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SPARK SOURCE MASS SPECTROGRAPHIC ANALYSIS
OF PLUTONIUM-URANIUM OXIDES

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The United States is committed to the Liquid Metal Fast Breeder Reactor program. As part of that program, the construction of the Fast Flux Test Facility at Hanford is under way. A knowledge of the composition of the fuel materials going into that reactor is of extreme importance. Spark source mass spectrography which is an extremely powerful and versatile analytical tool should find widespread applicability in the analysis of fuel materials. It is capable of detecting any element in the periodic table to very low concentration levels. We have applied it to the determination of the more difficultly analyzed impurities such as tantalum, tungsten, and the rare earth elements in uranium oxide and in plutonium-uranium mixed oxides. Our instrument is a double focussing mass spectrograph employing Mattauch-Herzog geometry made by Japan Electron Optics Laboratory with photographic plate detection. A description of the instrument and associated equipment will be presented. The methods and procedures used in the analysis of a plutonium-uranium mixed oxide fuel sample will also be described.

Figure 1 shows a general view of the spectrograph with the glove box built around the ion-source end. Figure 2 shows a block diagram of the system which can be broken down into three main sections: ion source, analyzer, and detection. The sample is ionized in the source by being subjected to a high voltage radio frequency (RF) spark. The ions formed are extracted and beamed by an accelerating voltage (25 Kv) and slit system and separated according to their mass to charge (m/e) ratios in the analyzer section. The magnetic analyzer accomplishes this; but in addition, our system has an electrostatic analyzer. This is the Mattauch-Herzog double focussing feature. The electrostatic field is required because the RF spark method of ionization produces a considerable spread in energy, direction, and velocity of the ions. The electrostatic field provides direction and

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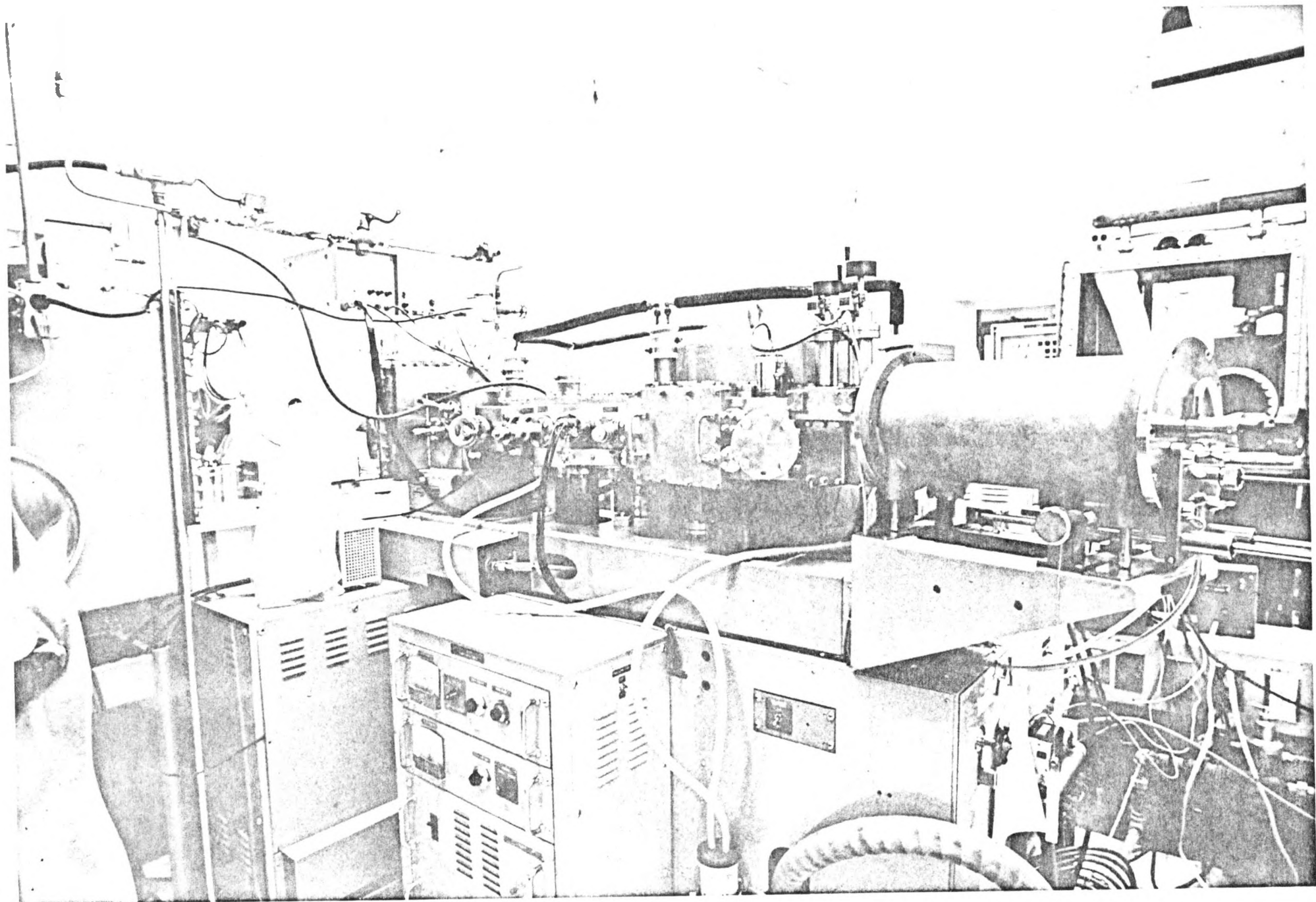
velocity focussing. All these things must take place in a high vacuum. After the analyzer section, the ions emerge focussed in a single plane, the focal plane, and this is where the photographic plate is placed.

In order to ionize Pu-U oxides, the sample must be crushed, pulverized, mixed with a conductor, and pressed into a suitable electrode shape for sparking. We mix our samples with graphite, put the mixture into 3/16-inch holes in a polyethylene plug which is placed in the die shown in Figure 4, and press them at 10 tons. We do this sample preparation in the glove box shown in Figure 3. The electrodes made in this way are inserted into holders shown in Figure 5. A RF spark is applied across the sample electrodes. Figure 6 shows a view taken through the viewing port looking directly into the source. The spark discharge between the two electrodes can be seen.

After the exposure, the plate is developed and interpreted. Varying exposures are made--i.e., varying coulombs of charge as indicated by a monitor electrode, which intercepts half of the ion beam, are collected. The photoplate thus shows lines of decreasing densities (since usual practice is to collect largest charge first and successively decreasing charges thereafter). Since separations are made according to m/e ratios, lines appear at whole mass units and successively weaker lines at fractional mass units. In addition, space charge effects contribute odd lines, molecules (oxides, carbides) and polymers contribute other lines so that the interpretation of the photoplates is not always a simple matter. It is interesting to note that carbon polymers up to C_{19} have been observed.

Concentrations are estimated from the least exposure producing a just visible line. Without too much effort and time a concentration level of 1 ppm is easily detected. By increasing the exposure, fractional parts per million may be detected.

For the analysis of Pu-U oxides, U oxide standards have been used to calculate sensitivity factors (S.F.). For most of the elements, the S.F. is near unity. Figure 7 shows results obtained in the analysis of U_3O_8 and of UO_2 - PuO_2 .



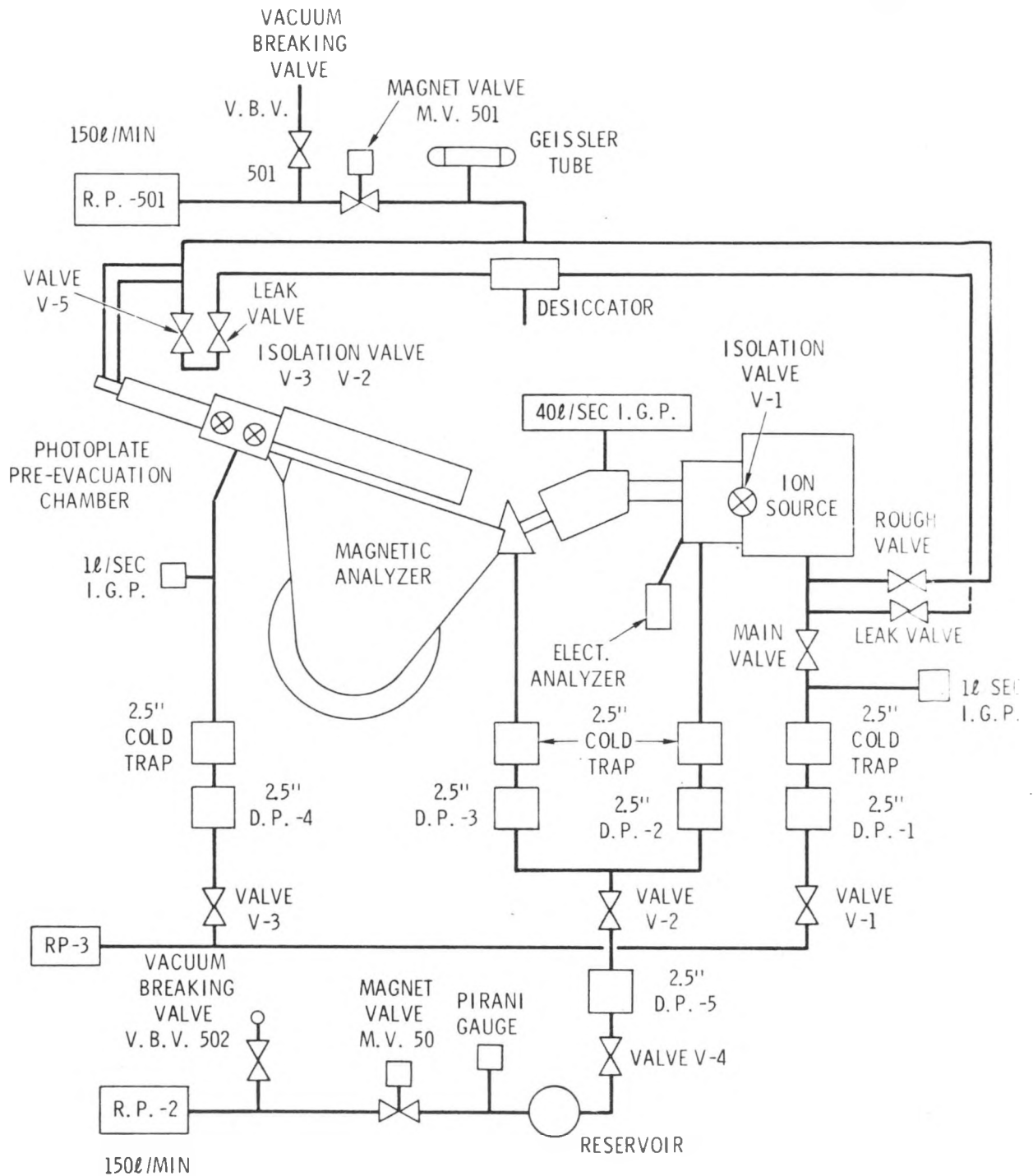
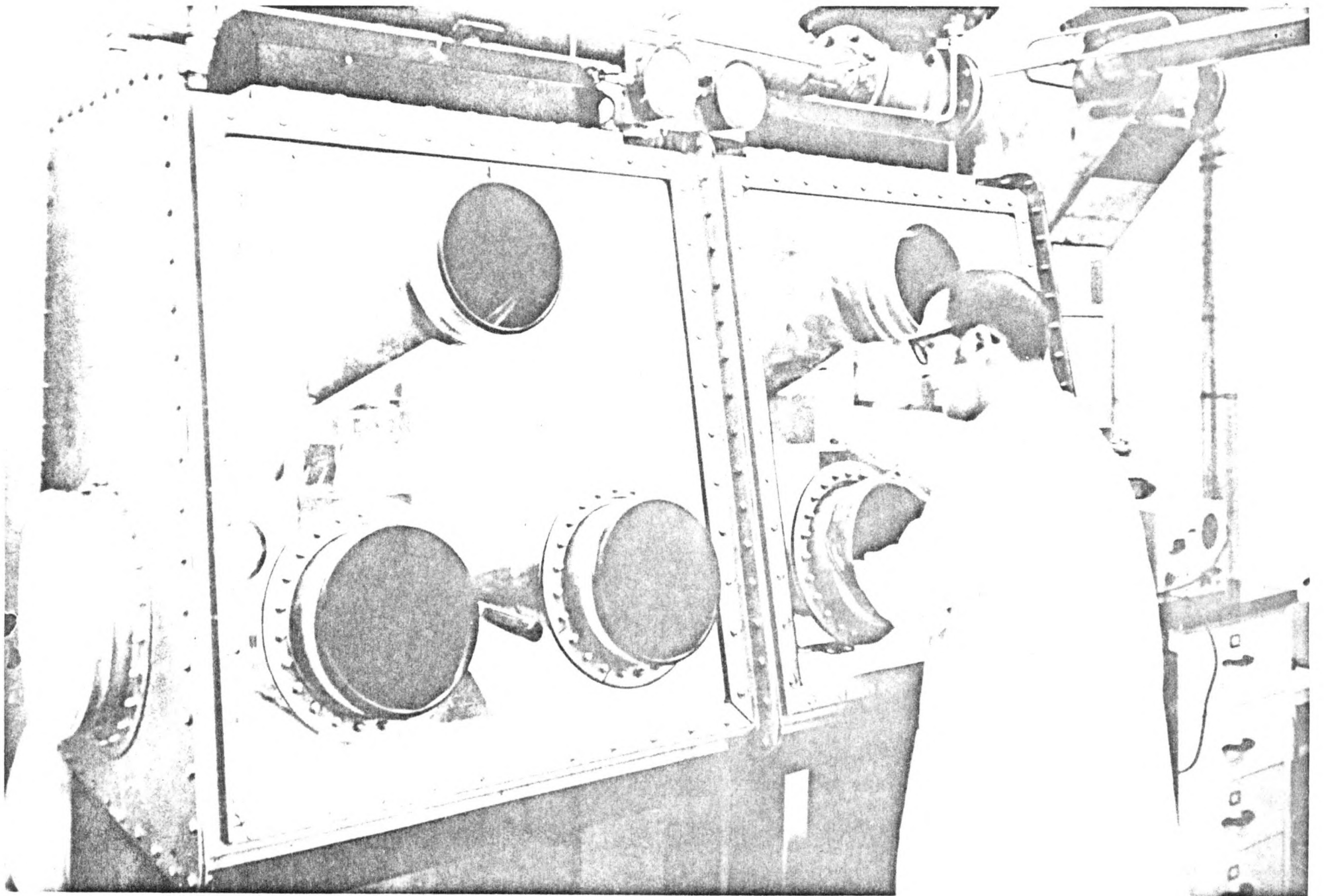
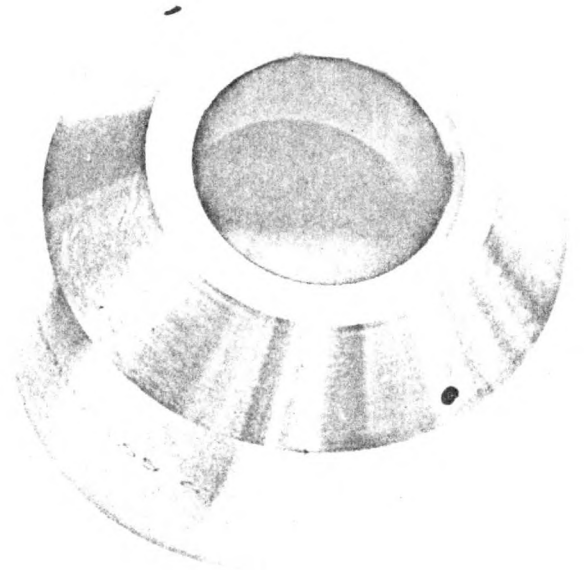
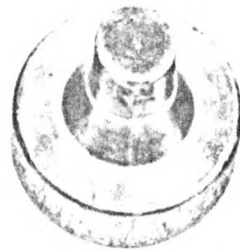
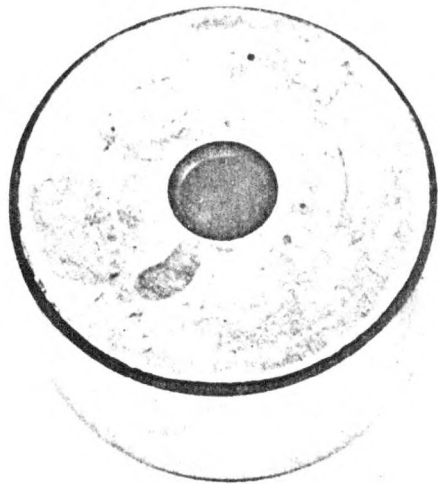
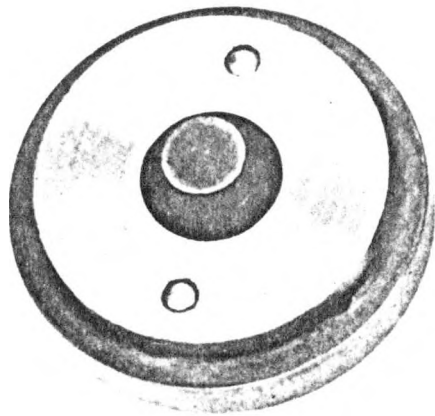
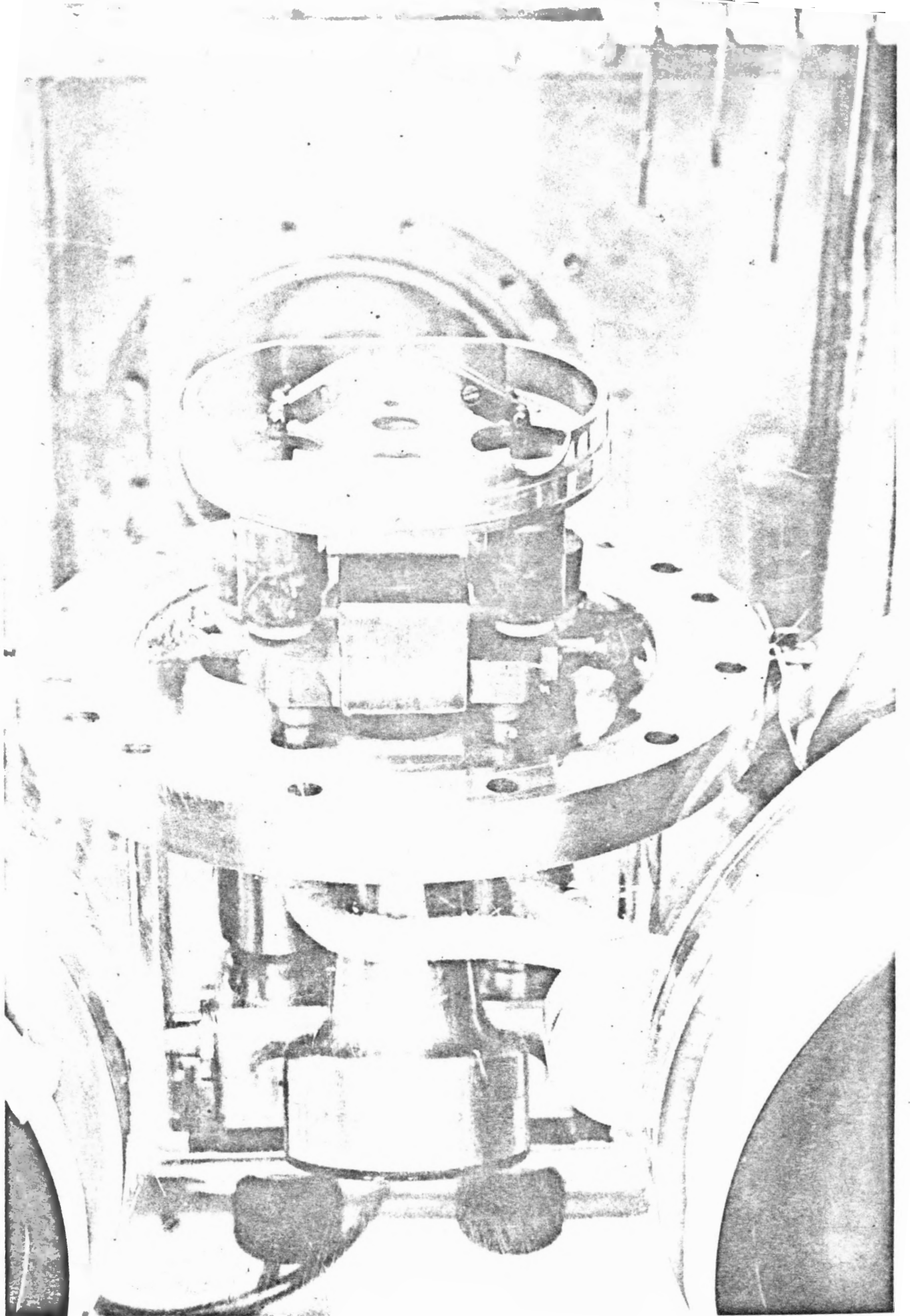
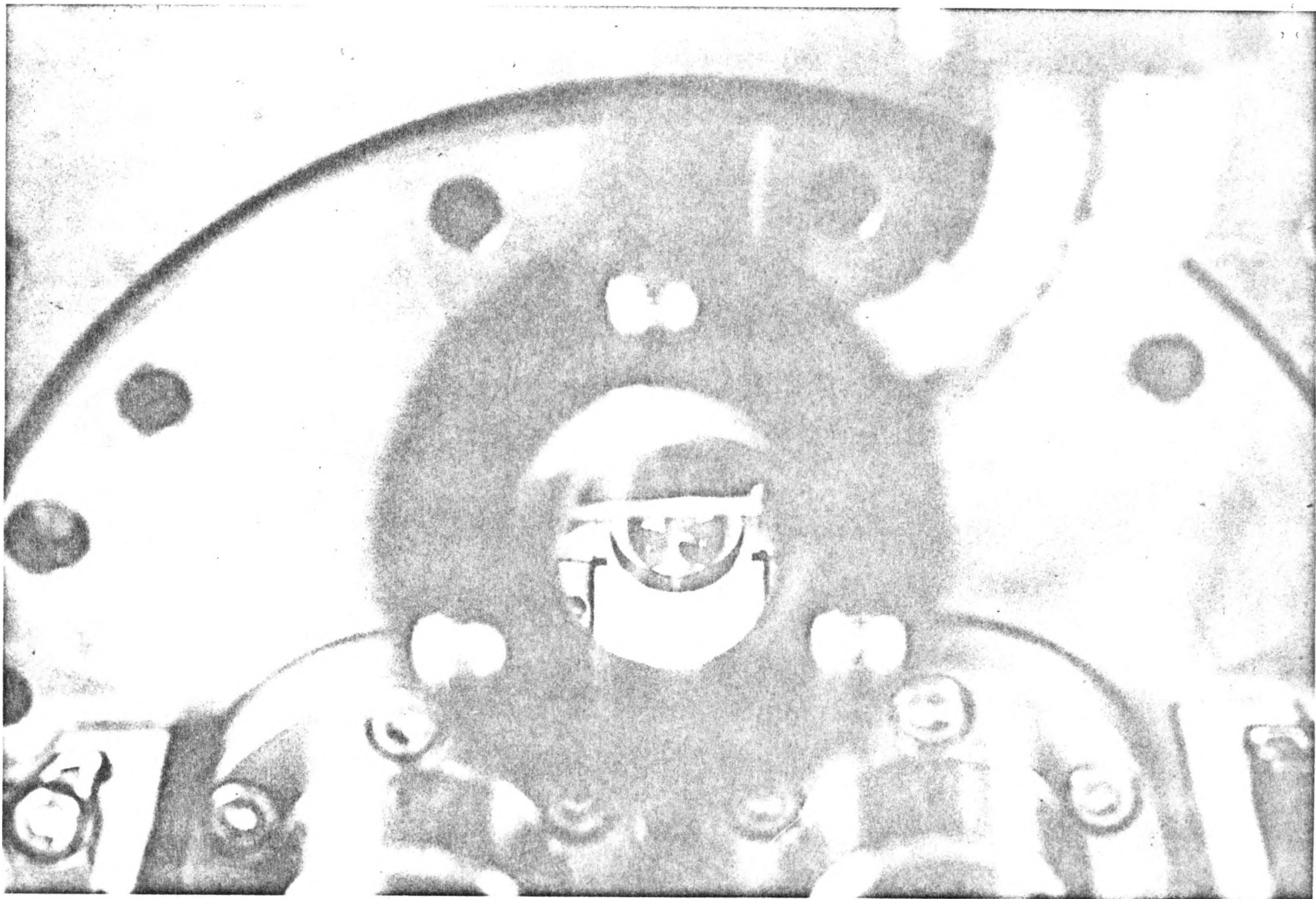


FIG. 1









RESULTS OBTAINED WITH VISUAL ESTIMATION
IN $\mu\text{g/g}$

	<u>Sm</u>	<u>Eu</u>	<u>Gd</u>	<u>Dg</u>	<u>Ta</u>	<u>W</u>
<u>IN U_3O_8</u>						
ADDED	50	50	50	50	200	50
FOUND	30	50	40	80	200	40
<u>IN $\text{UO}_2\text{-PuO}_2$</u>						
ADDED	50	50	50	50	200	80
FOUND	60	50	20	20	200	60

FIG. 7