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A RADIOCHEMICAL STUDY
OF THE MECHANISM OF POLISHING GLASS

by

JOHN GORDON SMITH

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ABSTRACT

In an effort to determine whether glass flows when polished uranium glasses were fused to non uranium glasses and polishings carried out in a direction from the active to the inactive side. Soda-lead-silicate and phosphate glasses were polished with rouge and ceric oxide. To locate any of the uranium glass which may have been transferred during the polishing, nuclear track plates were used. Thus by autoradiographs taken before and after polishing, any alpha particles from the uranium glass transferred to the side of the non uranium glass would have registered on the developed plates. This method was capable of detecting a 4.5 A° flowed layer or chips of uranium glass 0.019 mms in diameter. No evidence of any flow, greater than over a distance of 0.2 mms , was found by the authors.

Experiments were also carried out using radioactive ceric oxide. Thus if the contact temperatures reached during polishing approached the softening point of the glass it was reasoned that the agent might have become fused with the polished surface. By using this method it would have been possible to detect 2.3×10^{-7} grams of CeO_2 spread over 6 cm^2 on the surface of the glass. By assuming that the CeO_2 would fuse with and become part of the glass it was possible to detect a 15 A° layer of this changed glass, if 10% of this changed glass was CeO_2 . The authors found no evidence of any fusion of the CeO_2 with the glass under the conditions of polishing used in this project.

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INTRODUCTION

Before polishing glass, the surface is ground to the desired contour with an abrasive such as emery, alumina, or carborundum slurried with water on a revolving iron form. This process is recognized to be one of chipping particles of glass away from the surface and leaves the latter rough and translucent.

In the ensuing polishing, a finer powder is used, slurried with water on moving felt or wax. The powder particles must be hard and insoluble and have a melting point higher than the softening point of the glass being polished. They must also not be too fine as shown by Sir H. Jackson's failure to polish¹ glass with the very fine powder obtained from an iron arc. Rouge and ceric oxide are both in common use today and were used in our work.

There are three schools of thought about the mechanism of the polishing operation. The first claims that it is simply a continuation of the grinding operation, a chipping off process, continued to such a degree of fineness that the reflected light waves cannot show any measurable amount of straying. The second believes that the frictional heat of polishing raises the surface temperature to the softening point of the glass and thereby smooths the surface by an actual flow of glass. The third claims a combination of these two mechanisms.

The evidence for these alternatives will now be considered.

POLISHING IS A CONTINUATION OF THE GRINDING PROCESS

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Sir George Beilby reported that the flowed layer on crystals, such as calcite, was vastly tougher than the crystalline material below. He interpreted this toughening as being due to a non-crystalline flowed layer. This layer was considered to be tougher for it was able to resist the cutting action of the agent by which the original surface had been satisfactorily ground. Preston,³ however, found nothing of this sort with glass for he was unable to detect any progressive toughening of the glass with prolonged polishing. He found that material came off steadily at the same rate whether the glass was quite "grey" or had been polished for many hours. From this he concluded that the flowed film, if it existed at all, had no direct influence on the process and that mechanical abrasion, i.e., the complete removal of glass was the conspicuous feature.

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Further evidence supporting this mechanism may be obtained from the method Strong recommends for the polishing of precision lenses and prisms. He uses wax instead of felt as the base, and recommends that the contour of the wax base be altered in order to remove any high spots which may have been left on the surface after the initial polishing. This procedure of altering the wax base is continued until the surface becomes planar.

This method of eliminating high spots suggests that the important feature of the polishing operation is that of a removal of glass.

THE SURFACE IS SMOOTHED BY AN ACTUAL FLOW OF GLASS

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Dreyer and Ertel used a technique which afforded a means of differentiating between a surface which had been polished and a surface which had been ground. They applied a light polarizing solution called "polacoat" to the surface being studied. This solution on drying takes on the orientation of the surface with which it is in contact. When this solution was applied onto a polished surface a uniform polarized film resulted and left a visible record of the surface, while with a rough surface the polarization was non-uniform. They removed a polished layer on plate glass by rubbing with whiting, an abrasive material. Then, when "polacoat" was applied to this surface, the marks made in the original grinding of the glass reappeared. They explained this by a flow, i.e., the original grinding marks were flowed over during polishing.

Another argument in favour of the flowed layer is the so-called burn which is usually made when the table is allowed to run too dry. Burns may be incrustations of rouge on the glass surface. Chemical tests show these to be insoluble in all organic solvents, and in a great many cases may be removed by hydrofluoric

acid only. However, burns are not necessarily incrustations of rouge and in some cases rouge may be absent. A burn is readily recognizable for it is always raised above the surface of the glass. In view of this it would appear that they may be particles of glass which have been removed during the polishing and re-attached later due to high contact temperatures. Hence it may be reasoned that if such high contact temperatures are reached during polishing it may be possible that there is a smoothing action at work.

POLISHING IS A COMBINATION OF FLOW AND REMOVAL

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Bayer, in 1938, pointed out that when an attempt was made to improve a poor polish by further polishing, the effort proved fruitless. He interpreted this as a different process at work during polishing than during grinding. He also stated,

" With proper fine grinding, after twenty minutes of polishing there will be no unevenness, but only scattered depressions of grinding holes. The surface has already assumed an incipient shine and appears to the layman as 'finished'. At closer observation, however, there is still a general blueishness on the glass, which disappears with further working.

This is occasioned by the flowing process,
which takes place with dry polishing material,
when the temperature of the glass surface is
so high that shell edges of extraordinary
small height extenuate into the shell cups ".

Those who claim the operation is a combination of the two processes maintain that glass is removed during the initial stages of polishing. They consider the wet polishing to be a continuation of the grinding, the difference being one of rate. From such considerations it is evident that further polishing will not improve the resulting surface. For if the areas which require polishing lie in surface depressions they would remain untouched during subsequent polishing. They believe that these areas are, however, flowed over during the dry polishing.

GENERAL METHOD OF ATTACK

To determine whether glass flows during polishing a piece of radioactive glass was fused to a non radioactive sample and polishing carried out in a direction from the radioactive side to the non radioactive side. Autoradiographs taken before and after polishing were used to show whether active glass flowed over to the inactive side. Uranium was used as the source of radioactivity because it is an alpha emitter. One reason for

using an alpha emitter was because of the very short range of these particles. Hence alphas originating below the surface would not activate the emulsion for they would be effectively absorbed in the glass. This would result in a sharp boundary between the two glasses on the ensuing autoradiograph. If a beta emitter had been used a distinct boundary would not have resulted because beta particles originating below the surface would have caused a darkening on the side of the inactive glass in the resulting autoradiograph. The other reason that an alpha emitter was chosen is due to the fact that alpha particles register as distinct individual tracks in nuclear track plates. This affords a very sensitive method to detect minute amounts of such radioactive materials. Soda-lead-silicate and phosphate glasses were the types polished and were chosen because of their relatively low softening points.

Another phase of this project was that of using a radioactive polishing compound. Thus if sufficiently high contact temperatures were reached during the polishing it was reasoned that the agent might become fused with the glass, indicating that a flow would be possible. Polishings were carried out using radioactive ceric oxide. After neutron irradiation ¹⁴¹Ce emits beta particles and gamma rays. A Geiger counter was used for detecting beta particles emitted from the radioactive agent incorporated

with the polished glass. In cases where there was a sufficient amount of activity transferred to the surface autoradiographs of the sample were taken to determine the distribution of this activity.

EXPERIMENTAL

PREPARATION OF GLASSES

(a) Silicate Glasses

Fig. 1 illustrates the electric resistance furnace used to prepare the soda-lead-silicate glasses. The resistance wire was 50 mil Platinum 10% Rhodium imbedded in high grade alumdum cement. Some of the advantages this alloy possesses over pure platinum are: the tensile strength (at the operating temperature of the furnace) of the alloy is considerably higher than that of 100% platinum; the rate of vaporization is less than platinum; and the melting point of this alloy is 1860 °C whereas that of 100% platinum is 1773 °C. The temperature of the furnace was controlled by a Wheelco Potentioltrol to ± 20 °C.

Glasses of the following compositions were prepared:

TYPE "A"

Non Uranium Glass

SiO ₂	56.5 per cent
Na ₂ O	14.0 " "
PbO	29.5 " "

SCHEMATIC DIAGRAM OF
ELECTRIC RESISTANCE FURNACE

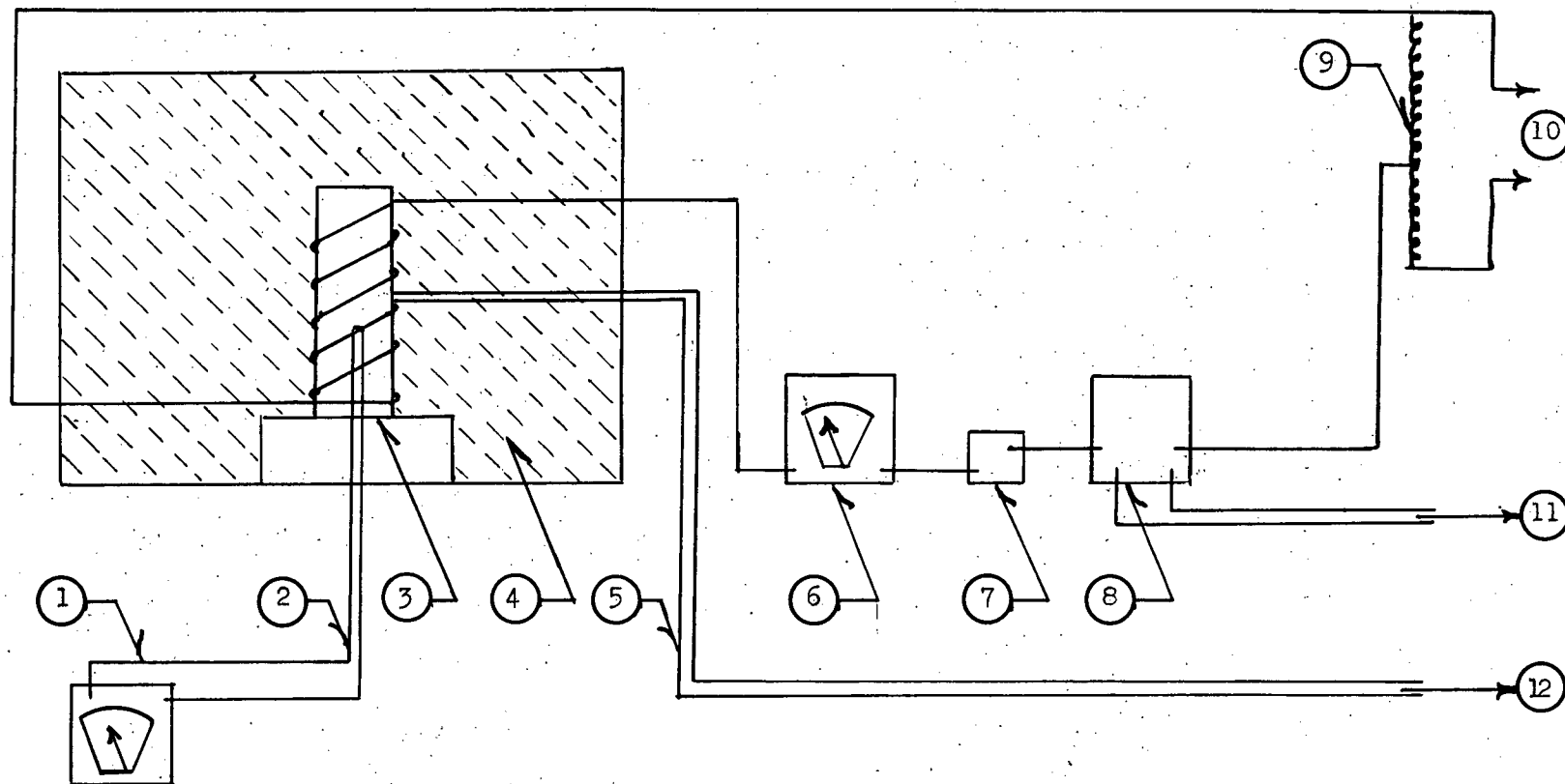


Fig. I

KEY TO DIAGRAM

- (1) Potentiometer.
- (2) Chromel - Alumel Thermocouple leading to inside of oven.
- (3) Oven - clay core consisting of sixteen turns of the alloy.
diameter - 9 cms
height - 17 cms
- (4) Insulation - Finely divided clay.
Size of circular furnace
diameter - 60 cms
height - 43 cms
- (5) Chromel - Alumel Thermocouple leading to outside of oven.
- (6) Ammeter.
- (7) Fuse.
- (8) Cut-off Control.
- (9) Variac.
- (10) Source - 110 volts.
- (11) To Potentiometer.
- (12) To Potentiometer.

TYPE "B"

Uranium Glass

This is composition "A" plus 5% of its weight of U_3O_8 .

SiO_2	53.7	per cent
Na_2O	13.3	" "
PbO	28.2	" "
U_3O_8	4.8	" "

Batch materials used for the above compositions were:

SiO_2 , PbO , Na_2CO_3 , and $UO_2(NO_3)_2 \cdot 6H_2O$. The SiO_2 was in the form of a very fine powder and was obtained from Linde. The batch was melted in a cylindrical platinum crucible, 5 centimeters in diameter and 4.5 centimeters in height, at a temperature of 1160 °C for a period of 12 hours after the batch had become molten. Immediately prior to being poured it was heated to 1260 °C, then poured onto a previously heated graphite tray and placed in an annealing oven. It was kept at a temperature of 532 °C for 30 minutes, then allowed to cool to room temperature at a rate of 5 °C per minute.

(b) Phosphate Glasses

Glasses of the following compositions were prepared:

TYPE "C"

Non Uranium Glass

P_2O_5	67.0 per cent
Al_2O_3	4.0 " "
BaO	19.0 " "
CaO	10.0 " "

TYPE "D"

Uranium Glass

This is Type "C" plus 5% of its weight of U_3O_8 .

P_2O_5	63.7 per cent
Al_2O_3	3.9 " "
BaO	18.0 " "
CaO	9.5 " "
U_3O_8	4.9 " "

TYPE "E"

Non Uranium Glass

This is composition "D" but contains PbO instead of U_3O_8 .

P_2O_5	63.8 per cent
Al_2O_3	3.8 " "
BaO	18.1 " "
CaO	9.5 " "
PbO	4.8 " "

Batch materials used for the phosphate glasses were:

H_3PO_4 , $\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$, BaHPO_4 , $\text{Ca}(\text{H}_2\text{PO}_4)_2 \cdot \text{H}_2\text{O}$, PbO , and $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$.

In the preparation of these glasses the batch was thoroughly mixed in a beaker then added to a clay crucible and placed in an ordinary laboratory blast furnace. After the batch had become molten the glass was allowed to remain in the furnace for a period of 1 hour and 30 minutes. It was then poured onto a previously heated graphite tray and placed in an annealing oven, kept at a temperature of 500 °C for a period of 30 minutes, then allowed to cool to room temperature at the rate of 5 °C per minute.

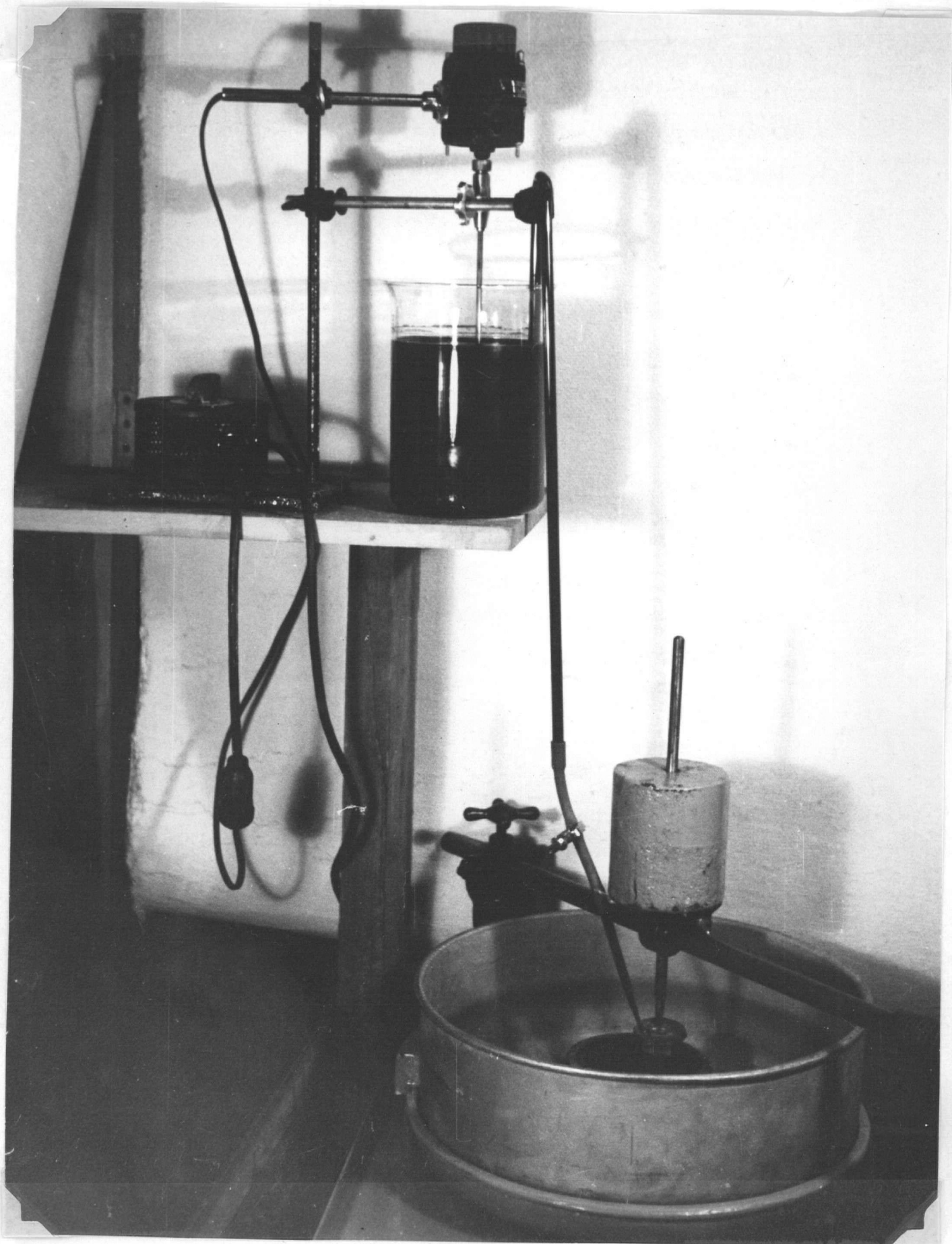
When an attempt was made to join the uranium phosphate glass (Type "D") to non uranium phosphate glass (Type "C") it was found that upon annealing and cooling a fracture developed at the join. To surmount this difficulty it was decided to use Type "E" instead of Type "C". It was hoped that by adding the same percentage of PbO as of U_3O_8 the coefficients of expansion of these two glasses would be similar. However, this also proved fruitless with fractures occurring in the same place. In an effort to make such a join two pieces were placed end to end in a graphite holder, after the ends had previously been polished. Then the holder and glass were placed in a furnace and the temperature raised to 670 °C.

The glass flowed, hence a join was made and the ensuing sample annealed by allowing the furnace to cool to room temperature in a period of 15 hours. This did not completely remove all the strain, for all the samples developed a fracture near this join in the subsequent grinding and polishing procedure. However, only those samples which did not fracture all the way across the join were used. In some cases these fractures resulted in a complete break between the two glasses, the breaks occurring upon the removal of the samples from the discs after polishing, the samples having been adhered to the discs with wax.

GRINDING AND POLISHING EQUIPMENT

The machine used in this experiment was a standard commercial model, the rotating iron disc being 12 centimeters in diameter. The initial fine grinding was performed on this disc, using a fine grade of emery, slurried with water. Polishings were then carried out by using a felt pad 10 centimeters in diameter glued onto the disc. In most of the polishings the standard weight (5.5 kilograms) supplied with the machine was used. To regulate the flow of slurry a gravity fed arrangement was constructed as shown in the attached photograph I. A pinch cock controlled the rate of flow. This was measured before and after polishing and averaged. During polishing the sample was held by hand in such a manner as to ensure that the direction of polish was always in a direction from the active to inactive side.

PHOTOGRAPH I (To follow page 11)



In the polishings made with radioactive ceric oxide the slurry was not applied by this arrangement but poured onto the disc from a beaker. In this case pressure was applied by hand and not by the standard weight. For these experiments the polishing disc was completely covered by a lucite dome with a pair of heavy duty rubber gloves fitted into two conveniently placed holes cut into its wall. In addition to this dome a small circular cellophane holder was built around the rotating disc. These precautions were taken to prevent radioactive contamination to workers and surroundings. Photograph II demonstrates this arrangement.

AUTORADIOGRAPHIC MEASUREMENTS

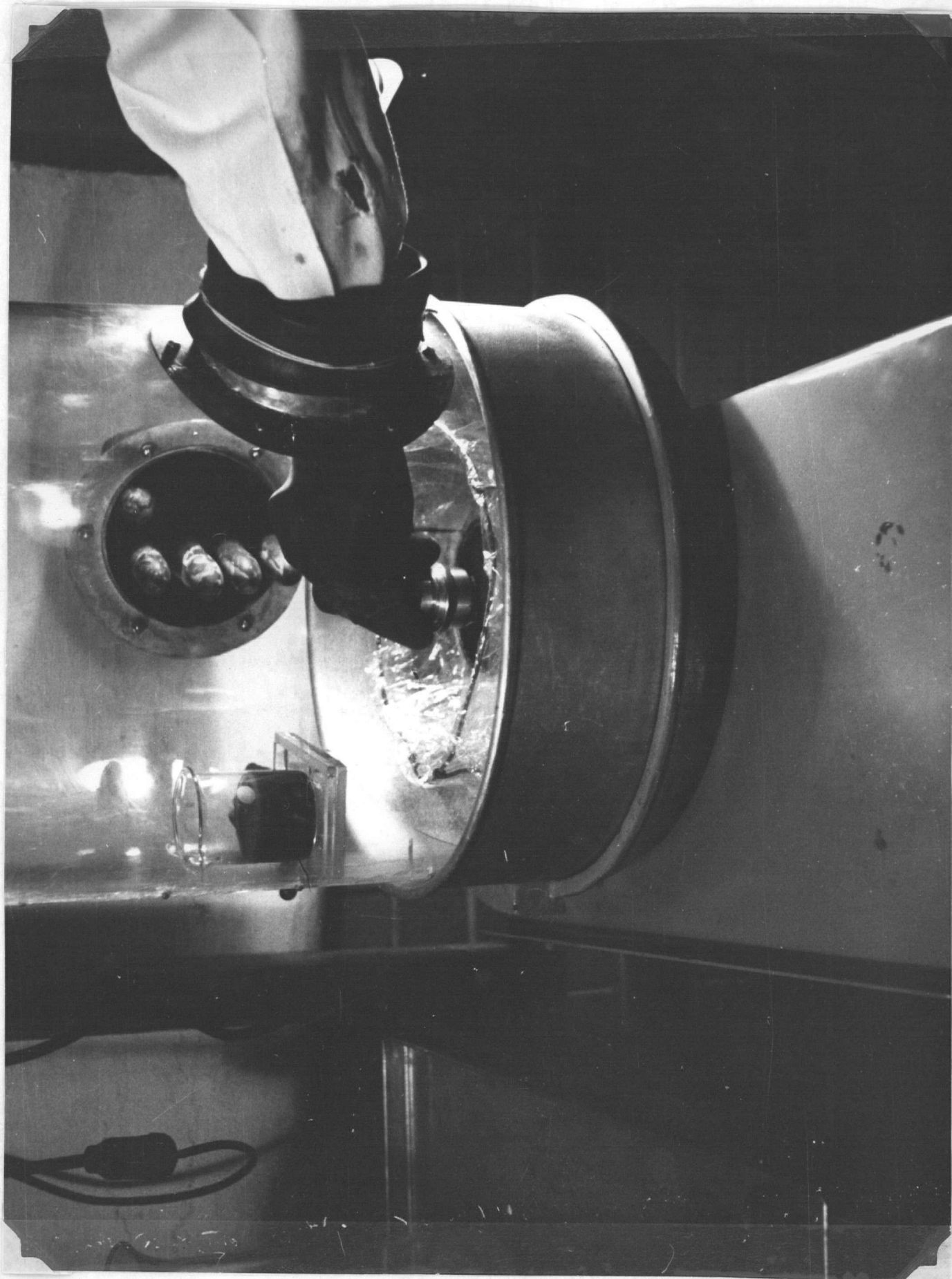
(a) Emulsions

For alpha measurement NTB - 3 plates were used. An alpha particle entering this emulsion is registered as an individual track upon subsequent development of the plate, thus enabling a minute amount of an alpha emitter to be detected. The alpha tracks registered on the plates were counted on a B & L microscope with magnification of 560 X, with an ocular disc in the eye piece.

(b) Development of Plates

The procedure used for the development of the nuclear track plates is given in detail below.

PHOTOGRAPH II (To follow page 12)



- (1) The plates were developed in D - 19, diluted with an equal volume of water at a temperature of 4 °C for a period of 30 minutes. Then two volumes of water at a temperature of 22 °C were added and the resulting solution kept at room temperature for a further period of 30 minutes.
- (2) The plates were then placed in a 2% acetic acid bath at 4 °C for a period of 30 minutes. Strong nitrogen agitation was applied while the plates were in this stop bath.
- (3) Upon removal from the stop bath the plates were placed in the fixing solution, (30% hypo) maintained at a temperature of 4 °C for a period of 1 hour. After this time the fixing bath was allowed to come to room temperature and strong nitrogen agitation was again applied.

The plates began to clear approximately 5 hours after the commencement of the fixing. The fixing was continued for a period of 15 hours. Following this the plates were washed in running tap water for 3 or 4 hours.

(c) Sensitivity of the Photographic Method

Cook and Hudswell⁷ were able to prepare thin films of U_3O_8

on platinum foil or stainless steel by,

- (1) Defining the area to be covered by the U_3O_8 with a solution of nitro-cellulose in ether-alcohol.
- (2) Applying uranyl nitrate in pyridine to this area.
- (3) Heating gently to drive off the pyridine and to destroy the boundary, then they ignited to convert the uranyl salt to U_3O_8 .

They were able to determine the amount of U_3O_8 by weighing, finding it necessary to follow this procedure since losses of uranyl salt or oxide may have occurred during ignition.

For the purpose of this experiment it was not practical to weigh out very small amounts of uranium, therefore it was decided to use the same method as Cook and Hudswell, but to omit the ignition, hence eliminating the necessity of weighing.

A minute amount of uranyl nitrate in pyridine was prepared,
-5
 6.132×10^{-5} grams per ml of solution. Of this solution 0.5 ml was pipetted onto a previously cleaned piece of platinum foil.
2
An area of 9 cms² was defined on the platinum by painting a border with collodion. The pyridine was allowed to evaporate at room

temperature. Finally, to expel any excess pyridine, the sample was very gently warmed. At this point an autoradiograph was taken of this area. The track plate was placed directly over this area and clamped. A calculation giving the theoretical number of alpha tracks that this activated area will give per cm^2 is given below.

Concentration of uranyl nitrate	=	6.132×10^{-5}	gms/ml
Quantity of uranyl nitrate used	=	3.066×10^{-5}	gms
Area covered	=	8.85 cm^2	
Concentration of uranyl nitrate	=	3.47×10^{-6}	gms/ cm^2
Time of exposure of autoradiograph	=	2 hours	
$t_{\frac{1}{2}}$ of uranium	=	4.5×10^9	years
Molecular weight of uranyl nitrate	=	502	gms

The rate of decay of uranyl nitrate for 3.47×10^{-6} gms per hour is given by,

$$\begin{aligned} \frac{dx}{dt} &= -\lambda N \\ &= \frac{.693 \times 6.02 \times 10^{23} \times 3.47 \times 10^{-6}}{4.5 \times 10^9 \times 365 \times 24 \times 502} \\ &= 74 \text{ disintegrations per hour} \end{aligned}$$

But U^{234} is in equilibrium with U^{238} therefore there are two alpha emitters.

Therefore rate of decay, if this amount is spread evenly over one square cm.

$$148 \text{ disintegrations/hour/cm}^2$$

However, only one half of the alpha particles will activate the film. For a 2 hour exposure this will represent

$$148 \text{ alpha tracks/cm}^2 \quad (\text{Theory})$$

Experimental results:

Three different areas of this plate were counted. When this plate was observed under the microscope the distribution of alpha tracks appeared to be random and no clumps were noticed. To determine the background two blank plates were also counted.

Below are the results of these counts.

	<u>Area Counted</u>	<u>No. of alpha tracks</u>
Blanks with no uranium	0.4228 cm ²	59
	0.3881 "	53
Total area	0.8109 cm ²	
Total number of alpha tracks	112 ± 10.6	
Therefore number of alpha tracks/cm ²	138 ± 13	
Sample	0.4101 cm ²	114
	0.4536 "	128
	0.4040 "	111
Total area	1.2677 cm ²	
Total number of alpha tracks	353 ± 18.8	
Therefore number of alpha tracks/cm ²	280 ± 14.7	

Therefore the number of alpha tracks/cm² from the uranyl nitrate is

$$\begin{aligned} 280 & - 138 \pm \sqrt{(13)^2 + (14.7)^2} \\ & = 142 \pm 17 \text{ tracks cm}^2 \end{aligned}$$

If the exposure were increased to a period of 200 hours, 3.47×10^{-8} grams of uranyl nitrate would give this same number of tracks. Thus, the reasonable limit of the sensitivity of this method is reached, for in order to decrease the amount of the nitrate by a factor of 10 this would necessitate prolonging the exposure to 2,000 hours which is impractical.

A similar calculation shows that 1.98×10^{-8} grams of U_3O_8 will give 148 tracks/cm² with a 200 hour exposure. If during the polishing operation 1.98×10^{-8} grams of U_3O_8 were transported from the active side to the inactive side and assuming the glass transported was spread evenly over a square cm of the surface, the depth of this flowed layer would be,

Density of glass 2.5 gms/cm³

Composition of glass 4.8% U_3O_8

Therefore number of grams of glass which contain 3.47×10^{-8} grams of U_3O_8

$$= \frac{100}{4.8} \times 1.98 \times 10^{-8}$$

$$= 4.1 \times 10^{-7} \text{ gms of uranium glass}$$

Volume of this layer

$$= \frac{4.1 \times 10^{-7}}{2.5} = 1.64 \times 10^{-7} \text{ ccs.}$$

This would represent a layer of 16.4 \AA .

However, it would be possible to detect an increase in track count of 40 tracks/cm^2 over the background instead of 148 tracks/cm^2 . This would represent a layer of 4.5 \AA .

This then is the minimum thickness of a layer which could be detected by this photographic method.

(d) Determination of Polishing Compound Transferred to Glass

To detect any of the activated polishing agent that may have fused with the glass during polishing, it was decided to first use the Geiger counter, then when positive results were obtained autoradiographs of the surface were taken. From the following calculation it may readily be seen that the method of detection of betas by the Geiger counter is more sensitive than the method of autoradiography.

20 mcs of activated CeO_2 were available for this project.

$1.0 \text{ mcs} = 3.7 \times 10^7 \text{ disintegrations/sec.}$

$20 \text{ mcs} = 7.4 \times 10^8 \text{ disintegrations/sec.}$

The 20 mcs were contained in 5 grams of the activated CeO_2 .

Therefore 5 grams of activated $\text{CeO}_2 = 4.4 \times 10^{10} \text{ disintegrations/min.}$

If the sample is placed 1.25 cms from the window of the tube in the counter approximately $\frac{1}{4}$ of the total count will register. Hence for the purpose of counting 1.1×10^{10} counts per minute were

available. It was planned to use activated CeO_2 in the ratio of 1:20 with ordinary CeO_2 .

Therefore 10 grams of the mixture would equal 1.1×10^9 cts/min.

It is possible to detect with ease a count of 25 cts/min over the background.

Therefore $\frac{25}{1.1 \times 10^9} \times 10 = 2.3 \times 10^{-7}$ grams of

the mixture are needed to give this count of 25 cts/min over the background.

One of the best films for detecting beta particles is No-screen X-ray film. To darken this film sufficiently to detect beta particles 10^7 disintegrations/cm² are needed. The sensitivity of the photographic method will now be considered and compared with the sensitivity obtainable with the Geiger counter.

$t_{\frac{1}{2}}$ of $\text{Ce}^{141} \sim 21$ days
 $\sim 3 \times 10^4$ mins

Now 25 cts/min on the counter is equivalent to approximately 100 disintegrations/min.

$$\frac{dx}{dt} = -\lambda N$$

Therefore $N = \frac{100 \times 3 \times 10^4}{.693} \sim 4.3 \times 10^6$ atoms of Ce^{141} .

From this it may readily be seen that it is impossible for the film to be darkened by 10^7 beta particles if the activity is spread uniformly over one square cm. However, if the total

count were 1000 cts/min it is obvious from the above that 4.3×10^7 atoms of Ce^{141} would be present. Since one half of the beta particles activate the film approximately 2×10^7 atoms would be available. Therefore it would require approximately 21 days to activate the film. From previous calculations it may be seen that 2.3×10^{-6} grams of the mixture would be necessary to give a total disintegration rate of 1000 dis/min. Thus the counter method is more sensitive than this photographic method by a factor of approximately 10.

The following calculation illustrates the depth a disturbed layer must be in order that it may be detected by the counter method. It is necessary, for the purpose of this calculation, to assume that the glass into which the ceric oxide is incorporated contains 10% of this activated agent. As previously calculated the minimum amount of CeO_2 detectable is approximately 2.3×10^{-7} grams.

$$\text{Number of grams of glass detectable} = 2.3 \times 10^{-6}$$

$$\text{Density} = 2.5 \text{ grams/cc}$$

$$\text{Volume of glass} = 9.2 \times 10^{-7} \text{ ccs}$$

$$\text{Area of the sample polished} = 6.2 \text{ cm}^2$$

$$\text{Therefore depth of the layer} = \frac{9.2 \times 10^{-7}}{6.2}$$

$$= 15 \text{ A}^\circ$$

The following equation gives the relation between the depth of this layer and percentage of incorporated CeO_2 .

$$d \times \% = c$$

Where

d = depth of layer in Angstrom units.

c = constant, in this case equal to 150.

RESULTS

A series of blank autoradiographs were taken at random whenever a new batch of photographic plates were opened and used. Listed below are the results of these blanks from which it was possible to determine the background count. In addition to these blanks autoradiographs of glass, ground from the inactive to the active side, were also taken. In these cases it was most unreasonable to suppose that there would be any probability of glass being flowed onto the inactive side. This series also acted as a check in determining whether there happened to be any radioactivity in the material used to prepare the glass, excluding uranium, of course.

BLANKS

<u>Sample No.</u>	<u>Area Counted</u>	<u>No. of Alpha Tracks</u>
1	0.377 cm ²	52
2	0.325 "	42
3	0.390 "	54
4	0.372 "	51
5	0.342 "	44

Total area 1.806 cm²

Total number of alpha tracks 243 ± 15.6

Therefore number of alpha tracks/cm² 135 ± 8.6

GROUND GLASS AUTORADIOGRAPHS

<u>Sample No.</u>	<u>Area Counted</u>	<u>No. of Alpha Tracks</u>
1	0.403 cm ²	58
2	0.390 "	53
3	0.387 "	52
4	0.396 "	53
5	0.345 "	46

Total area 1.921 cm²

Total number of alpha tracks 262 ± 16.2

Therefore number of alpha tracks/cm² 137 ± 8.5

In the results tabulated below, the method used to record the number of alpha tracks per square centimeter was to mark off

a small square (approximately 0.5 cm^2) on the resulting autoradiograph, about 0.2 cms from the boundary between the active and inactive side. The number of alpha tracks in this area was then counted, use being made of a microscope with magnification of 560 X. For any positive results (by positive results it is implied that uranium glass was flowed over to the non uranium glass) one half of the total of the alpha particles from the uranium would register as distinct tracks in the photographic emulsion. It was possible to arrive at this factor of one half because the glass surface was clamped tightly to the plate. There would have been no correction necessary for absorption of alpha tracks by the glass. Any amount flowed would have been so small that the absorption of alpha particles by this flowed glass would have proved negligible.

The following series of polishings had the undermentioned standard conditions :-

SERIES A

- (1) Type of glass lead glass (Type A & Type B)
- (2) Finely ground with fine emery
- (3) Composition of slurry 25 gms of CeO_2 /1000 mls of water
- (4) Rate of flow of slurry 40 mls/min
- (5) Standard weight of 5.5 kilograms
- (6) Time of exposure of autoradiograph 100 hours

<u>Sample No.</u>	<u>Time of Polish</u>	<u>No. of Tracks per cm²</u>
1A	5 secs	130
2A	10 "	132
3A	20 "	128
4A	30 "	134
5A	1 min	132
6A	2 mins	131
7A	4 "	136
8A	8 "	132
9A	10 "	132

In the above series the time of polishing was increased continually from five seconds to ten minutes, thus, if a flow did take place, it would have been possible to determine at which time during the polishing operation this flow was most pronounced, and if it varied with the degree of polish.

By comparing the number of tracks recorded per cm² on the above autoradiographs with the number of tracks per cm² on the blanks, it is at once obvious that there was no flow during this set of polishings.

It was then decided to increase the amount of polishing agent in an effort to determine whether this variable had any effect on flow. Hence, the amount of polishing agent in the next set was increased from 25 grams to 50 grams of CeO₂/1000 mls of water. Other than this change in composition of the slurry, this series "B" had the same standard conditions as the previous series.

SERIES B

<u>Sample No.</u>	<u>Time of Polish</u>	<u>No. of Tracks per cm²</u>
1B	5 secs	140
2B	10 "	138
3B	20 "	134
4B	30 "	135
5B	1 min	131
6B	2 mins	136
7B	4 "	130
8B	8 "	132
9B	20 "	143

There was no indication of flow, even with twice the rate of addition of ceric oxide.

If the manner of recording results is considered, it is possible to reason that there may have been flow in the previous polishings, but this flow was over such a minute distance that it was impossible to detect it because no glass would have flowed all the way from the boundary to the area counted, i.e., a distance of 0.2 cms. In an effort to answer this question counts were made as close as possible to the boundary for all plates in the "A" and "B" series. This was done by making a single sweep of the autoradiograph, the edge of the field being 0.218 mms from the outermost position of the boundary between the inactive and active sides. This dividing line could readily be seen on the plate, it was very distinct both visually and under the microscope.

<u>Sample No.</u>	<u>Area Counted</u>	<u>No. of Tracks</u>
1A	0.015 cm ²	1
2A	0.033 "	3
3A	0.028 "	4
4A	0.040 "	5
5A	0.034 "	2
6A	0.042 "	6
7A	0.041 "	7
8A	0.043 "	6
9A	0.031 "	4
1B	0.011 "	2
2B	0.026 "	3
3B	0.012 "	4
4B	0.020 "	2
5B	0.019 "	2
6B	0.026 "	3
7B	0.020 "	4
8B	0.021 "	3
9B	0.014 "	3

Total area counted 0.476 cm²

Total number of alpha tracks 64 ± 8

Therefore number of alpha tracks/cm² 138 ± 17

Again no evidence of any increase in track count is apparent, thus indicating that if flow does take place it is over a distance of less than 0.22 mms.

After negative results were obtained using ceric oxide as the polishing compound it was decided to use rouge in order to determine whether this type of agent had any effect on a flow process. Because in the previous series the amount of agent appeared to have no effect in a flow it was deemed unnecessary to vary the amount of the polishing compound. The following set of polishings had the undermentioned standard conditions.

SERIES 'C

- (1) Type of glass lead glass (Type A & Type B)
- (2) Finely ground using fine emery
- (3) Composition of slurry 50 gms of Fe_2O_3 /1000 mls of water
- (4) Rate of flow of slurry 40 mls/min
- (5) Standard weight of 5.5 kilograms
- (6) Time of exposure of autoradiograph 100 hours

<u>Sample No.</u>	<u>Time of Polish</u>	<u>No. of Tracks per cm^2</u>
1C	5 secs	129
2C	20 "	136
3C	50 "	135
4C	100 "	133
5C	200 "	136

Again there was no evidence of any flow.

A phosphate glass was then polished with the thought that its lower softening point would be more conducive to flow. Again it was deemed unnecessary to vary the amount of the agent supplied for reasons already given. The following series had the under-mentioned standard conditions :-

SERIES D

- (1) Type of glass phosphate glass (Type D & Type E)
- (2) Finely ground using fine emery
- (3) Composition of slurry 50 gms of Fe_2O_3 /1000 mls of water

- (4) Rate of flow of slurry 40 mls/min
- (5) Standard weight of 5.5 kilograms
- (6) Time of exposure of autoradiograph 100 hours

<u>Sample No.</u>	<u>Time of Polish</u>	<u>No. of Tracks per cm²</u>
1D	5 secs	137
2D	15 "	130
3D	50 "	141
4D	120 "	136
5D	240 "	129

Again it is readily seen that there was no increase in the background count indicating no flow. Hence this left the alternative of using ceric oxide instead of rouge as the polishing agent, therefore, in the following series the only change in the standard conditions of polishing was that ceric oxide was used in place of rouge. The composition of the slurry being 50 grams of ceric oxide per 1000 mls of water.

<u>Sample No.</u>	<u>Time of Polish</u>	<u>No. of tracks per cm²</u>
1E	5 secs	133
2E	15 "	138
3E	50 "	135
4E	120 "	130
5E	240 "	129

Again there was no evidence of any flow taking place.

In the final set of polishings both types of glass were polished with ceric oxide and rouge. The procedure of polishing was altered in that force was applied by hand and not by means of

the standard weight. The method employed was that of polishing for a period of twenty minutes using a slurry composed of 50 grams of the agent in 1000 mls of water, the rate of flow of slurry during this wet polish being 40 mls per minute. At the end of 20 minutes the table was allowed to run dry and there were no further additions of the polishing compound. This operation of the dry polish was continued for a period of 10 minutes, following this an autoradiograph with a 100 hour exposure was taken of the resulting polished surface.

SERIES F

Lead Glass

<u>Sample No.</u>	<u>Type of Agent</u>	<u>No. of alpha tracks/cm²</u>
1F	Ceric oxide	133
2F	Rouge	131

Phosphate Glass

3F	Ceric oxide	129
4F	Rouge	129

There was no indication of flow even when the table was allowed to run dry. Dry polishing was continued for a period of 10 minutes after 20 minutes of wet polishing.

Counts were again made as close as possible to the boundary in a similar manner to that done in series "A" and "B". The outermost point of the boundary being 0.218 mms from the edge of

the field and again a single sweep taken of the autoradiograph.

<u>Sample No.</u>	<u>Area Counted</u>	<u>No. of alpha tracks</u>
1F	0.033 cm ²	5
2F	0.021 "	2
3F	0.045 "	7
Total area counted		0.099 cm ²
Total number of alpha tracks		14 ± 3.7
Therefore number of alpha tracks/cm ²		141 ± 37.4

Again there appears to be no evidence of a flow, however, the probable error is great because of the limited number of plates. Therefore these results are not conclusive in themselves and serve only as an indication.

The following procedure was carried out when the glasses were polished with radioactive ceric oxide.

- (1) 2 mcs of irradiated ceric oxide were mixed with non-irradiated ceric oxide to give 10 grams of the mixture. This was then slurried in 200 mls of water.
- (2) Approximately 5 mls of this slurry was rubbed into the felt and polishing carried on for 15 seconds. At the end of this time the table was stopped and a further 5 mls of the slurry added, this

procedure being repeated every 15 seconds.

Pressure was applied by hand.

Picture (2) represents this technique. However, the slurry in the beaker was poured through a funnel in the lucite dome.

When polished, the samples were thoroughly washed and scrubbed with a nylon brush in running water, alcohol, a soap solution and finally more water.

Below is an example of the procedure followed in recording the counting rate of all samples. A standard sample was first counted to determine whether the plateau of the tube had changed. Following this the background was taken and finally the sample counted.

Example

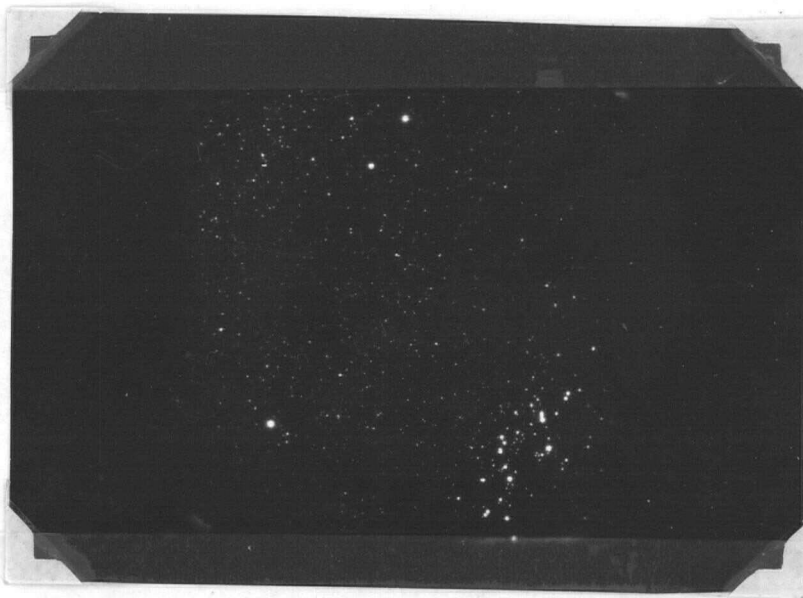
In one determination of the background, 294 counts were recorded in 5 minutes and the sample recorded 406 in the same period of time.

Therefore background rate is

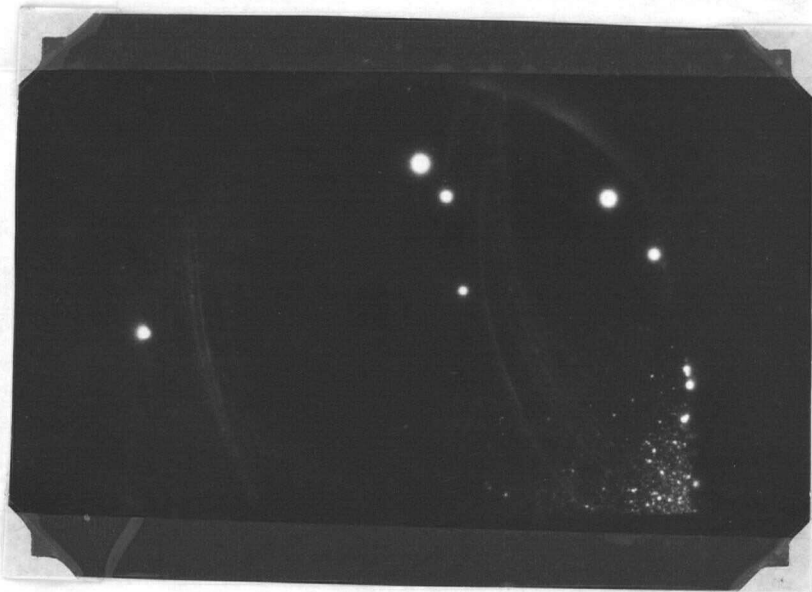
$$\begin{aligned}\frac{294 \pm \sqrt{294}}{5} &= \frac{294 \pm 17.0}{5} \\ &= 59 \pm 3.4 \text{ cts/min}\end{aligned}$$

Total rate of sample is

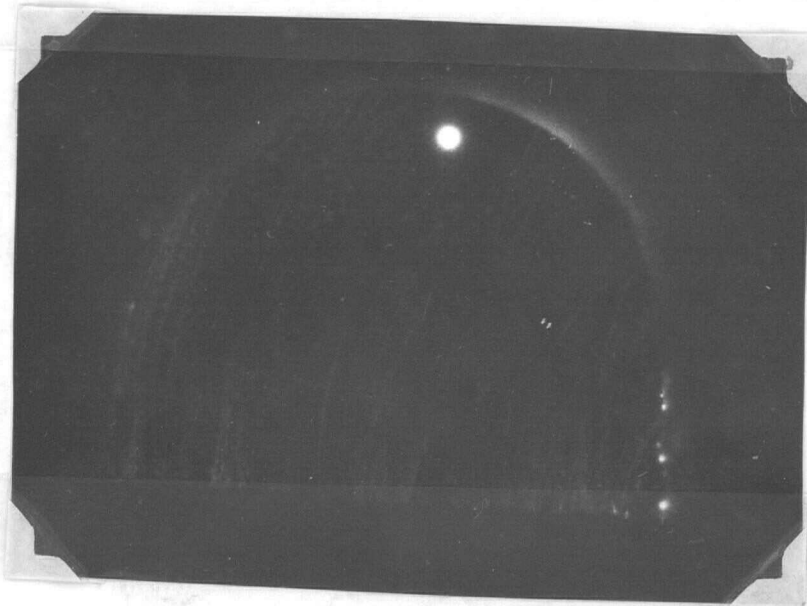
$$\begin{aligned}\frac{406 \pm \sqrt{406}}{5} &= \frac{406 \pm 20.1}{5} \\ &= 81 \pm 4.02 \text{ cts/min}\end{aligned}$$



Autoradiograph I



Autoradiograph II



Autoradiograph III

The sample was then placed in a solution of 6 N HCl and KI for twenty seconds at 70 °C. This solution readily dissolves CeO₂. The count taken of the sample before and after being immersed in this solution is given below.

Count before being placed in the acid solution	59 ± 6 cts/min
" after " " " " " "	13 ± 5 " "

An autoradiograph with an exposure of 72 hours failed to show any activity on the polished surface. Only a faint blackening of the film was observed from activity lodged in the roughly ground sides of the sample.

Sample No. 2 Phosphate Glass Type "C" 6.4 cm²

This sample was polished and treated in the usual manner, total polishing time being 8 minutes.

Net rate	22 ± 5 cts/min
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An autoradiograph taken of this surface gave only a few scattered spots similar to radiograph No.1, but they were less dense. This lack of density made it impossible for a print to be made of this film. Again the spots corresponded exactly with minute pits in the polished surface of the glass.

Sample No. 3 Soda-lead-silicate Glass 3.0 cm²

This sample was polished 6 minutes and treated in the

usual manner.

Net rate 384 ± 9 cts/min

The resulting autoradiograph (2) taken with an exposure of 24 hours again showed the activity to be accumulated in spots on the surface which corresponded with minute pits in the polished surface of the glass. This sample was polished for a further 15 minutes, giving a total polishing time of 21 minutes.

Net rate 242 ± 9 cts/min

Autoradiograph (3) had an exposure of 44 hours. Again it is seen that the activity was accumulated in spots. Scratches visible to the naked eye appeared in this sample when the polishing felt was changed five minutes before the end of the polishing. They were attributed to particles on the new felt. In the resulting autoradiograph (3) darkening appeared on the film directly above these scratches, indicating the presence of CeO_2 . To improve the polish of the sample, polishing was carried on for an additional 21 minutes.

Net rate 12 ± 2 cts/min

The autoradiograph of this sample showed no darkening whatsoever.

DISCUSSION

The method used to investigate the problem of determining whether glass flows during polishing was primarily one of being able to detect any radioactive glass flowed over to a non radioactive glass. As previously shown the photographic method used enabled the detection of a layer 4.5 \AA in thickness. However, if during the polishing operation particles of glass are removed and later re-attached these particles would leave clumps of alpha tracks on the resulting autoradiograph. For example, if 6 alpha tracks were left in a clump this would represent :-

1.98×10^{-8} grams of U_3O_8 give 148 alpha tracks/cm² on an autoradiograph with a 200 hour exposure.

Therefore
$$\frac{6}{148} \times 1.98 \times 10^{-8} = 8.04 \times 10^{-10} \text{ grams of } \text{U}_3\text{O}_8 \text{ are required to give such a clump.}$$

Composition $4.8\% \text{ U}_3\text{O}_8$

Therefore
$$\frac{100}{4.8} \times 8.04 \times 10^{-10} = 1.67 \times 10^{-8} \text{ grams of uranium glass will give such a clump.}$$

Density 2.5 gms/cc

Therefore volume occupied by this weight of glass is

$$\frac{1.67 \times 10^{-8}}{2.5}$$

$$6.7 \times 10^{-9} \text{ ccs}$$

Hence a clump of 6 alpha tracks observed in the resulting autoradiograph would have represented a chip of 6.7×10^{-9} ccs of uranium glass, had this chip been removed from the active side and later incorporated on the inactive side. Assuming such a chip to be spherical the diameter would be 0.019 mms.

Such clumps were noticed in a few radiographs. However, upon re-washing and re-scrubbing these particular samples the clumps were not reproduced in the ensuing autoradiographs. Because the particles were so easily removed it was concluded that they were merely trapped in the surface and not due to any flow.

If during polishing a flow did take place it was reasoned that the sharp boundary would have disappeared leaving instead a gradient. In all the autoradiographs taken there was no evidence whatsoever of this phenomenon, the boundary remained sharp and distinct.

From the results of the experiments conducted using radioactive glass, it was concluded that the most conspicuous feature during polishing was that of removal of glass, rather than that of a smoothing or flowing action resulting from the softening point of the glass being reached. This statement is modified to include the type of glass polished and the conditions of polishing. It may also be pointed out that even when the table was allowed to run dry there was still no evidence of any flow taking place. Thus if flow occurs, it is over a distance of less than 0.22 mms even when the polishing is done with a dry felt under $.3 \text{ kg/cm}^2$ for 10 minutes.

The results of the experiments conducted with activated CeO_2 indicate that some of this polishing agent remains on the polished surface in spots after polishing is ceased.

If ceric oxide is entrapped in the crevices of the surface this would account for the spottiness of the resulting autoradiographs. It would also account for the decrease in activity with longer polishing times for with prolonged polishing a finer polish is naturally attained, thus fewer crevices are available to entrap the ceric oxide. The pits and lines in the surface corresponded exactly with the dark areas on the autoradiographs, as could be seen with the naked eye. Further evidence which suggests that the CeO_2 is merely entrapped in the surface was

gained from the experiments in which the ceric oxide was so readily dissolved by the acid solution. If the CeO_2 were fused with the glass it would be expected that the rate of attack would have been much slower. The acid solution will attack the glass, therefore this evidence is not at all conclusive and is presented as a suggestion.

If fusion of the CeO_2 with the surface glass took place it was reasoned that there would probably have been an increase in the amount of the agent incorporated with the glass as the polishing time was prolonged. However, there was no increase in the amount of ceric oxide on the surface with prolonged polishing times but rather a decrease. This mechanism, therefore, does not appear to be the one involved.

To account for the impossibility of removing CeO_2 from the surface by washing it was possible to reason that during polishing crevices were flowed over, thus securely entrapping this polishing agent. If this were the case it was expected that a considerable amount of activity would have been present on the surface after prolonged polishing. Such was not the case, hence it was again reasoned that the most conspicuous feature of the polishing operation was that of glass removal.

FUTURE WORK

- (1) Use radioactive rouge to determine whether this agent fuses with the glass during the polishing operation.
- (2) Further work might be carried out on the study of the flowing over of scratches. A possible method of attacking this problem would be that of scratching a well polished surface then rubbing radioactive ceric oxide or rouge into the scratches. After this treatment the glass could be polished in an attempt to flow over these scratches. A failure to remove any of the radioactive material by washing would indicate that the scratches had been flowed over. By using a glass that is not readily attacked by a solution which would dissolve the radioactive agent it would be possible to determine the depth of the glass flowed over. The rate of attack of this solution on the glass would have to be determined.
- (3) The work done on phosphate glasses might be checked to determine whether the fractures interfered with a flow. A study of the coefficients of expansion of this glass would first be necessary in order that a good join be made. When a join is made in a flame the boundary is more distinct than if it is made in a furnace.