuel particles, and (iii) dissolve the fuel particles.

Glazes applied to BeO and fuelled BeO were initially assessed by macro-examination at 20X magnification. Those free from obvious defects were sectioned and polished by normal metallographic techniques and a closer study of their microstructure was made. Selected compositions of the various series were submitted to a thermal cycling test in which the specimen temperature was varied between 40°C and 900°C with a heating and cooling rate as shown in Figure 1. Any mismatching of expansion between the BeO matrix and the coating could be detected by the appearance of crazing on the flat surface or peeling at the edges.

3.3 Glaze Types

The glazes developed form two groups: Group I, containing no BeO, and Group II of varying BeO content. Group I glazes can be classed as one of three types according to whether their structures were largely amorphous, semi-crystalline or predominantly crystalline. Group II glazes were predominantly crystalline.

3.4 Group I Glazes

An established glaze was used to initiate development of this group. Its composition is typical of a "frit free" high temperature glaze used for most commercial applications. The composition A, Table 1, has a maturing temperature of 1290°C. Preliminary observations showed good adherence between the glaze and BeO matrix, and metallography revealed that the glaze had an amorphous structure. The glaze was not affected after 50 thermal cycles. Several modifications of varying composition, aimed at attaining a maturing temperature of 1450°C, were developed from this glaze, and of these composition B was selected. Glaze B was free from surface defects and gas bubbles when applied to BeO cubes and withstood 50 thermal cycles without rupture. To determine the effect of application over fuel particles, the glaze was sprayed onto fuelled BeO cubes; on firing, bubbles developed in the glaze directly above a high proportion of the fuel particles, producing a small crater in the glaze surface. A brown discoloration was observed in the glaze near the fuel particles (under low magnification) and was attributed to dissolution of the $(UTh)O_2$. Likely contributors to dissolution of the $(UTh)O_2$ were thought to be the high K_2O and SiO_2 content of the glaze (see Section 5).

The high silica content would also be an objectionable component in chemical reprocessing of the fuel should such a coating be incorporated on fuelled spheres in a reactor system. To reduce the K_2O and silica contents, several glazes were developed from composition B in which the feldspar was decreased and the silica reduced by replacement with ZrO_2, ZrO_2–TiO_2, and TiO_2. The most significant features of these glazes are now described.

3.4.1 $SiO_2 - ZrO_2$ based glazes

The free silica in composition B was progressively replaced by ZrO_2. The glazes with above 9.5 mole per cent. of ZrO_2 were too viscous and had defective surfaces when fired at 1450°C. Those below 9.5 mole per cent. ZrO_2, on firing at 1450°C, showed pinhole-free surfaces but contained occasional entrapped bubbles. Three that are representative of the latter group are compositions C, D and E, Table 1. Glaze C, when applied to unfuelled BeO, showed a largely amorphous structure by metallographic examination (Figure 2). C and D showed an increase in crystalline phase with increased ZrO_2 content; all three withstood 50 thermal cycles. Glazes C, D, and E and others containing less than 9.5 mole per cent. of ZrO_2 were applied to fuelled BeO and fired at 1450°C. In each case voids formed in the glaze above the fuel particles.

3.4.2 $SiO_2 - ZrO_2 - TiO_2$ based glazes

The silica in the preceding series was then further replaced by TiO_2. Most of the compositions examined by firing on BeO resulted in glazes having good surfaces free from porosity and all withstood 50 thermal cycles. Metallographic examination showed that their structure was part-crystalline, with a crystalline phase dispersed in an amorphous matrix, the amount of crystalline phase increasing with TiO_2 content. Three selected glazes of this series are F, G and H, Table 1; Figure 3 shows the microstructure of glaze H indicating a predominantly crystalline structure. Glazes of this series, like those of the $SiO_2 - ZrO_2$ series, showed void formation above fuel particles when applied to fuelled BeO. Evidence of reaction with the fuel particles can be observed from metallographic examination of glaze G, Figure 4. All glazes in this series withstood 50 thermal cycles, with the exception of H, in which cracks developed after 20 cycles.

3.4.3 $SiO_2 - TiO_2$ based glazes

Complete replacement of the zirconia by TiO_2 led to a series from which I and J, Table 1, were selected as the most interesting. TiO_2 addition lowered the fusion temperatures, I and J maturing as low as 1310°C. When sprayed onto BeO, both glazes withstood 50 thermal cycles without rupture. Metallography revealed that both consisted of an amorphous matrix with a high proportion of crystalline phase. When applied to fuelled BeO, no void formation was observed above fuel particles and no obvious evidence of attack of the fuel particles was detected by metallographic examination.

The significant difference between this and the two preceding series was the ZrO_2 content. The absence of voids above fuel particles was thought to warrant development of this glaze to give a

higher fusion temperature without appreciably altering the TiO_2 content. A progressive series with increasing SiO_2 and Al_2O_3, and decreasing CaO and BaO, content was prepared from which composition K , maturing at 1450 °C, was selected. This composition gave the best fired surface, was free of entrapped porosity, and was unaffected by 50 thermal cycles. When applied to fuelled BeO, voids occurred above fuel particles with metallographic evidence of fuel particle attack; this is shown in Figure 5.

3.5 Group II Glazes

Incorporation of BeO in the glaze compositions was expected to give a coating approaching more closely the nuclear and physical properties of the fuelled matrix. Initial coatings contained BeO, K_2O, MgO, CaO, Al_2O_3, and SiO_2 in varying molecular proportions. The most fusible of these compositions was L , Table 2, which fused at about 1500 °C. This coating appeared dense and crystalline but badly cracked (Figure 6). Cracking resulted from high shrinkage during fusion and the low proportion of glass present in the melt. Feldspar additions were made to L to lower the fusion temperature and provide additional glass to increase the plasticity of the glaze during fusion.

Other variations in composition were investigated and from the series prepared, M, N, O, P, Q, R, S, and T have been selected as the most attractive. Compositions containing more than approximately 40 mole per cent. of SiO_2 when applied to BeO, showed considerable cracking after firing. Glazes below 40 mole per cent. showed no cracking or apparent defects; all showed a largely crystalline structure in an amorphous matrix. These latter compositions were applied to fuelled BeO and were found to cover the fuel particles without voids; some attack of the fuel was revealed by metallographic examination as shown by composition T (Figure 7). Glazes Q, R, S, and T after 50 thermal cycles had crack-free surfaces, and no physical change in the glaze structure could be noted after 500 hours at 900 °C in air.

3.6 Miscellaneous Coatings

A predominant feature of all glaze compositions in Groups I and II is the presence of K_2O incorporated (as feldspar) to act as the primary flux and promote glass formation. Compositions low in feldspar showed poor surfaces and invariably "crawled". The observed tendency for dissolution of the fuel by these glazes could have been enhanced by their K_2O content (see Section 5). A large range of feldspar-free compositions was studied but all showed an inherent tendency to "ball up" on firing. Table 3 shows a selection of these coatings. Compositions A – F, although dense, were unsatisfactory in some cases because they would not cover the fuel particles, and in others because of glaze – fuel particle interaction. Compositions G and H sprayed onto BeO cubes and fired at 1500 °C formed very porous coatings. These compositions were formed into small compacts at 1/4 t.s.i. and when fired at 1500 °C attained a density of 93% theoretical. Negligible increase in densification was achieved with higher compaction pressures. Additional problems were foreseen in applying coatings requiring compaction for densification, particularly in achieving a uniform thickness in a thin coating (0.004 inch or 100μ).

4. TESTS ON GLAZED SPECIMENS

4.1 Fission Product Release Tests

At this stage in the development of a suitable fission product retentive glaze, the ability of a glaze to retain fission products within fuelled BeO was investigated. Two of the most likely compositions were selected. Group I glazes all showed visible evidence of dissolution of the fuel, and void formation above the particles. Compositions Q, R, S and T in Group II showed some reaction with the fuel but no void formation, and thus appeared more attractive. Of these, Q and T were chosen for retention tests since they differed significantly in BeO and TiO_2 content. Both were sprayed onto fuelled BeO specimens trepanned from a hot-pressed dispersion of 100 – 200μ $(UTh)O_2$ in BeO of overall composition U:Th:Be = 1:50:900. The average glaze thickness after firing was 0.004 inch. Glazed and unglazed specimens were irradiated in a thermal flux of 1×10^{13} to 2×10^{13} neutrons/cm^2 for 8 hours at approximately 100 °C and then annealed out-of-pile at 800 °C, 1000 °C, and 1100 °C. The resultant Xe133 release was measured and compared for glazed and unglazed specimens; results are given in Table 4. Ideally the release from glazed specimens would be zero and the results for G and H glazes are therefore disappointing. As the glazes were non-porous, it appeared that fuel migration in the glazes could have been the primary reason for the high release values.

4.2 Microanalyser Examination

Fuelled specimens glazed with Q and T compositions were scanned across the glazed surface with an X-ray microanalyser probe. In both glazes, the presence of uranium and thorium was detected at the external surface, particularly in the region directly above fuel particles. Uranium concentrations in the glaze of up to 10 per cent. of that in the fuel particles were estimated, and this observation confirmed that considerable reaction between the glaze and fuel had taken place. This interaction apparently allows uranium atoms to reach the outer surface of the glaze and fission products can then readily escape by recoil (during irradiation) or via a short diffusion path (during annealing).

4.3 Fission Release From Fuelled BeO Specimens Acid-Leached Before Glazing

Fuelled BeO specimens, similar to those used in the tests described in Section 4.1, were treated four times with boiling 15N HNO_3 + 0.04N NH_4F to remove the fuel at the surface before applying a glaze coating; under low magnification the fuel appeared to have been completely dissolved. The leached specimens were glazed with compositions Q and T and subjected to fission product release tests as in Section 4.1. The results showed only a small decrease in fission product release compared with the unleached glazed specimens. The unexpectedly high release could result from either incomplete dissolution of fuel from narrow crevices or from glaze reaction with particles below the surface via interconnected pores.

4.4 Adhesion Tests of Glazed Surfaces

Tests were made to determine whether glazed coatings applied to spheres would result in adhesion between spheres under simulated reactor conditions.

Pairs of specimens with rounded ends, machined to a radius of ¼ inch, were glazed with composition T. The rounded ends were held together under a 60 lb compressive load for 5 – 7 days at 1020°C. Unglazed specimens were similarly tested. The glazed specimens were found to adhere, although the "neck" could be readily fractured. The unglazed specimens showed no adhesion or deformation at the contact points.

5. SUMMARY AND DISCUSSION

Glaze coatings maturing at about 1450°C have been developed from a conventional glaze. Metallographic examination of these coatings has shown them to vary in structure, depending on composition, from largely amorphous to predominantly crystalline, with no observable porosity. Such coatings would, in principle, appear able to prevent fission product escape by recoil and permeation. Some escape by recoil plus diffusion would occur, but at operating temperatures, owing to the slowness of this process, total fission product release over the fuel element lifetime could be negligibly low.

Two glazes selected as the most promising ones developed, were applied to fuelled BeO and subjected to irradiation to measure their ability to retain fission products. Release was higher than expected but could be attributed to dissolution of the fuel by the glaze, giving rise to fuel migration to the surface and fission product release therefrom because of the short diffusion path (on annealing). Common to these and most of the glazes developed was the incorporation of feldspar and this would appear to be the major contributor to fuel attack; at the maturing temperature, the K_2O in the feldspar would react to form uranates. Silica is known to form a liquid phase with UO_2 at approximately 1650°C (Lang and others 1956) and it would be expected that the temperature of formation of this phase would be considerably lower in the presence of K_2O. The pronounced solid solubility between ZrO_2 and UO_2 (Lambertson and Mueller 1953) would also be expected to enhance dissolution of fuel particles by ZrO_2– containing glazes. Elimination of the feldspar reduced fuel dissolution but resulted in glazes which cracked badly during firing, owing to the absence of glass.

Glaze type coatings applied directly to fuelled BeO do not therefore appear promising as a means of preventing fission product release; the load-bearing and non-adherence properties of glaze coatings under operating conditions are also in doubt. Further development work in this field has now ceased in favour of the development of glass-free sintered BeO coatings.

6. REFERENCES

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Lambertson, W.A., and Mueller, W.H., (1953). – J. Am. Ceram. Soc., 36, 367.

7. ACKNOWLEDGEMENTS

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TABLE 1 – GROUP I GLAZES

GLAZE COMPOSITION – MOLE PER CENT. COMPONENT

Glaze Designation	K_2O	CaO	BaO	Al_2O_3	SiO_2	TiO_2	ZrO_2	Maturing Temp. °C	Remarks
A	4.82	5.43	1.65	9.43	78.64	–	–	1290	Amorphous; void formation above fuel particles
B	4.73	2.74	0.88	1.24	90.40	–	–	1450	,, ,, ,,
C	3.87	5.30	1.86	10.21	72.03	–	6.72	1450	Some crystalline phase in an amorphous matrix; void formation above fuel particles
D	4.17	5.43	1.97	8.38	70.60	–	9.42	1450	,, ,, ,,
E	2.73	5.33	1.99	6.85	73.71	–	9.36	1450	,, ,, ,,
F	2.55	5.11	1.80	6.53	60.32	14.67	8.99	1450	Crystalline phase in an amorphous matrix; void formation above fuel particles
G	2.73	5.47	1.92	6.99	41.80	31.46	9.62	1450	,, ,, ,,
H	2.82	5.62	1.97	7.18	25.64	46.87	9.89	1450	Predominantly crystalline with small amount of amorphous matrix; void formation above fuel particles
I	3.16	1.31	5.45	8.06	28.69	53.32	–	1310	Some crystalline phase in an amorphous matrix; particles were covered without void formation
J	2.75	7.68	3.47	9.35	29.80	46.92	–	1310	,, ,, ,,
K	2.65	1.50	0.87	13.20	37.49	44.29	–	1450	Some crystalline phase in an amorphous matrix; void formation above fuel particles

TABLE 2 – GROUP II GLAZES

GLAZE COMPOSITION – MOLE PER CENT. COMPONENT

Glaze Designation	K_2O	CaO	BaO	BeO	Al_2O_3	SiO_2	TiO_2	Maturing Temp. °C	Remarks
L	0.336	0.453	0.170	8.364	9.006	81.66	–	1500	Crystalline; coating badly cracked
M	3.82	1.21	3.12	24.20	5.48	58.31	3.82	1450	" " " "
N	2.62	3.07	1.11	37.65	4.27	51.24	–	1450	" " " "
O	2.99	2.95	2.95	40.09	7.235	43.778	–	1450	" " " "
P	2.85	4.84	0.68	40.19	6.73	44.69	–	1450	" " " "
Q	3.25	3.05	4.27	28.90	5.30	23.55	31.67	1450	Crystalline; no cracking; covered fuel without void formation
R	7.17	1.64	4.20	39.83	8.35	38.81	–	1450	" " " "
S	4.45	1.45	3.78	47.70	7.56	35.04	–	1450	" " " "
T	4.30	1.48	3.59	55.20	6.55	28.88	–	1450	" " " "

TABLE 3 – MISCELLANEOUS COATINGS

MOLE PERCENT. COMPOSITIONS

Glaze Designation	Al_2O_3	CaO	MgO	English Ball Clay	ZnO	SiO_2	Calcined Wyoming Bentonite	UOX BeO	Sintering Temperatures	Remarks
A	62.0	38.0	–	–	–	–	–	–	2 hr 1450	Dense, "balled up" coating
B	60.0	15.0	25.0	–	–	–	–	–	½ hr 1450	" " "
C	53.8	26.8	9.0	10.4	–	–	–	–	½ hr 1450	Dense, almost continuous coating, does not cover fuel particles
D	51.5	26.5	8.5	–	9.0	4.5	–	–	½ hr 1450	Dense, continuous coating, does not cover fuel particles
E	–	6.0	–	47	–	–	47.0	–	½ hr 1450	Dense, almost continuous coating, reaction with fuel particles
F	30.0	–	–	–	–	–	70.0	–	½ hr 1450	Dense, almost continuous coating, reaction with fuel particles
*G	–	–	25.0	–	–	–	–	75	½ hr 1500	93% dense
*H	–	–	10.0	–	–	–	–	90	½ hr 1500	95% dense

* Compacts – pressed ½ t.s.i.

TABLE 4

Xe^{133} RELEASE COMPARED FOR GLAZED AND UNGLAZED FUELLED SPECIMENS

Glaze Composition	Annealing Temperature °C	Annealing Time (hours)	Xe^{133} Cumulative Fractional Release Relative to Unglazed Specimens
Q	800 1000 1100	1½ 19 6½	2% 8% 24%
T	800 1000 1100	3 18½ 7	2 – 3% 4% 8%

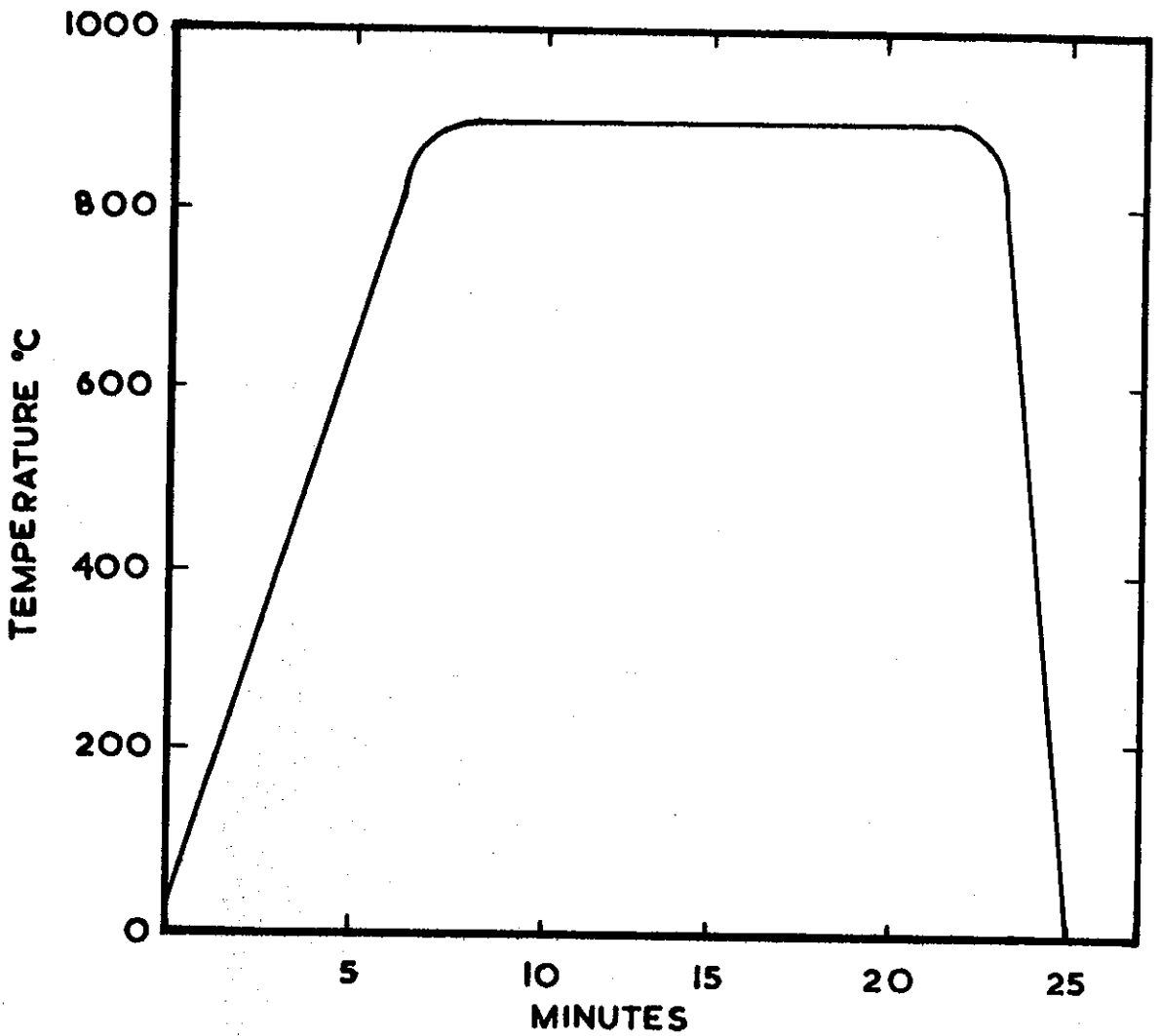
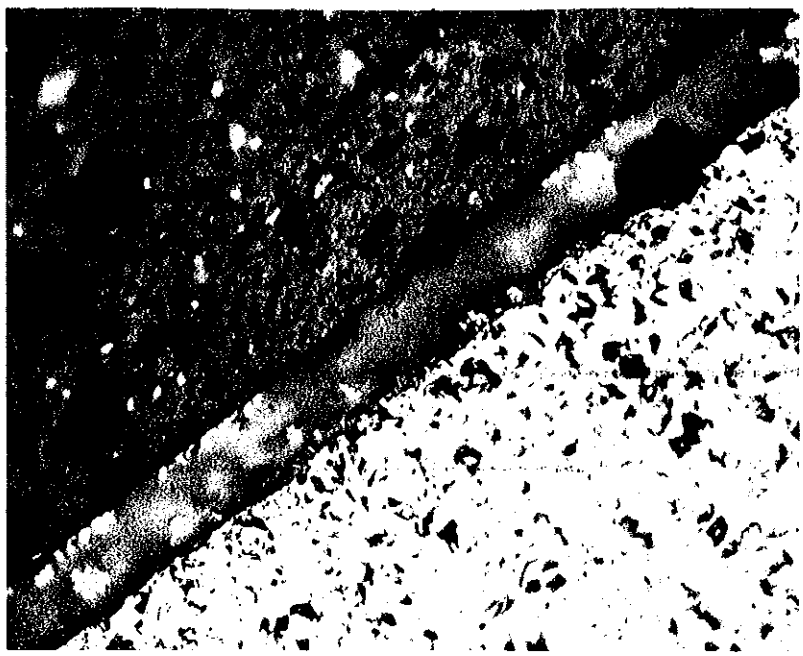


FIGURE 1. THERMAL CYCLING CURVE

P876

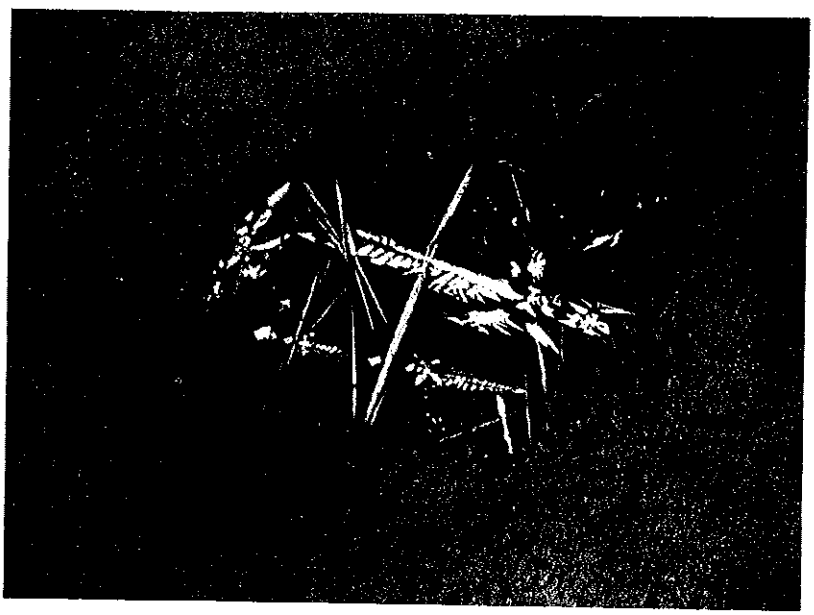


X 250

FIGURE 2 TYPICAL MICROSTRUCTURE OF AN ALMOST AMORPHOUS GLAZE (GLAZE C, TABLE 1)

X 250

FIGURE 3 TYPICAL MICROSTRUCTURE OF A PREDOMINANTLY CRYSTALLINE GLAZE (GLAZE H, TABLE 1)

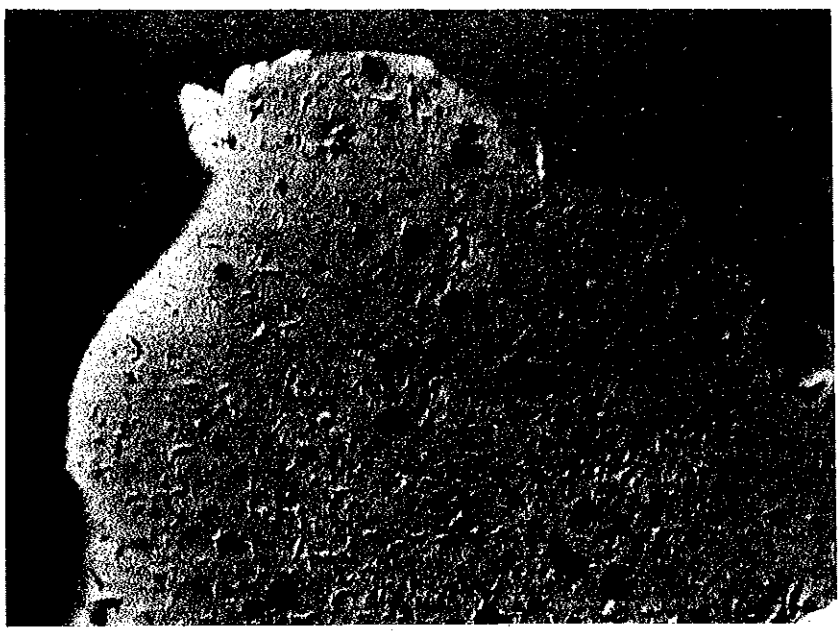


X 250

FIGURE 4 COMPLETELY DISSOLVED FUEL PARTICLE
(GLAZE G, TABLE 1)

X 250

FIGURE 5 PARTIALLY DISSOLVED FUEL PARTICLE
(GLAZE K, TABLE 1)



X 30

FIGURE 6 BADLY CRACKED CRYSTALLINE GLAZE
(GLAZE L, TABLE 2)

X 250

FIGURE 7 PARTIALLY DISSOLVED FUEL PARTICLE
(GLAZE T, TABLE 2)