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Author
Fulrath, Richard M.

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Richard M. Fulrath

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Richard M. Fulrath

Department of Materials Science and Engineering,
College of Engineering, and Inorganic Materials
Research Division, Lawrence Berkeley Laboratory,
University of California
Berkeley, California 94720

Abstract

A commercial hot stage for the JSM-U3 has been modified to give a device capable of long-time operations to temperatures of 1600°C. The design changes center around the construction of a reliable heater, increased radiation shielding, and modification of the thermal electron suppression grid to allow a shorter working distance and hence greater resolution. Magnifications to 5000X at 1600° have been obtained.

Major problems encountered in high temperature operation are surface contamination, temperature measurement, and image recording.

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Richard M. Fulrath
Department of Materials Science and Engineering,
College of Engineering, and Inorganic Materials
Research Division, Lawrence Berkeley Laboratory,
University of California
Berkeley, California 94720

Introduction

In all areas of microscopy (optical, scanning electron, or transmission electron) there exists the desire to examine specimens at other than ambient temperature. Metals and ceramic materials are subjected to high temperatures in processing, and many of the processing steps involve material transport. This is especially true in sintering compacted metallic or ceramic powders to form products. During sintering, mass transport occurs to eliminate porosity in the powder compact and significant geometric changes occur. The various theories of sintering describe the geometric changes in pore morphology and shrinkage of the compact. Direct observation of neck growth between particles and linear shrinkage has been made at temperature by optical microscopy.[1] However, the limited depth of field and magnification obtainable in optical microscopy have discouraged its use. The scanning electron microscope with its inherent advantages over optical microscopy is an obvious choice for an instrument to observe mass transport at high temperatures in basic sintering studies. During the fall of 1970 and throughout 1971 modifications of a commercial specimen heating device were made to achieve a temperature capability of 1600°C at a useful magnification of 5000X and to allow the continuous observation of specimens for periods of hours at temperature.

The primary goal of this development work was directed to sintering studies of metallic and ceramic powders. Individual particle shape and pore morphology changes during sintering of compacted powders were to be observed at temperature.

Original Hot Stage Design

The specimen heating device (SMU3-SHDH) for the JSM-U3 supplied by JEOL was a modification of a hot stage originally developed for the electron probe X-ray microanalyzer. The SEM heating device consisted of a main stage inserted into the front part of the microscope replacing the normal stage assembly. This stage provided an air-lock chamber for specimen exchange with an observation window. The air-lock chamber was also equipped with suitable electrical connections so that the specimen stage could be heated and the temperature recorded for either visual observation or outgassing. The stage allowed the specimen stage (when inserted into the main column) to have translation in the X and Y direction of 10 mm and vertical movement of approximately 3.2 mm. No rotation or tilt motion was provided. Because the stage was developed for secondary electron, back scatter electron, X-ray, and absorbed electron analysis, the working distance was fixed at 32 mm. This large working distance decreased the maximum magnification available, but did allow the mounting of a thermal electron suppression grid between the specimen stage and the secondary electron detector.

When materials are heated to high temperatures in vacuum, electrons may be emitted from the surface and collected on an electrode that is at a positive potential relative to the heated material. The secondary electron detector in the SEM can, therefore, act as a collector for thermally emitted electrons from heated metals or ceramics. Because the surface will emit these low energy electrons more or less uniformly from the entire surface, their intensity can easily exceed the electron beam produced secondaries and completely mask the secondary electron image. A negative potential on a grid between the heated sample and the secondary electron detector can repel these low energy thermal electrons and allow higher energy secondaries to be collected and form the secondary image.

The specimen stage consisted of an alumina or beryllia porcelain crucible surrounded by a non-inductively wound tungsten wire heater. An outer alumina porcelain insulator protected the tungsten heater from a nickel positioning cylinder. Tantalum foil radiation shields around the sides and bottom of the heating assembly reduced radiation heat losses. A flat metal top cover plate with a hold-down clip positioned a thermocouple which contacted the specimen and provided some radiation shielding in the vertical direction. In the design it was expected that the specimen surface would be nearly flush with the top cover plate so that X-ray analysis could be made using a 30° take-off angle.

Initial work with this specimen stage gave satisfactory results at temperatures below 900°C. Above 900°C short heater life was encountered. Also, the heater could not be repeatedly thermal cycled without breakdown. The major difficulty was that the insulating coating applied to the tungsten heating element broke down and allowed adjacent turns of the non-inductively wound heater to short out.

Modified Hot Stage Design

Initial Considerations

The intended use of the hot stage did not require X-ray or back scatter analysis; therefore, increased radiation shielding could be used. The main requirement of a long-life stable heater was the most difficult problem. Contrary to then current thinking, it was discovered that an inductively wound heating coil did not significantly effect the primary electron beam when used with a ripple free D.C. power source.

Heater Design

Aluminum oxide tubes of high density and of a 99.8 weight percent alumina composition were obtained. A nominal 1/4 in. I.D. by 3/8 in. O.D. tube was diamond ground to an O.D. of 0.360 in. This tube was threaded by diamond grinding to give 0.020 in. deep V-grooves with 28 threads per inch. The tube was cut to give sections 0.320 in. in length. A second tube with nominal dimensions of 3/8 in. I.D. by 1/2 in. O.D. was cut to 0.320 in. lengths, and then a 0.10 in. slot was cut in the tube wall. The smaller tube was mounted on a post and wound with six turns of 0.015 in. diameter tungsten wire as shown in Fig. 1. The tungsten wire was then bent and the larger tube slipped over the assembly (Fig. 2). Single bore alumina insulating tubes were slipped over the tungsten wire and the whole assembly covered with a high alumina cement (Fig. 3). Six molybdenum radiation shields were made of 0.003 in. foil with diameters ranging from 0.6 in. to 0.75 in. in 0.03 in. increments. After cutting V-notches in the shields, they were slipped over the heater lead wires as shown in Fig. 4.

Specimen Stage Carrier

A dovetailed metal block originally supplied with the SEM heating device was modified slightly to allow easy access to electrical connections. Aluminum oxide insulators on both sides of the block and one end were used to isolate the five electrical connections needed. An exploded view of the carrier is shown in Fig. 5. The connections shown on the right of the figure are copper connectors used to provide current to the heating coil. The other two electrical connections on the alumina side rails are thermocouple connections. The lower connector is platinum and the upper one of a platinum -10% rhodium alloy. The insulated copper connector at the rear (left) of the carrier is for electrically connecting the top cover plate described later. These five external lugs slide into clips on the main stage when the specimen stage is inserted. The clips for the thermocouple connections are of platinum and platinum -10% rhodium to prevent any junction effects.

Specimen Stage Assembly

Figure 6 shows the heater in place in the specimen stage. Five or six dimpled molybdenum foil disks 0.75 in. diameter placed under the heater provide bottom radiation shielding. The sample cup is either molybdenum or platinum with a platinum-platinum 10% rhodium thermocouple spot welded to the upper rim. The cups are 1/4 in. O.D. by 0.2 in. high with a 1/32 in. wall giving a sample cavity 3/16 in. diameter by 0.170 in. deep. Two high purity aluminum oxide disks 1/4 in. diameter by 1/16 in. thick insulate the sample cup from the bottom radiation shields and position the cup's upper rim flush with the face of the heater assembly. Figure 7 shows the assembled specimen stage and the bottom side of the top cover. The top cover is made by drawing a 3/4 in. diameter by 1/16 in. deep depression in a 0.015 in. thick platinum sheet. Five dimpled 3/4 in. platinum foil radiation shields are held in place by spot welded platinum foil strips. Holes drilled through these foils allow the entrance of the primary beam and exit of the secondary electrons. Holes from 1/8 in. to 3/16 in. diameter have been successfully used. Figure 8 shows the completely assembled specimen stage assembly. The top cover plate is held in position by screws at the front and rear. The front screw is insulated from the top cover by two aluminum oxide washers. The cover is supported on the sides by the aluminum oxide side rails which protrude 0.050 in. above the metal specimen carrier block. The rear screw provides electrical contact to the rear connecting lug. The thermocouple protection tube shown in Fig. 7 prevents the heater or side radiation shields from contacting the top cover.

Main Stage Modifications

To improve resolution, a shorter working distance was required. By transferring the thermal suppression function from the massive grid originally placed between the specimen stage and the pole piece to the top cover of the specimen stage, the stage could be raised the required 19 mm. However, this required tilting the specimen stage slightly toward the air lock so it could be inserted in the main stage. The specimen carrier holder of the main stage was removed and a 19 mm block of stainless steel used to raise it. The specimen stage when inserted in the main stage, therefore, is tilted approximately 15° to the electron beam. Figures 9, 10, and 11 show three views of the main stage. Figure 9 shows the specimen carrier holder with the electrical connection clips. The wires from the thermocouple clips are platinum and platinum-10% rhodium respectively, to place the cold junction as far from the heated stage as possible. The chromium plated metal plate directly above the specimen carrier holder is a beam shutter for alignment of the primary beam. It may also be used in long-time runs to shield the pole piece and aperature from radiation from the heated stage.

Figure 10 shows the specimen stage in place, and Fig. 11 shows the dimensional relations between the beam shutter and the specimen stage.

Hot Stage Operation

Column Pressure and Specimen Contamination

One of the first problems encountered in operation above 1200°C was specimen contamination. The major source of carbon buildup was traced to back diffusion of diffusion pump oil and subsequent cracking of the hydrocarbon gas on the

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hot specimen surface. Installation of a liquid nitrogen cold trap directly above the diffusion pump greatly reduced this problem and resulted in a pressure reduction in the column from 2 x 10^{-5} to 3 x 10^{-6} torr. At 1600°C operation, a buildup of surface contamination has been noted after 1/2 hr. At 1500°C for periods up to 4 hrs. the problem has not been serious enough to impair observations at 3000X. Efforts are continuing to reduce sources of surface contamination.

Temperature Measurement and Control

Two power sources are used for heating the stage. A constant current D.C. power supply is used for manual operation. Temperature drifts of approximately 20°C per hour have been observed. By manual adjustment, a constant temperature is easily maintained.

A variable voltage D.C. power supply driven by an automatic recorder-controller is used for long-time runs or where controlled heating rates are desired. Samples of metallic or ceramic powders have been heated to 1500°C in 5 min.; however, the maximum heating rate is usually limited by the samples outgassing characteristics.

A power of 12 watts to the heater will heat the stage to 800°C and 20 watts to approximately 1050°C. To operate at 1600°C, approximately 60 watts are required.

Temperature measurement at high temperatures has been a continuing problem. Melting copper or nickel in a platinum sample cup has indicated that when the thermocouple is spot welded to the cup rim it produces the best results. Differences of ±10°C from the published melting points have been observed. Having the thermocouple bead in contact or buried in powders or in contact with metallic specimens invariably indicate low temperatures. Samples will melt at an indicated temperature 20° to 50°C below their reported melting points. Spot welding a thermocouple to the bottom or side of the sample cup invariably gives high readings.

Above 1400°C stray fields and leakage currents will affect the potentiometric recorder. The best results are achieved by allowing the temperature measuring circuit which includes the sample cup to float electrically. No noticeable affect is observed on grounding a floating circuit relative to the secondary electron image in either TV or slow scanning modes.

SEM Operation with the Hot Stage

To date most of the hot stage work has been done by TV scanning because of the rapid changes occurring in the samples. Accelerating voltages of 35 to 45 KV and beam currents of 1 to 3 x 10^{-8} amps have given the best results. With these high beam currents and TV scanning the maximum useful magnification is approximately 3000X. Increased resolution and acceptable micrographs have been obtained over 5000X with the temperature at 1600°C by reducing the beam current to approximately 3 x 10^{-10} amps and using slow scanning speeds.

The present stage design gives interesting performance characteristics as a function of temperature. At room temperature with a working distance of 13 mm and using a 1/8 in. diameter hole in the top radiation shield assembly, the TV picture has extremely poor contrast. As the stage is heated, the picture will suddenly improve dramatically. The exact temperature where this change occurs is dependent on the specimen. For silicate glasses this temperature is around 300°C; for nickel microspheres, 600°C; and for aluminum oxide microspheres, 800°C. After this change, excellent contrast and image intensity is maintained until thermal electron emission starts to occur around 1100°C. A negative voltage applied to the top cover plate will then be necessary to repel the thermal electrons from the sample. As the temperature is increased, the necessary thermal suppression voltage increases. Observing either aluminum oxide or tungsten microspheres at 1600°C requires a voltage of approximately 5 volts.

Recording of the TV image has been by video tape, 16 mm time lapse movies, or by a 35 mm still camera.

Over 300 hrs. of hot stage operating time have been accumulated to date. During this period no heater has burned out. The only heater failures have been due to handling. Samples have been continuously observed for 1 hour at 1600°C and to 5 hours at 1500°C.

A variety of metallic and nonmetallic materials have been observed. Microspheres in the 1 to 100 μm size range of Cu, Ni, W, CaF_2, glass, and Al_2O_3 have been heated, many to their melting point. Powders of TiC, Fe_2O_3, and combinations of materials have also been observed. CaF_2 was observed evaporating after heating to a temperature where its vapor pressure was an order of magnitude higher than the column pressure. In no case have any conductive coatings been applied to the samples. Loose powders or powder compacts are placed in the sample cup and heated. A few steels have been observed and phase transformations through surface topography changes noted.

Summary

The details of the modification of a SEM hot stage for the examination of powders at temperatures to 1600°C have been given. The stage design incorporates a stable, long-life heater and provision for thermal electron suppression. Major problems in high temperature operation include

surface contamination, temperature measurement, and image recording.

Acknowledgment

This work was done under the auspices of the United States Atomic Energy Commission.

References

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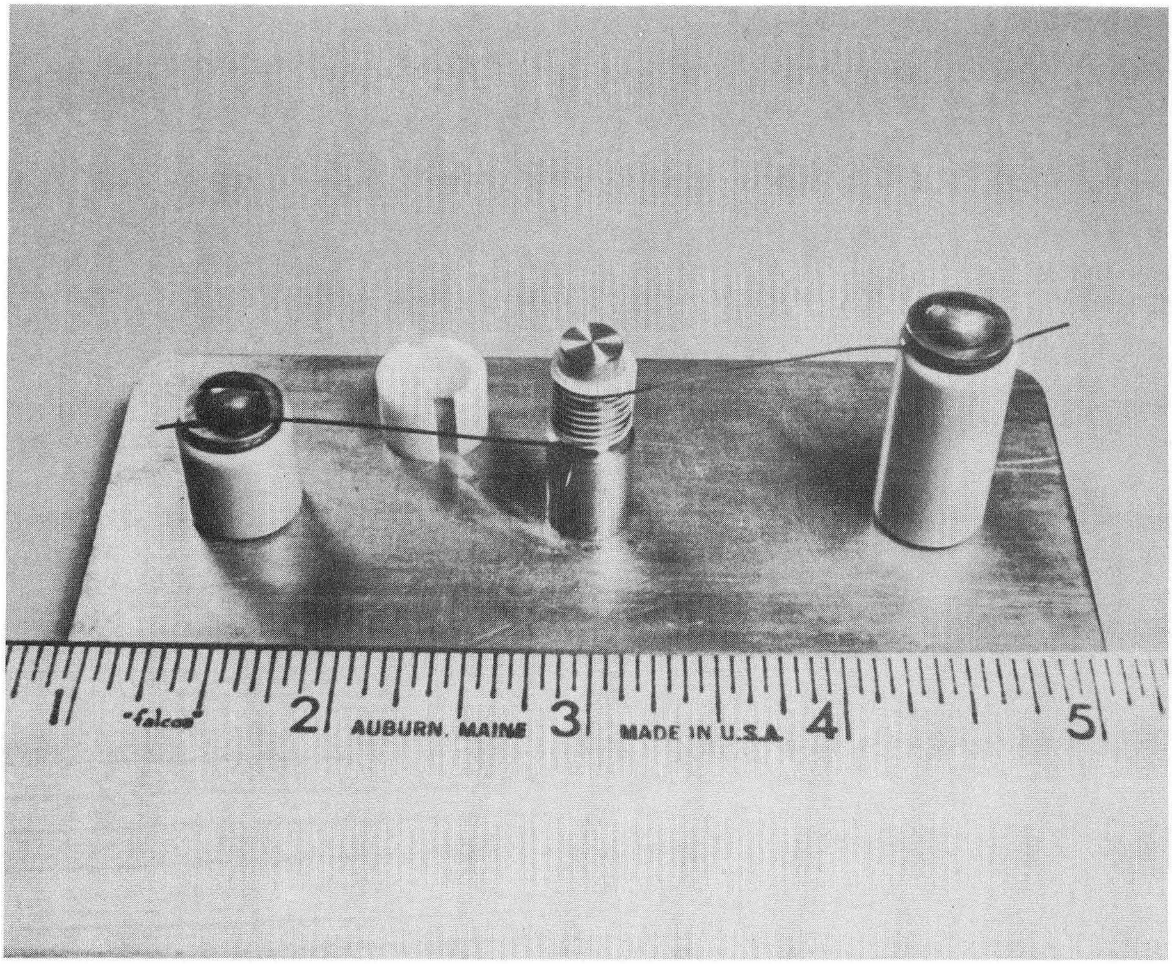

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Fig. 1. Tungsten heater wound on an alumina core and heater wire retaining ring.

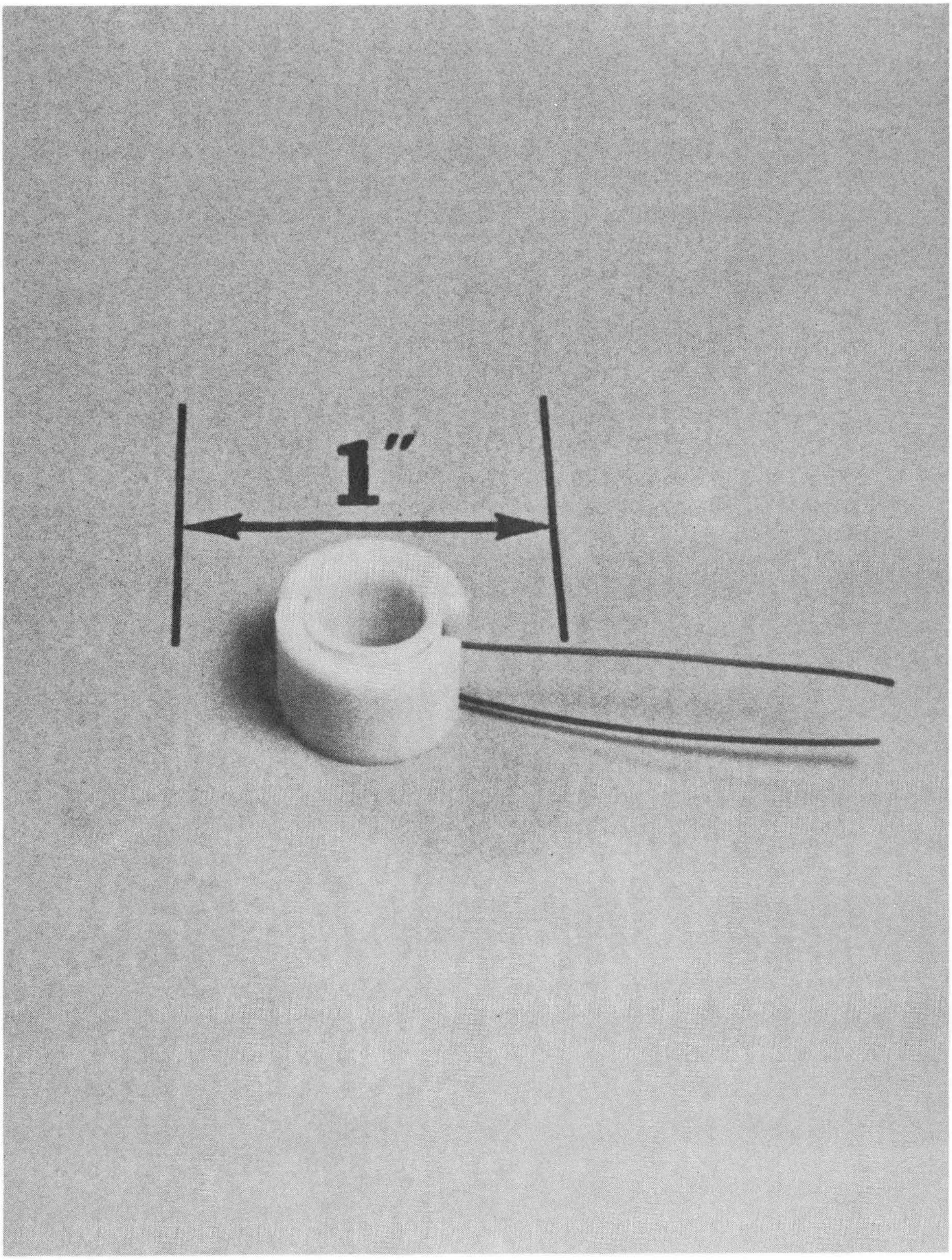

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Fig. 2. Assembled heater.

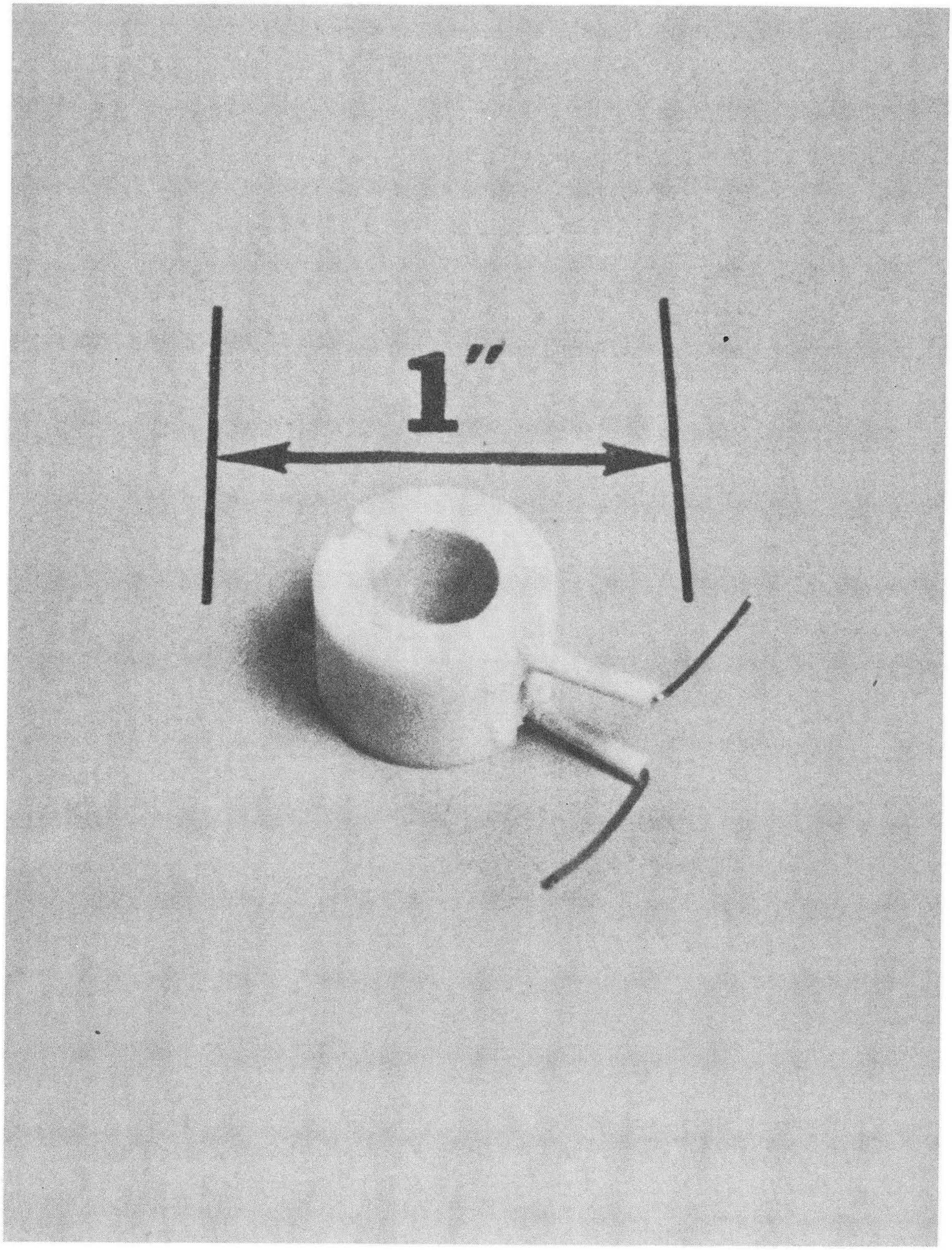

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Fig. 3. Completed heater after cementing.

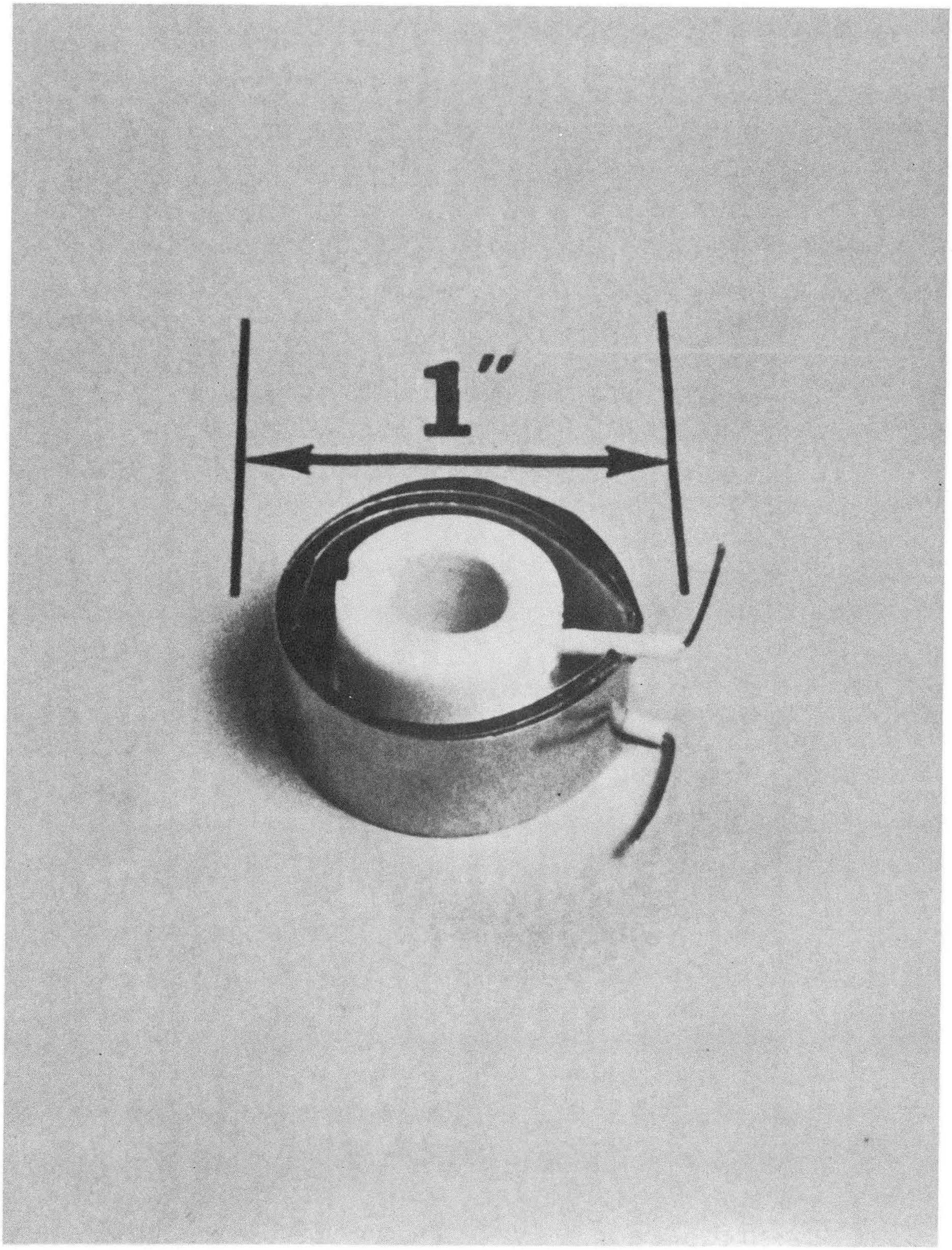

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Fig. 4. Heater and side radiation shields.

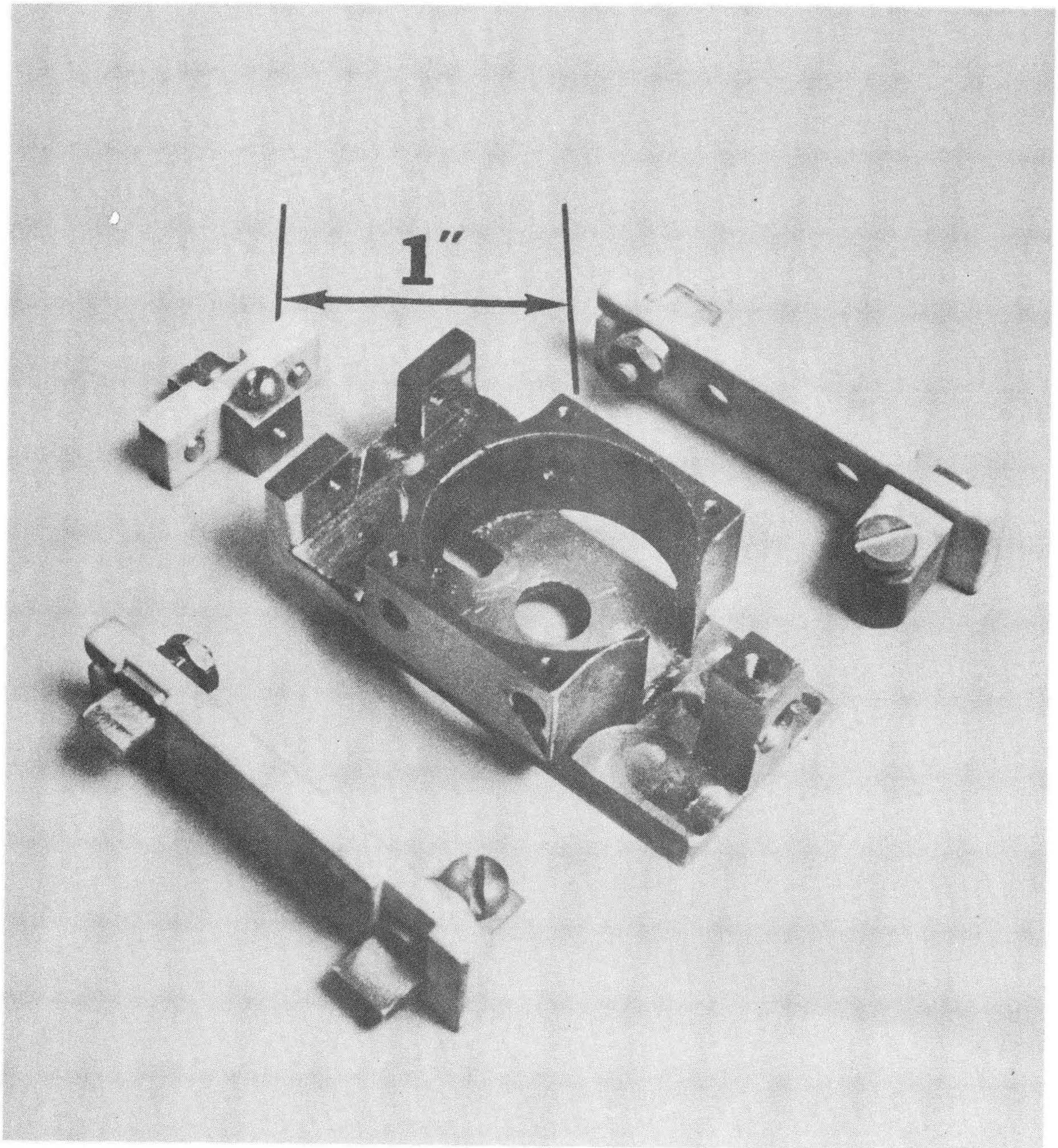

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Fig. 5. Exploded view of the specimen stage carrier block, alumina insulators, and electrical connection posts.

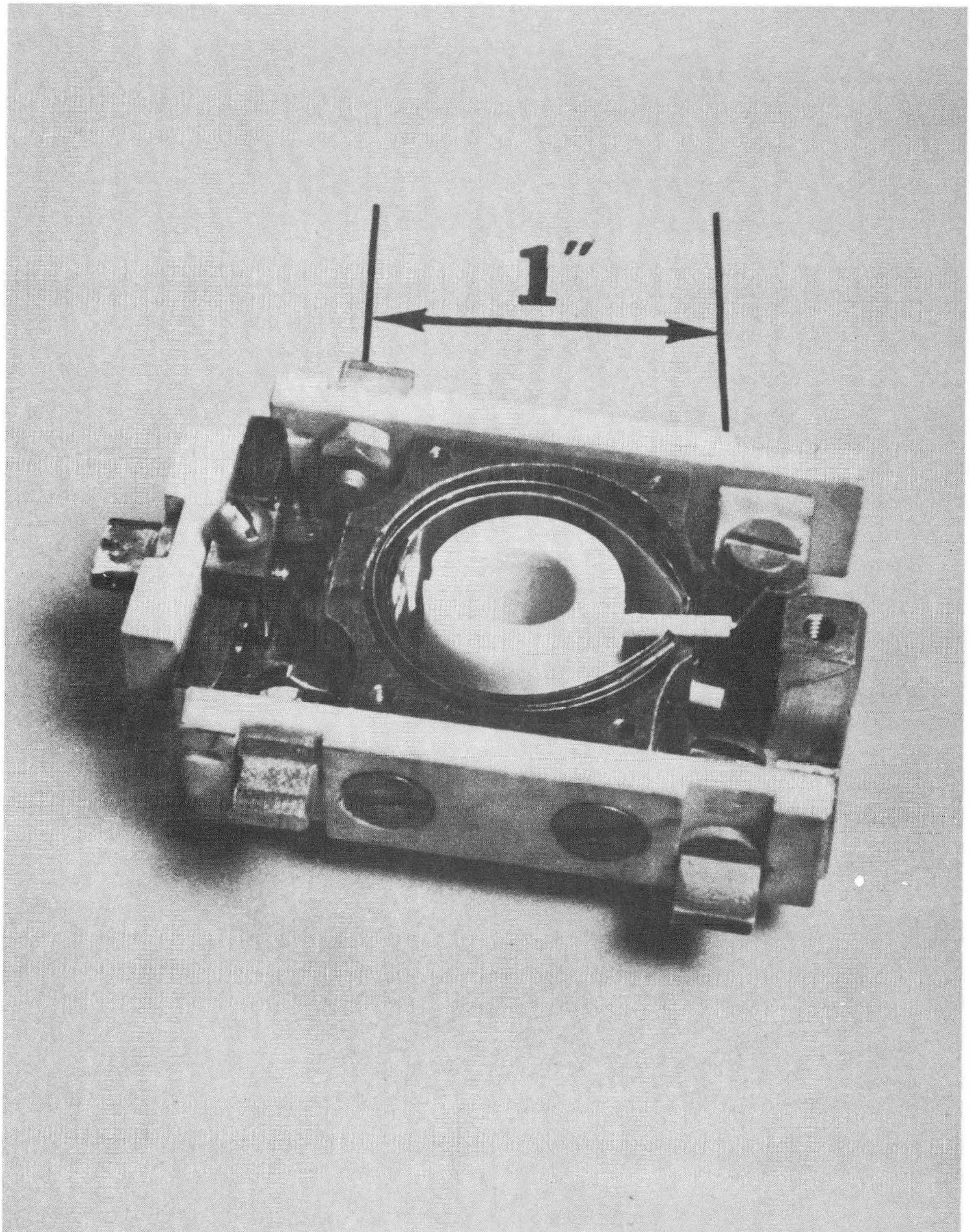

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Fig. 6. Heater assembly mounted in the specimen stage carrier block.

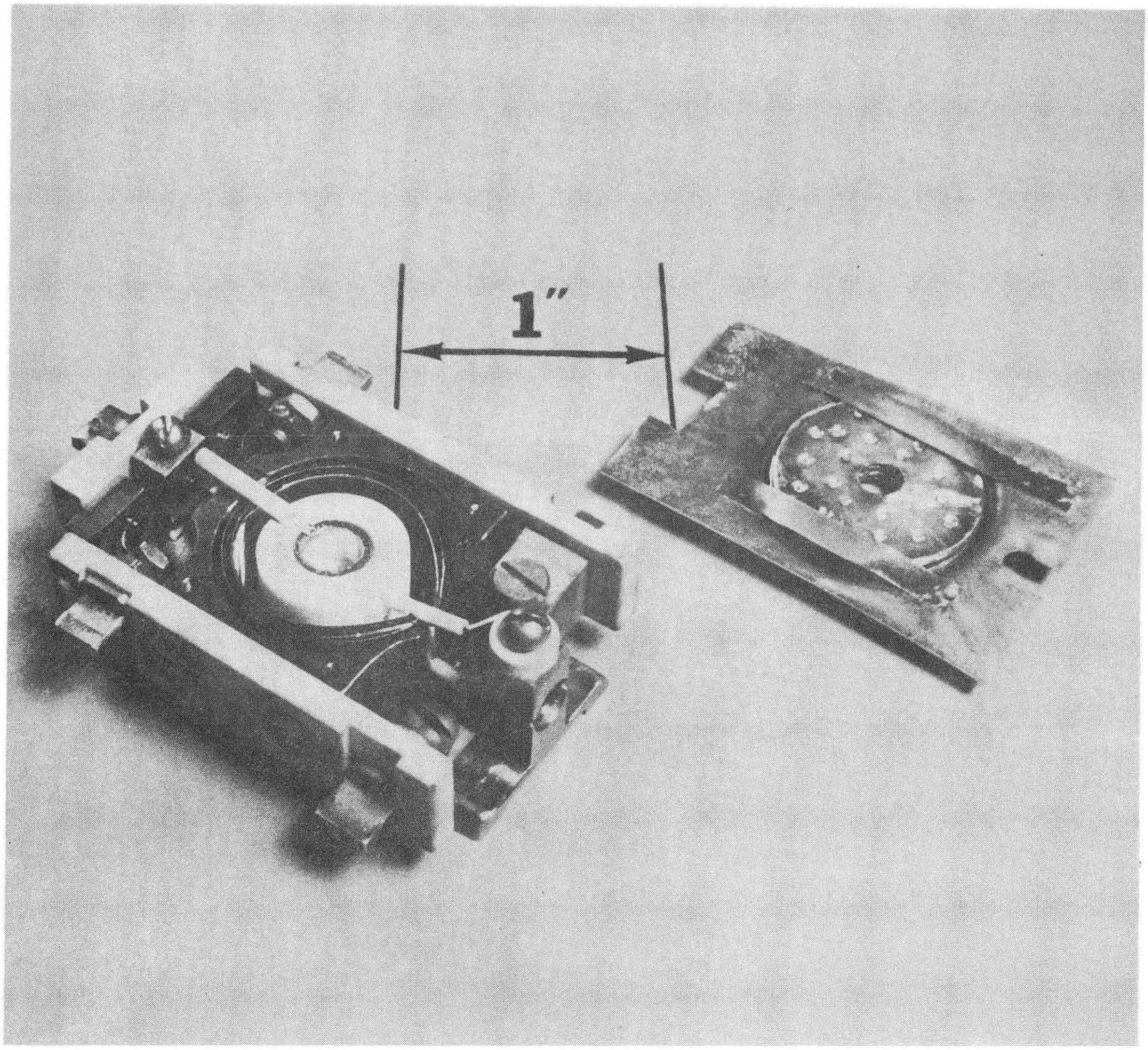

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Fig. 7. Sample cup installed in the heater. Also shown, bottom detail of the top radiation and thermal electron suppression shield.

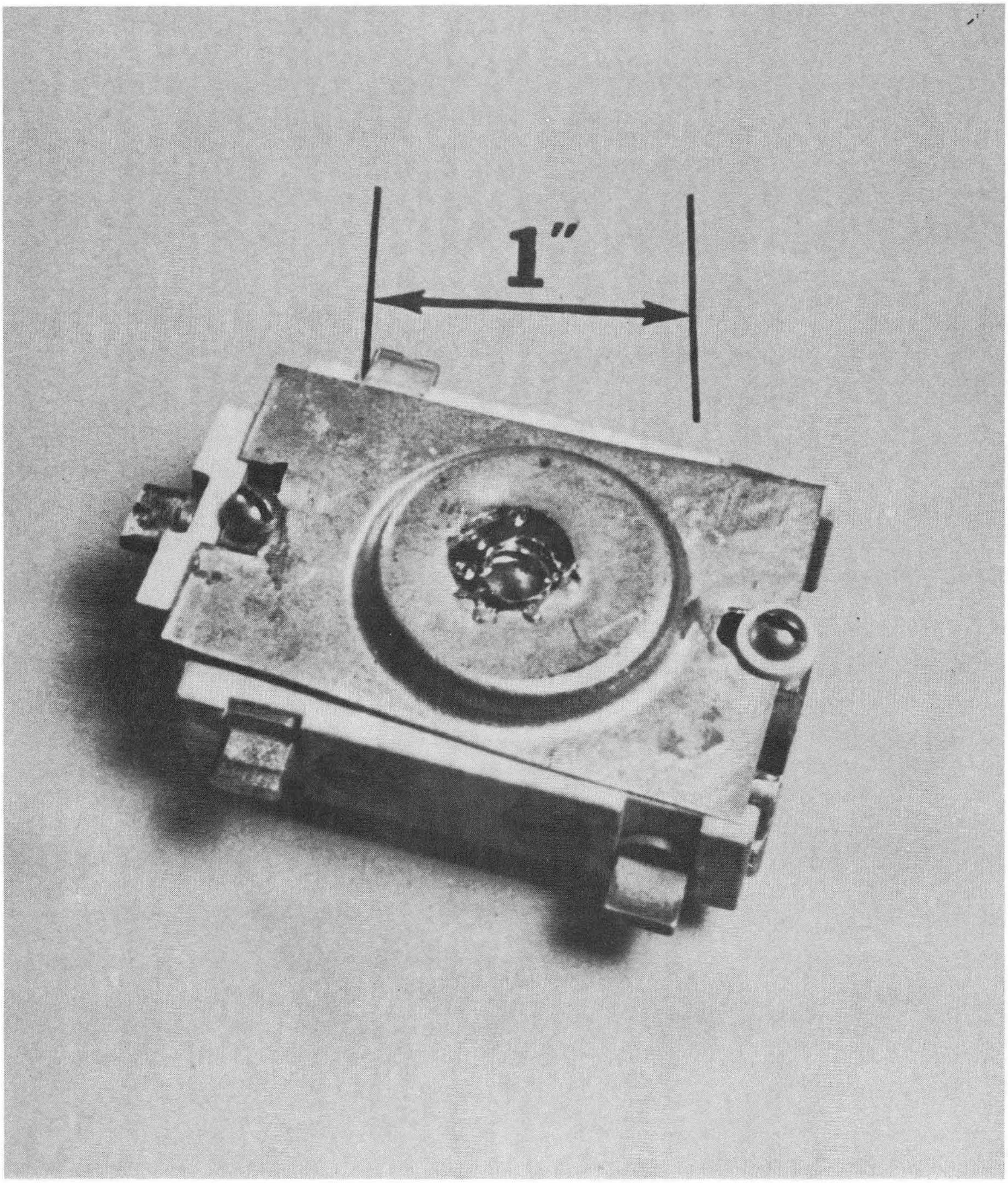

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Fig. 8. Complete specimen heating stage.

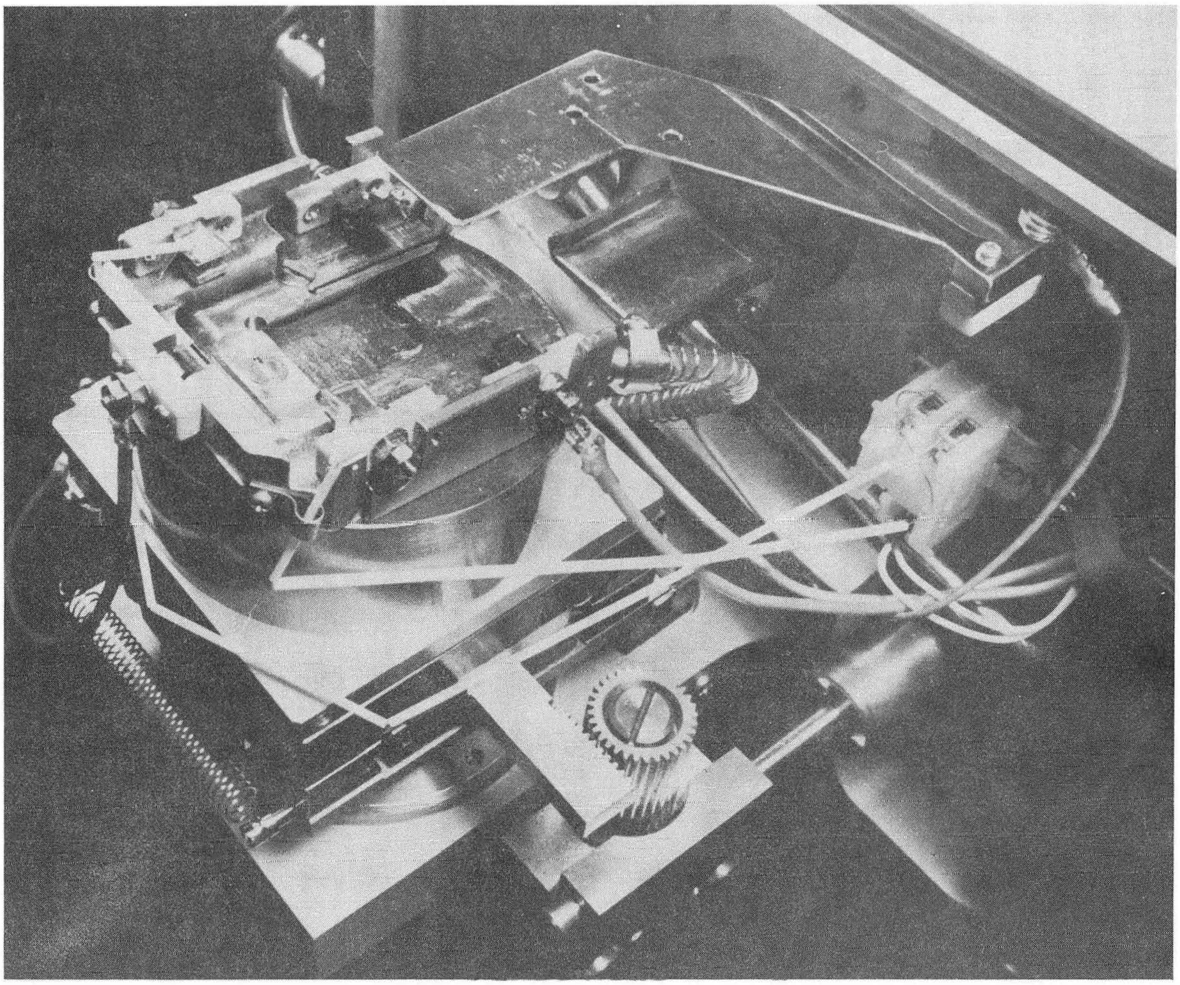

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Fig. 9. Main stage and specimen stage holder detail.

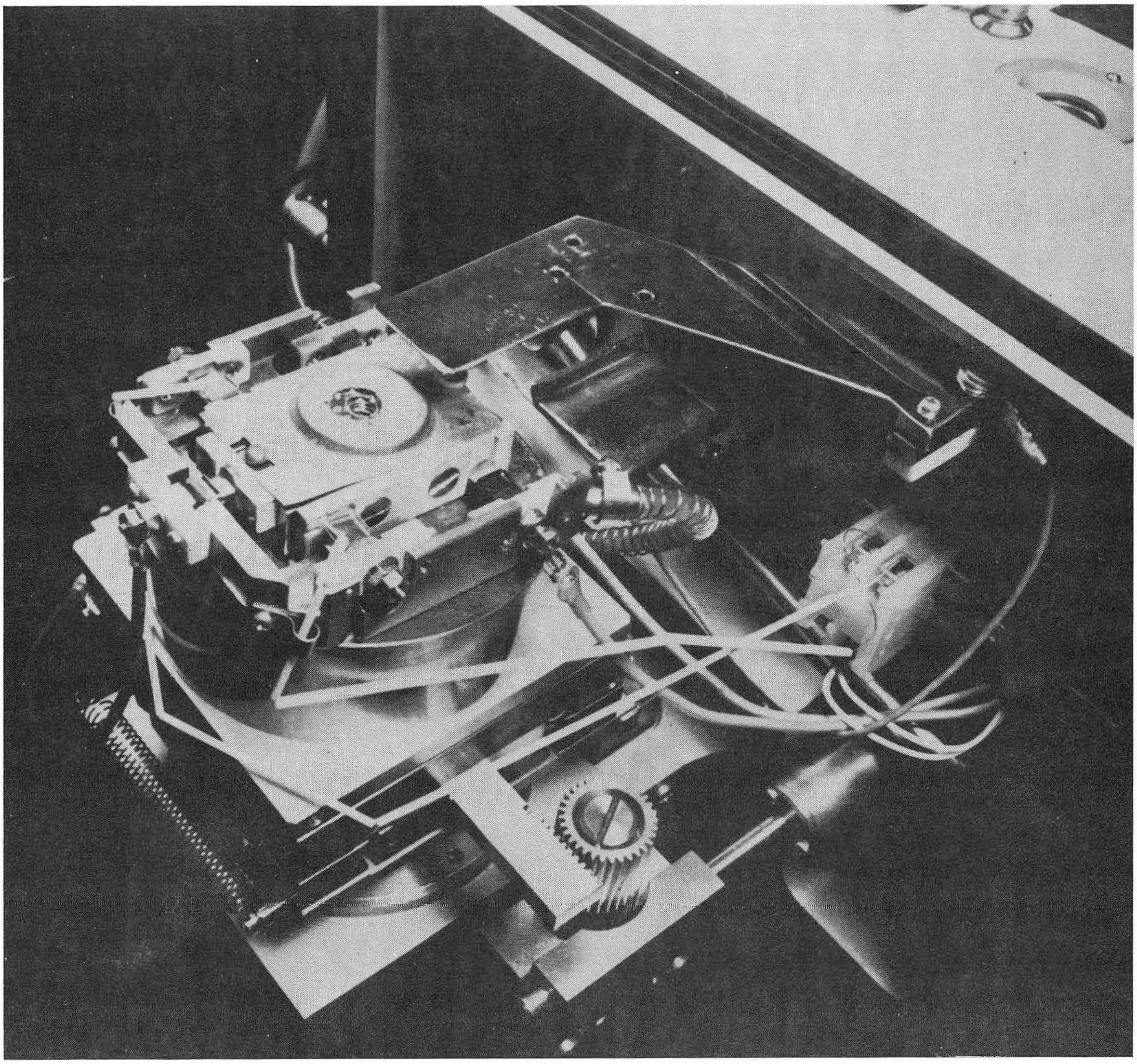

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Fig. 10. Main stage showing specimen stage in its operating position.

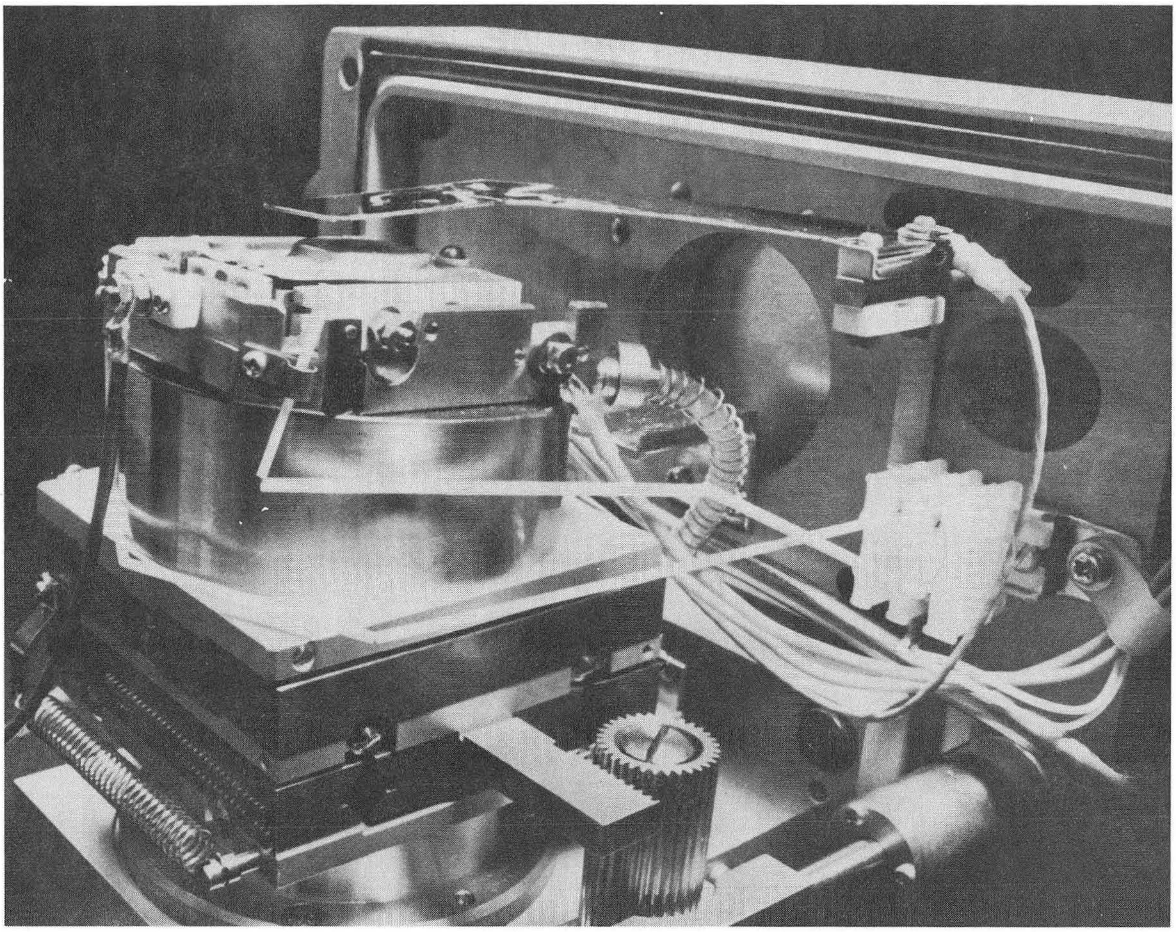

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Fig. 11. Main stage with specimen stage in position and beam shutter closed.

TECHNICAL INFORMATION DIVISION
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720