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PREPARATION OF A MOCK FISSION NEUTRON SOURCE

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Abstract

A procedure is described by which a neutron source with a fission spectrum (mock-fission) was prepared. 26.6 curies of Po was used as the source and the target material was a mixture of NaBF_4 and $2\text{NaF}_2 \cdot \text{BeF}_2$ (mol ratio B/Be $\approx 96/4$). A gas stream distillation technique was used for the preparation, and the resulting Po-target material mixture was pressed into a hard pill which was contained in a cylindrical capsule, 0.317 inches in diameter and 0.25 inches long. When assembled the source fitted inside a sphere 1.03 cm in diameter. The neutron yield was 3.85×10^6 n/sec or 1.46×10^5 n/curie sec.

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PREPARATION OF A MOCK FISSION NEUTRON SOURCE

Summary

A procedure is described by which a neutron source with a fission spectrum (mock-fission) was prepared. 26.6 curies of Po was used as the α source and the target material was a mixture of NaBF_4 and $2\text{NaF} \cdot \text{BeF}_2$ (mol ratio B/Be = 96/4). A gas stream distillation technique was used for the preparation, and the resulting Po-target material mixture was pressed into a hard mill which was contained in a cylindrical capsule, 0.317 inches in diameter and 0.25 inches long. When assembled the source fitted inside a sphere 1.03 cm in diameter. The neutron yield was 3.85×10^6 n/sec or 1.46×10^5 n/curie sec.

Introduction

Richards (1) reported that the neutrons formed by Po α 's with a BF_3 target possessed an energy spectrum similar to the spectrum of neutrons formed by fission of 25 and 49. It was pointed out by him that compact mock fission sources could be prepared by mixing Po and sodium fluoborate crystals. Two such sources were prepared for use by the Research Physics Division. The spectrum of these sources, designated by Richards as Source #1 and #2, have been described in a previous report (2). Source #1 contained 2 curies of Po and was prepared by evaporating a dilute HCl-Po solution to a few microliters and adding crystals of the salt. This method did not appear practicable for use with stronger sources. Therefore, a gas stream distillation technique was used for the preparation of Source #2. The target material for this source was NaBF_4 with 2 mol % of BeF_2 , and it contained 8 curies of Po. It was indicated

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by Richards (2) that the mixing of the Po and the target material was rather poor.

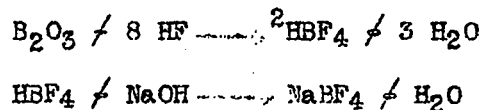
Accordingly, for Source #3, which was requested, although a gas stream distillation technique was used for adding the Po to the target, a separate mixing and pressing operation was performed to achieve a more uniform and stable mixture. The procedure was tested in preparing a number of weak sources containing 100 - 500 millicuries, before source #3, specified to contain 25 curies, was made. To increase the number of high energy neutrons the mol ratio of B/Be was increased to 96/4. This report describes the procedure employed in the preparation of Source #3.

- (1) Richards, H. T. - LA-85
- (2) Richards, H. T. - LA-201

Materials

The target material was a mixture of sodium fluoborate, NaBF_4 , and sodium fluoberyllate $2\text{Na F} \cdot \text{BeF}_2$ with a mol ratio of B/Be \approx 96/4. These materials were prepared in this laboratory.

The NaBF_4 was prepared by E. S. Robinson using a modified procedure described by Booth, H.S., "Inorganic Synthesis", McGraw-Hill Co., New York (1939). The reactions employed were:



91.9 g of 48% HF was weighed out in a Pt dish. The dish was placed in an ice bath and reagent grade B_2O_3 was added to the solution slowly. The dish was left in the ice bath 1.5 hours and then remained over night at room temperature.

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The solution was neutralized with 91 ml of 5 N NaOH. 95% ethyl alcohol was added to precipitate the salt, and finally the crystals were filtered out on a sintered glass funnel. The crystals were transferred to a platinum dish and dried for four days in a vacuum desiccator over H₂SO₄. The final product was in the form of very fine crystals.

The sodium fluoberyllate was prepared by the following reaction:



1.099 g of NaHF₂ was weighed out in a Pt dish. 60 ml of water and 3.1 ml of ca 5 N NaOH was added to this salt. It was necessary to warm the dish over a steam bath to effect complete solution. The resulting pH was about 4 as indicated by universal test paper. The BeF₂ available was a sticky glass-like solid of questionable purity. It was ground in a mullite mortar and 0.85 g weighed into a platinum dish. The NaF₂ solution prepared previously was added to these crystals. The BeF₂ dissolved only slowly while the dish was heated on a steam bath. As the BeF₂ dissolved light crystals formed which were presumably the fluoberyllate. When it appeared that all the BeF₂ had dissolved, the solution was evaporated to dryness under an infra-red lamp. The rather large plate-like crystals remaining were ground in a mullite mortar, they had none of the characteristics of BeF₂ and a different appearance from NaF₂ formed by evaporation.

To prepare the target mixture with a mol ratio of Be/B = 4/96, 1.00 g of the 2NaF·BeF₂ and 20.12 g of the NaBF₄, prepared by the above methods, were weighed out into a mullite mortar. The mixture was ground and mixed by hand with a platinum spatula for 40 minutes.

The Po used for preparing the source was deposited on both sides of 2 mill Pt strips, 0.5 cm wide and 2 cm long. The deposit densities varied from

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1.0 to 3.0 curies/cm². Pt foils supplied by the Monsanto Chemical Co. Laboratory were used for this source. They were removed from the shipping containers just before insertion into the distillation apparatus.

Apparatus

The Po was added to the target mixture by a gas stream distillation. The distillation was performed in an apparatus sketched in Figure 1. It consisted of the quartz tube shown which was supported in a vertical position. This quartz tube had a constriction 3 mm I.D. for producing a narrow jet of gas. The Po foils were inserted after removing the standard taper cap, which also contained a thermocouple well. A platinum wound resistance furnace was fitted closely around the quartz distillation tube for heating the region containing the Po foils. It was important that this furnace also heat the constricted portion of the tube to prevent the condensation of Po on the quartz surface in this region. In operation the furnace rested on the horizontal surface of the quartz tube just below the constriction.

A condenser containing the target material was located in the wide portion of the quartz distillation tube below the constriction. To load the target material, a brass plug was first slipped into the top of the condenser. A 100 mesh platinum gauze was laid on top of the brass plug, and then a Pt sleeve slipped into the condenser. The crystals of target material were loaded inside this sleeve by a Pt microspatula and were supported by the Pt gauze. The condenser was then inserted into the bottom of the distillation tube and held in place by the rubber stopper. In this way the bed of crystals was supported directly in the path of the gas jet. Steam was used as the cooling agent for the condenser. For a distillation, after the Po foils had

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been loaded into the tube, He was passed into the cap at the top of the tube. The furnace was then heated so the He passed down over the hot foils, through the jet, down through the bed of crystals and finally escaped through the center tube in the bottom of the condenser while the Po condensed on the surface of the target material. A glass tube containing a small piece of zinc sulphide fluorescent screen was placed just below the condenser to detect the presence of any Po that might penetrate the bed.

It was found that the fine crystal powder could be readily jarred or blown by gas currents out of the bed. However, if a drop or two of ethyl alcohol was added to the crystals after they were loaded, the particles were cemented together, but the bed still remained porous to the flow of gas. Preliminary experiments also indicated that a considerable fraction of the Po condensed on the inside surface of the Pt sleeve in the condenser. Consequently, the following procedure was developed. A slurry of the $\text{NaBF}_4 \cdot 2\text{NaF} \cdot \text{BeF}_2$ mixture was made by adding a few drops of water to some crystals. This slurry was then painted on the inner surface of the Pt sleeve with a camel's-hair brush. When the water was evaporated by heating in a drying oven, a reasonably adherent crust remained. The weight of the crust, obtained by difference, amounted to about 0.10 - 0.15 g.

Since the large amount of Po present in the finely divided source mixture was felt to constitute a serious health hazard, all operations performed with the unconfined crystals to which Po had been added were carried out in a small wooden dry box. A pyrex tray was placed on the floor of the dry box so any material that was spilled could be readily recovered. Thin latex obstetrician gloves were used in the dry box to facilitate the handling of the small objects.

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To package the source in the smallest possible volume it was necessary to press the materials into a compact pill. The crystals containing the polonium were therefore placed in the brass capsule, 1 inch long, whose details are shown in Figure 2. The brass capsule could be placed in the split die, details in Figure 3, which was assembled by means of four cap screws. The plunger, Figure 4, could then be inserted into the hole in the die. A 20 ton hydraulic press was available for pressing the plunger down against the powder. The shoulder on the plunger meeting the top surface of the die limited the compression to the desired pill size. Very close tolerances were necessary for the matching parts of the brass cup, the die, and the plunger; and the surface of the plunger and the cavity of die were of the highest possible finish. A special tube cutter, Figure 5, was built for cutting off the cartridge flush with the top of the pressed pill. The cartridge with the plunger protruding was laid in the V block of the cutter, the base of the cartridge and plunger were then rotated under the cutting wheel and pressure applied by the thumb screw. After the upper section was removed, the cap, Figure 2, could be slipped over the top of the cartridge. It was found that 450 mg of crystals could be pressed into a compact pill with a hard, smooth surface by forces of less than 1,000 lbs. If somewhat more material was used so that about 5,000 lbs. was required, the brass capsule was seriously distorted. A small portion of the top surface usually cracked away from the pill during the cutting operation. These fragments could be placed inside the cap before it was slipped over the cartridge. The special tube cutter that was used for this operation was made in the laboratory shop with very small clearances so the cartridge could be cut closely to the desired length.

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Procedure

Three separate distillations were employed to add the Po to the target crystals. A crust of the salts weighing 140 mg was placed on the inner surface of the Pt sleeve inserted into the condenser. About 100 mg of powder was placed inside the sleeve and a few drops of alcohol added to cement the particles together. The condenser was inserted into the distillation tube. Three foils bearing a total of 6.94 curies of Po were dropped into the distillation tube. A helium flow of 45-50 ml/min at room temperature and 590 mm Hg (one Los Alamos atmosphere) was started, and the power turned on for the furnace. After an interval of 19 minutes when the chromel-alumel thermocouple in the well indicated a temperature of 850°C, the fluorescent screen in the effluent line was observed to glow brightly in the dark. The heating was immediately stopped and the apparatus allowed to cool. It was found that a hole had formed in the bed of salt, which had permitted the Po to pass through without condensing. Only a fraction of the Po had distilled at this time. Therefore, another 75 mg of crystals were added to the bed and wet with alcohol. Also a new glass tube with fluorescent screen was inserted in the effluent line. The apparatus was reassembled and the distillation continued again with a 45-50 ml/min He flow. The distillation tube was maintained at a temperature of 1030°C for a period of two hours. There was no further indication of Po penetrating the condenser. After the apparatus had cooled, 60 mg of crystals were added to the bed, and again a drop or two of alcohol added. The Po foils were replaced by three additional foils bearing 10.18 curies. The distillation was again performed with 45-50 ml/min He flow at 1030°C for a period of two hours. After the addition of 60 mg of crystals, a third distillation under similar conditions was made with 12.94 curies on three foils. Thus a total of 30.06 curies had

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been used in the three distillations. Finally, 30 mg of crystals were added to the bed. There was a heavy black deposit over the topmost crystals in the bed following each distillation and over the surface of the crust on the sleeve. The body of the salt exhibited a strong orange fluorescence. The condenser assembly was transferred to the dry box for the subsequent operations. The bed was cracked out into a small agate mortar. The Pt sleeve, gauze, and brass plug were pushed out into the mortar by means of a rod inserted into the effluent tube of the condenser. The salt was dusted off the gauze and plug with a camel's-hair brush. The crust on the inner surface of the sleeve was readily cracked away into the mortar by means of a small Ni spatula. The material was then ground with an agate pestle and mixed with a Pt spatula. The neutron emission at this time could be detected by a BF₃ proportional counter which was situated 20 feet away. The grinding and mixing operation was observed to significantly increase the neutron emission. The final mixture was a pale gray-lavender color. The strong orange fluorescence could be observed in a shadow from the room lights. This fluorescence of the crystals provided a nice indication of the uniformity of mixing and indeed furnished sufficient illumination for these operations to be performed in the dark. The resulting powder was transferred from the mortar to the brass cartridge rather easily by means of a V-shaped platinum spatula.

The cup was removed from the dry box and placed in the split die which was then assembled. The plunger was inserted into the hole of the die. After the die assembly was placed in the hydraulic press, the plunger was depressed to the stop. It was left under pressure for two hours. The plunger and cap together were removed from the die and placed in the dry box. By means of the special tube cutter, described previously, the upper portion of the brass

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cartridge was cut off. The cap was then slipped over the cartridge. The capsule was removed from the dry box and burnished with steel wool. A layer of solder was applied over the bottom of the source with a small iron and resin for flux to seal the cap to the cartridge. The surface was burnished further with steel wool to reduce any contamination on the outside. It was noted that the source was quite hot because of the large quantity of energy dissipated in stopping the α 's.

It is to be noted that the experience gained in rehearsal preparations with a few hundred millicuries of Po was invaluable in making the strong source.

Recovery of Polonium

Each of the source foils used for the distillation were measured afterwards in an integrating chamber. On none of the foils had more than one or two millicuries remained. Various portions of the distillation and handling apparatus were leached in acid after the preparation was completed. Quartz, agate, and platinum were stripped with c HNO₃, brass and copper with 6 N acetic acid, and neoprene tubing with c HCl. These solutions were then assayed. The greatest amount was recovered from the quartz distillation tube, 2.95 curies. A blue fluorescence of the quartz was observed in the vicinity of the constriction during and following the distillations. 0.41 curie was recovered from the brass condenser. A total of 3.47 curies was stripped from all the parts. Therefore, the source contained at most 26.6 curies.

Results

The final source was a cylinder 0.317 inches in diameter and 0.25 inches long. It is to be noted that the cylinder of maximum volume inscribed in a sphere of given radius has a ratio: diameter/length = 2. The source

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was slightly longer than was planned originally because the cap did not seat against the rim of the cup. The maximum dimension of the source was 1.03 cm.

The polonium content was at most 26.6 curies. The external contamination was measured in an ionization chamber and was found to be $3-5 \times 10^4$ counts per second. This was no larger than frequent hand contamination of the polonium chemists.

The neutron emission was measured by John William's group and found to be 3.85×10^6 neutrons/sec. This corresponds to a neutron yield of 1.45×10^5 n/curie second. A 400 millicurie source prepared by this technique yielded 2.4×10^5 n/curie second so there apparently was considerable loss of efficiency caused by the concentration of Po. When compared with the yield of Source #2, 2.0 curies - 1.3×10^5 neutrons/curie second and Source #3, 8 curies - 0.8×10^5 neutrons/curie second, it appears that the mixing and pressing technique was of considerable value. The neutron yield was reported to decay with the standard Po half-life.

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FIGURE 1

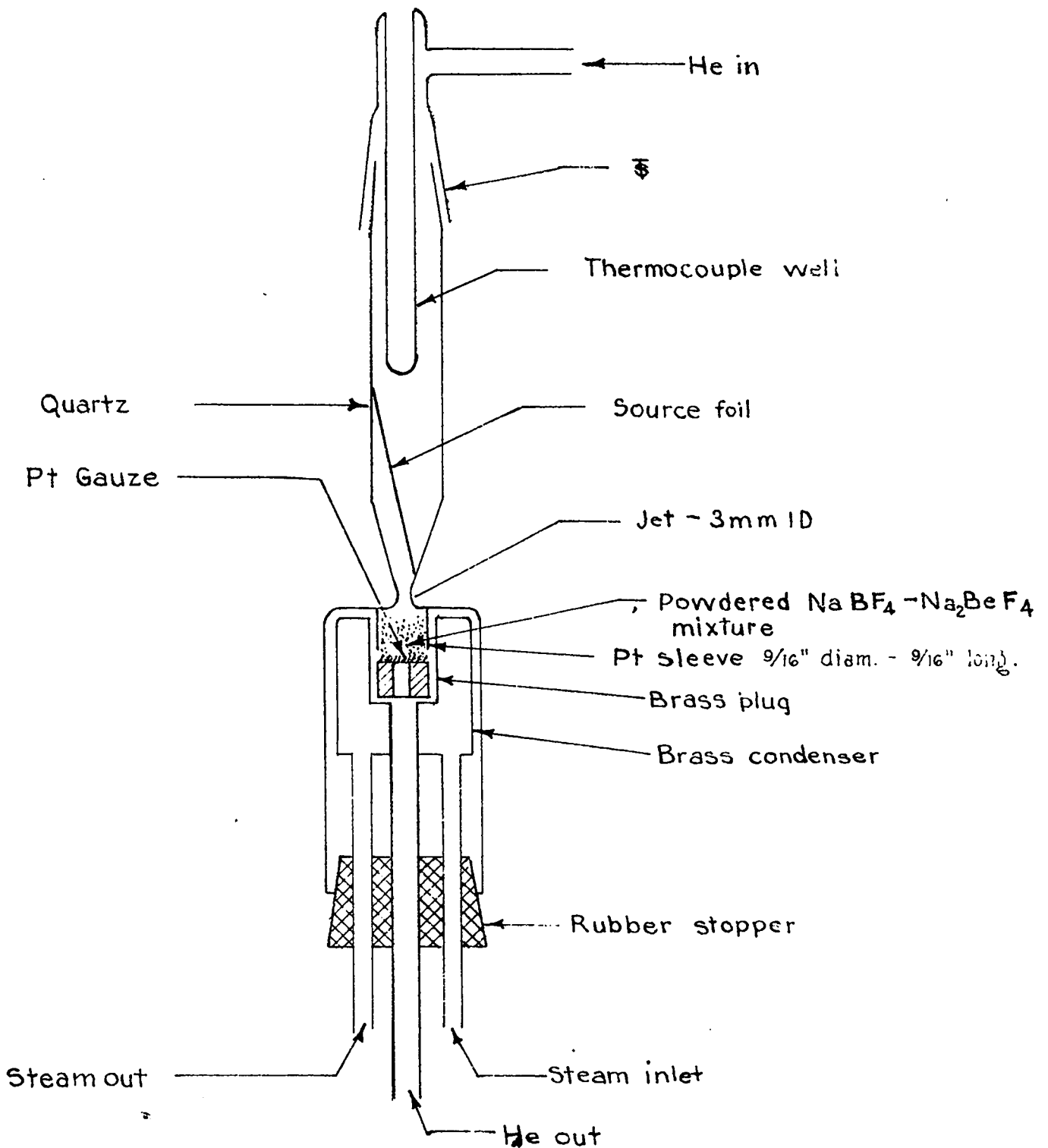
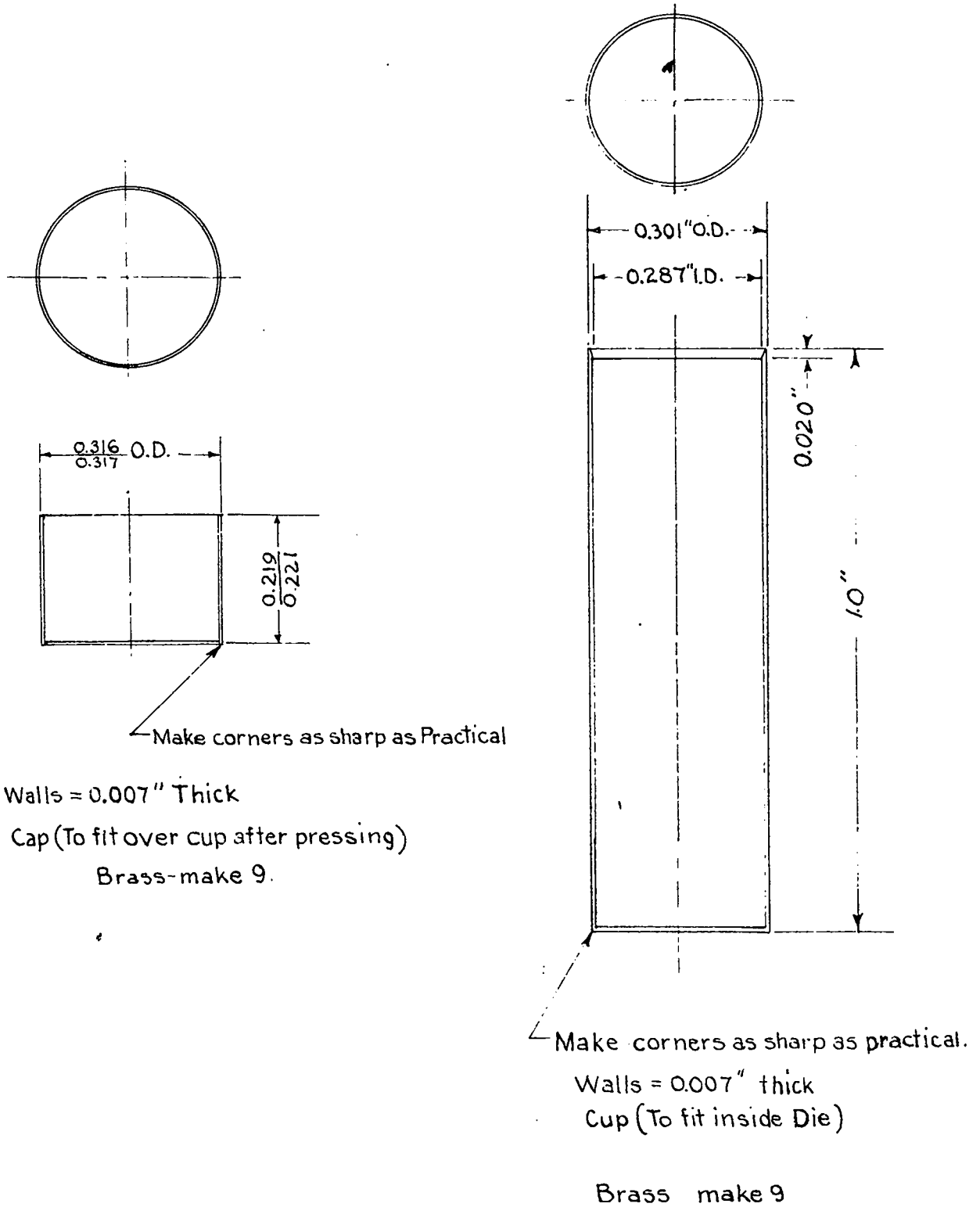
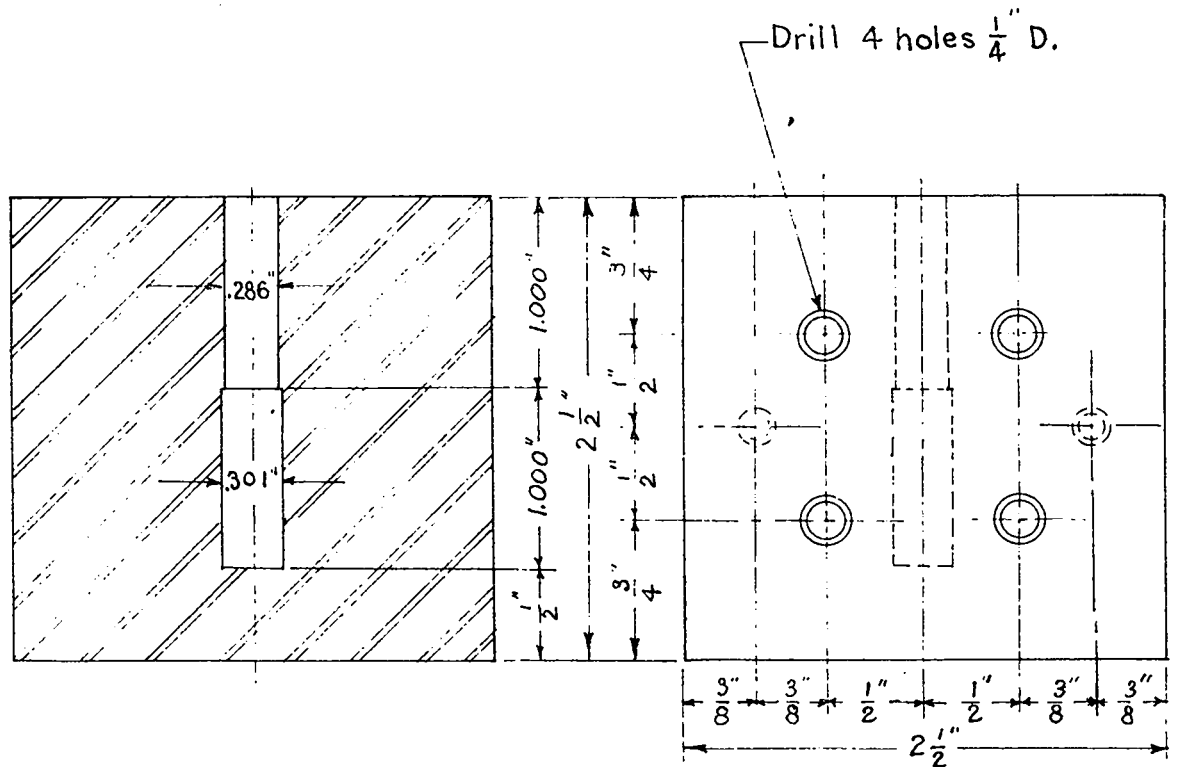
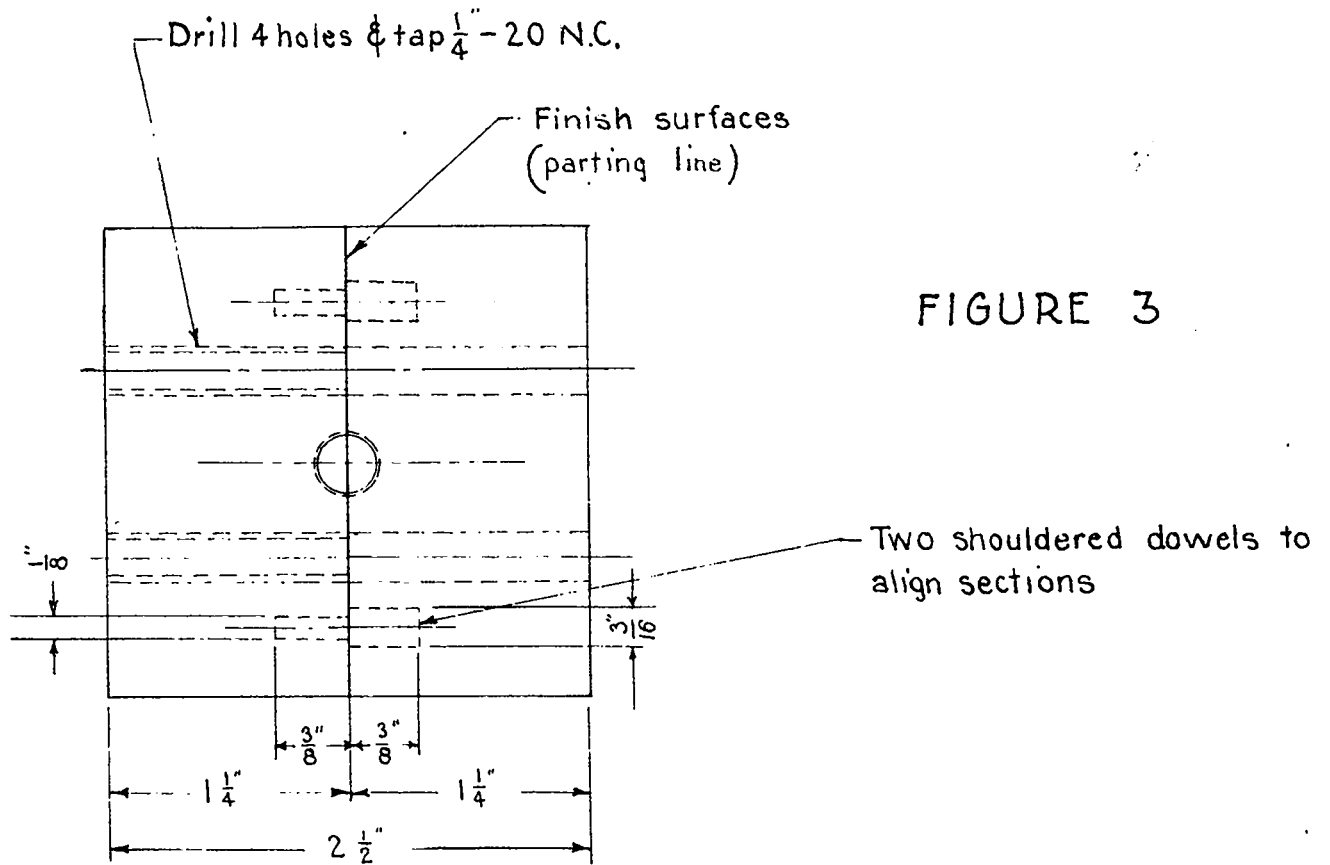


FIGURE 2





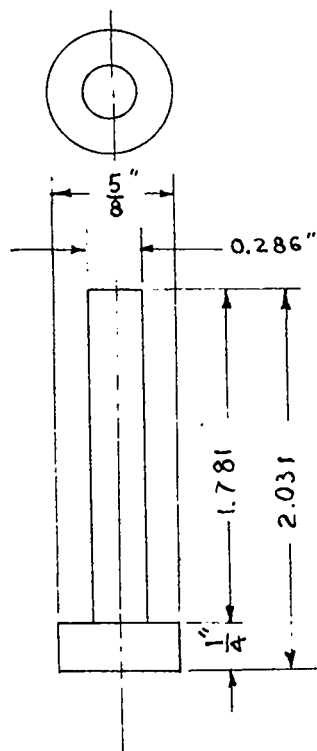
SPLIT DIE ASSEMBLY

Make of K46 steel

One req'd

Assembled with 4 Hex head $\frac{1}{4}$ " 20 N.C. cap screws

FIGURE 4

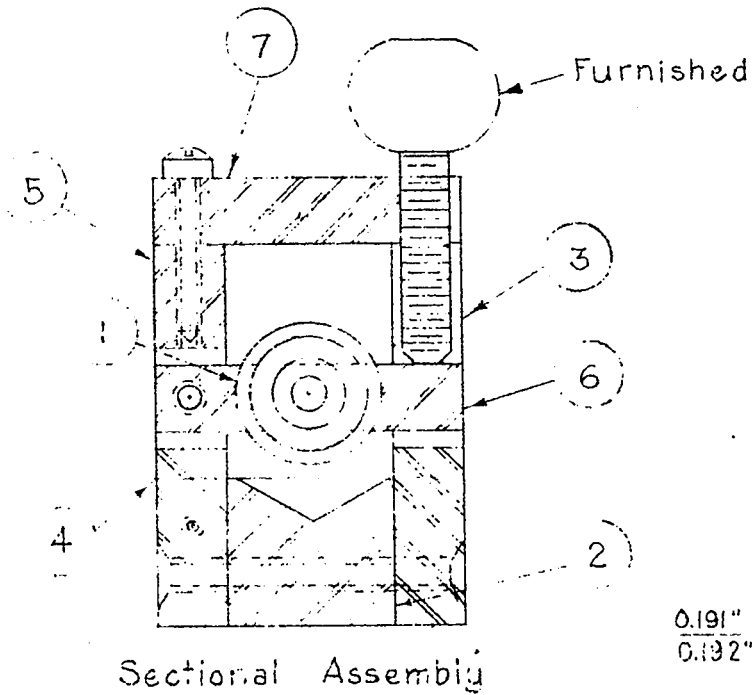


PLUNGER

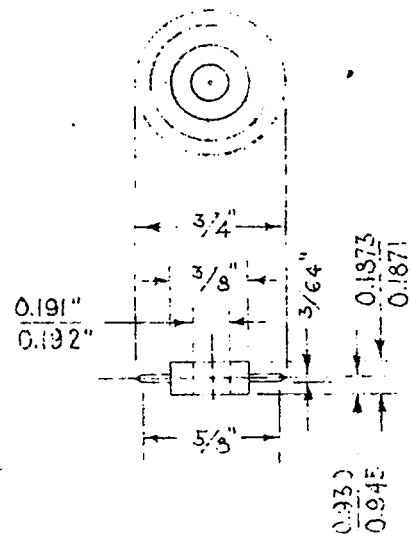
Made from K 46 steel - Harden surface

One Required

FIGURE 5



Sectional Assembly



Part 1
Matl: Tool Steel
Reqd: 1

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