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Techniques for examining radioactive materials by electron microscopy: Experience and developments in the U.K.A.E.A.

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TECHNIQUES FOR EXAMINING RADIOACTIVE MATERIALS BY ELECTRON MICROSCOPY: EXPERIENCE AND DEVELOPMENTS IN THE UKAEA

by

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SUMMARY

The problems of preparing radioactive specimens and examining them in electron-beam instruments have been considered for the two main types of equipment: the first type, including instruments such as the scanning transmission electron microscope (TEM/STEM), is usually used for examining small, low activity specimens (eg 3 mm discs or extraction replicas). Here the preferred practice is to have specially-designed shielded cells to prepare the specimens up to the final delicate preparation stage, when a shielded fume hood or glove box can provide adequate protection for the operator. It does not appear to be necessary or practical to shield TEM/STEM instruments themselves when examining these small specimens.

For the second type, including instruments such as the electron probe micro-analyser (EPMA) and the scanning electron microscope (SEM), the specimens are usually larger and more active, necessitating fully-remote preparation in the majority of cases. There are advantages in providing only partial shielding of the EPMA or SEM to facilitate maintenance and also in reducing operator dosage to a minimum by loading the sample remotely and separating the control console from the microscope column where possible.

The experience gained on a variety of radioactive specimens at three UKAEA Establishments (at Dounreay, Windscale and Harwell) is presented, together with an assessment of future developments in this field.

x AERE, Harwell
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1. INTRODUCTION

The radioactive specimens prepared for electron microscopy can conveniently be divided into two classes: firstly, the small specimens (usually 3 mm diameter discs or carbon extraction replicas) which are examined in the transmission electron microscope (TEM). Such specimens, by reason of their size and thickness, have a low activity but are fragile and easily damaged. The term TEM is used here to include those microscopes which are made more versatile by the addition of scanning (ie STEM) facilities for analysis.

Secondly, there are the somewhat larger and consequently more active specimens, typically up to 1 cm in size, which are metallographically prepared and analysed in the electron-probe micro-analyser (EPMA). Since the EPMA and the scanning electron microscope (SEM) are similar and sometimes combined instruments, the latter is generally included in this category here, although frequently examinations in the SEM are carried out on rough unprepared surfaces (eg fractures) rather than on polished sections.

The various techniques used for handling radioactive specimens for electron microscopy at Dounreay, Windscale and Harwell are described here, with an indication of future developments in this field.

2. SCOPE OF EXAMINATIONS

The Fuel Technology Laboratories at Dounreay have been engaged for some considerable time in examining by techniques such as transmission electron microscopy and electron probe micro-analysis[1-4], irradiated fissile and non-fissile materials in connection with the Fast Breeder Reactor Development Programme. The TEM examination work has been confined to non-fissile fuel pin cladding and structural components from the fuel pin sub-assemblies and to a lesser extent reactor components, whilst the EPMA work has been concerned mainly with fuel examination and fuel/clad interactions.

The necessary handling techniques for preparing the two specimen types are governed by the need to ensure adequate protection from the plutonium hazard indigenous in the fuel material and, also, to shield against the attendant high beta and gamma radiation fields. Two separate preparation routes have evolved, one for EPMA samples which are fuel bearing, and one for cladding and non-fissile structural materials which may have been in intimate contact with plutonium-bearing fuels or have been exposed to a plutonium-contaminated environment.

At Windscale Nuclear Laboratories, the majority of samples arise from irradiated thermal reactor fuel rods and other highly $\beta\gamma$-active materials, such as irradiated europia, Nimonic PE16 alloy and glasses containing fission product wastes. Short lengths of fuel rod or other material, including metallographically-mounted samples, are cut to provide 3 mm discs and thin slices for TEM and EPMA/SEM examinations respectively.

At Harwell, preparation of TEM specimens of non-fissile reactor materials is simplified because they are in the form of 3 mm discs of various thicknesses before they are irradiated; therefore after irradiation they require only grinding to 0.20 mm thick and thinning to electron transparency in the central region (using jet electropolishing) before examination. Irradiated fuel samples

are prepared in an inert atmosphere in shielded cells, the philosophy being to minimise the size and activity of the samples so that the complexity of shielding required on the EPMA is reduced.

3. PREPARATION OF TEM SPECIMENS FOR EXAMINATION

Shielded cells with lead walls typically 180-200 mm thick are used to prepare 3 mm discs from bulk cladding material, from which fuel residues have been removed. The material is cut using precision diamond-impregnated steel wheels, running at slow cutting speeds and suitably lubricated. Spark-machining is used to trepan the discs out of cladding. For ceramic materials, a rotating diamond-tipped hollow drill is used. Handling the small discs remotely is difficult; vacuum tweezers or adhesive-tipped sticks can be used to pick up the discs, whilst close-focussing TV cameras can be used to give a magnified image of the operations. Until recently, grinding of the discs to the required thickness has been done by hand, but the requirement to handle more active discs has led to the use of metallurgical grinding machines, specially adapted for this purpose (Fig. 1). Although the thinning process itself could be carried out remotely (eg by jet-electropolishing using a commercial system; see Fig. 2), the extreme fragility of the perforated discs makes some intermittent handling necessary.

However, discs 3 mm in diameter and 0.1-0.2 mm thick have only a small mass and, although still requiring to be shielded, the specimen activity is relatively low; if the discs are handled for only very short times using tweezers, the accumulated dose to the hands is comparatively small.

Calculations indicate that using the technique of brief handling times, together with efficient viewing and shielding, the accumulated dose to the hands from the preparation and examination of one specimen of Nimonic PE16 (43% Ni, 33% Fe, 16.5% Cr plus small amounts of Mo, Ti and Al), which has been irradiated for six 65 day periods in PFR with a final decay time of 6 months, should be approximately 0.1 Rem. Thus, even if 100 highly irradiated Nimonic PE16 discs are prepared and examined in one year, the total dose to the hands (10 Rem) is an acceptable fraction of the ICRP recommended for hands and forearms (75 Rem/yr). In practice many specimens will give a lower dose than this.

The concept of a cell capable of being opened has therefore been adopted at Harwell and Dounreay as a reasonable compromise and the lead cells (with 100 mm thick walls) for electropolishing of specimens have been designed so that the doors in the front faces may be opened, to enable the operators to manipulate the discs for short times by hand during the polishing process.

For very active ceramic materials, eg irradiated europia or UO_2 fuel, a petrographic method has been used at Windscale to prepare very thin samples in order to reduce the radiation hazard; the disc, initially 1-2 mm thick, is bonded with wax to a 0.05 mm deep recess in a glass slide, which is in turn bonded to a suitable mount. The disc is then ground using a cast iron lap and abrasive slurry until the sample is level with the glass surface. The complete 0.05 mm thick sample and mount is then cleaned and transferred to a shielded fume hood, for debonding and thinning to electron transparency. For ceramic specimens, this final thinning has been carried out using chemical polishing[5] or ion beam devices. At present this is done using only local shielding of the apparatus; however, with care, specimens having activities up to 1R/h $\beta\gamma$ (in the range 50 to 300 mR/h γ) at 50 mm can be safely handled.

Preparation of irradiated europia presents special problems due to its high specific activity, particularly of high energy gamma radiation, and its chemical reactivity, which has prevented the use of chemical thinning. Ion beam thinning has been tried but was found to introduce defects ~ 2 nm in size, even at low beam angles; this limits its use in radiation damage studies. A third method had to be adopted: a small fragment of the disc was ground in a pestle and mortar and dispersed on a carbon film for TEM studies; this method provided some electron-transparent regions for observation of radiation damage effects in the TEM (Fig. 4a).

The petrographic method is also to be used for preparing 3 mm disc samples from glass containing highly-active fission product waste, in order to study the form and type of precipitation present. Ion beam thinning would appear to be suitable for the final thinning of the 0.05 mm thick glass discs, although this has yet to be proved for active specimens.

The TEM column is designed to shield the operator from X-ray emission and proves adequate to shield him from the $\beta\gamma$-radiation emitted by the small irradiated samples that are examined in the microscope.

4. PREPARATION OF EPMA/SEM SPECIMENS FOR EXAMINATION

The preparation route at Dounreay for EPMA fuel samples has been well reported[2,3,4] and the latest practice differs only in that an internally and externally shielded JEOL JXA 50A instrument is now used, similar to the one installed at Harwell.

During routine ceramographic examination of fuel pin sections in a heavily shielded $\alpha\beta\gamma$ facility, features requiring EPMA examination are identified. A thin slice containing the fuel section is cut from the metallographic sample, using a high speed cutting wheel. The remnants of the original mount are removed and the fuel slice placed in a pre-prepared mount and impregnated with cold-setting resin. This pre-prepared mount contains definable features which permit accurate grinding to known depths. The ground sample is then conventionally polished with diamond abrasives. Following ultrasonic cleaning, the specimen is transferred in an $\alpha\beta\gamma$ transfer flask to a lightly shielded $\alpha\beta\gamma$ facility, where the outer part of the mount is broken away from the inner ring to give a small diameter mount containing the polished thin fuel slice. This small mount is further cleaned ultrasonically and loaded into the EPMA specimen holder. Conducting paint is applied to give electrical contact across the surface and the assembly is ready for transfer to the instrument using a specially designed shielded $\alpha\beta\gamma$ flask.

The adoption of comprehensive local shielding, together with sealed flask loading, has virtually eliminated the radiation and contamination hazard which previously existed when a sample was loaded into the earlier AEI SEM 2 instrument. Experience at Dounreay has shown that for most elements the X-ray spectrometer performance is not significantly affected, even with sample activities as high as 2 Curies (gamma).

Experience at Harwell in using an EPMA to examine radioactive fuels was reported in 1972[6], but the simple facilities described then have been enlarged to incorporate an inert atmosphere preparation suite, an inert atmosphere specimen transfer system and an EPMA with full computer control.

All specimen preparation is performed in a purpose-built cell, which is constructed to high standards and maintains a nitrogen atmosphere with less than 100 ppm oxygen. To minimise contamination the cell is divided into three compartments: two for coarse and fine polishing and one for decontamination, microscopy and despatch to the EPMA (JEOL JXA 50A).

In order to minimise the amount of radiation shielding needed on the machine, the size of the specimens is kept to a minimum. In the case of the polished sections this is achieved by first cutting a section about 2 mm thick from the fuel pin, mounting this on the face of a measured standard mount and then grinding carefully until the specimen is ca 100-150 μm thick, as determined by measuring the mount and specimen. Specimens are thoroughly cleaned by ultrasonic washing and, when possible, by taking Bex film replicas.

The specimen holder, shown in Fig. 3a, is a derivative of the standard JEOL holder and accommodates a 25 mm diameter metallurgical mount; this is used for both X-ray analysis of polished sections and for scanning electron microscopy of fracture surfaces. The holder has an interchangeable with analytical standard block (also shown in Fig. 3a), which accepts 15 of the 1.5 mm diameter Cameca type standard holders.

The specimen flask is shown in Fig. 3b. The specimen holder is mounted on the long shaft shown and is sealed into the flask by means of a vacuum-tight barrel valve, which also acts as a radiation shield. The spout on the front of the flask seals, by a toroidal sealing ring, on to the preparation cell and, when the valve is opened, the specimen holder may be introduced into the cell for loading. The flask is closed for transport and then seals on to the specimen air lock of the electron probe analyser. After the air lock is evacuated, the flask valve is opened so that the flask may also be evacuated; the specimen can be transferred directly to the microscope stage by the push rod. The "air" supply to the air lock is pure nitrogen so that, when the specimen is removed from the machine, the flask can be refilled with an inert atmosphere.

The EPMA has a Tracor computer control which has been modified to permit the examination of radioactive samples. These modifications consist of a development of the JEOL shielded secondary electron detector and biological and instrument shielding. The biological shield consists of a 75 mm lead wall in front of the machine (see Fig. 3c) and the instrument shielding is a 40 mm lead lining to the specimen chamber,together with a further 70 mm of lead in the spectrometer tank. All the machine controls are normally inaccessible and are operated by stepping motors through the instrument control computer.

In the X-ray analysis mode, specimens are normally of an activity of 1 Ci β and 30 mCi γ and against this background we expect a detection limit for most elements of better than 0.25% for a 30 second counting time and a beam current < 25 nA. In scanning electron microscopy the acceptable activity is somewhat higher: 4 Ci β, 400 mCi γ and the resolution is about 50 nm; Figs. 4b,c show some typical microstructures developed in a fast reactor fuel. These were obtained from a longitudinal fracture section of a 7 mm diameter fuel pellet irradiated to 7% burn-up and cooled for about 18 months.

Similar procedures are adopted at Windscale for reducing the size and activity of samples. For example, the petrographic method used for grinding 3 mm discs has also been used for EPMA/SEM samples, where it was required to produce a radial sector of a fuel pin for fission product analysis. The sample produced was ~ 7 mm long, 1.5 mm wide and ~ 0.4 mm thick; a typical sample of UO_2 fuel

at 35 MWd/kgU, cooled for about one year, had a $\beta\gamma$ activity of 1 R/h (50 mR/h γ), measured at 300 mm distance. Such specimens are not loaded remotely but 150 mm long handling tools are used to reduce operator dosage. The EPMA (a CSI Microscan V) is fitted with lead shielding (51 mm thick) around the specimen chamber; an air extract system is also fitted round the specimen loading port, in order to protect the operator from air-borne contamination. So far it has not been necessary to shield the two SEM's (CSI types S180 and S2A) in this way, but it is intended to provide air extract systems on the loading ports. For future EPMA and SEM instruments it is realised that there are advantages in separating the control console from the microscope column, where the design makes this possible.

Ceramic and glass specimens are nornally coated with gold or carbon before examination in the EPMA and SEM, to improve surface conductivity. The coating is done in commercially-available vacuum equipment, using local lead shielding to minimise the radiation hazard.

5. FUTURE TRENDS

The methods that have been used until now for examining radioactive specimens by electron microscopy have been proved to be safe and reliable, giving low cumulative radioactive exposures to the operators (typically $< 1/10$ of the permitted maximum); however, specimen activity levels are expected to increase and permissible exposure limits are likely to decrease and must be kept to a minimum. In order to achieve this, it is recognised that the small specimens have to be prepared in shielded cells specifically designed for the purpose, but with the capability of permitting limited handling in the final stages of preparation (Fig. 5). The three AEA establishments concerned are therefore improving their techniques and preparation facilities in this respect. Future developments in specimen handling and electropolishing techniques may eliminate the need to open the cell fronts, with consequential reductions in operator doses. One such improvement would be the development of a mini-manipulator which would facilitate the precise remote handling of such small objects as electropolished 3 mm diameter disc specimens. Engineering Division, AERE, is undertaking the design, development and manufacture of such a mini-manipulator and a prototype is now nearing completion (see Figs. 6a, b). The device is essentially a tele-operator: there is only electrical connection between the control unit and slave. The position of the controller is thus not fixed. The slave unit has seven movements to give a full simulation of finger, hand and arm movement, and has been designed to pass through a standard port in a cell wall. It will handle weights up to 0.5 kg with a precision of 0.1 mm and with slight modification may be wall or floor mounted. A mini-manipulator of this type is likely to have many applications and a more robust model based on similar principles and designed to handle weights up to 10 kg is now under consideration.

In future it may become necessary to load TEM specimens into the sample holders using a small shielded glove box, but it is not likely that any additional shielding of the microscope column will be needed for the examination of such small specimens. It will probably be necessary to provide yet more shielding for the EPMA and SEM instruments dealing with very active specimens and the need to control air-borne contamination is likely to increase in importance; the experience gained at Harwell shows that the combined use of moveable shield walls, local shielding of the specimen chamber and the use of a sealed transfer system for an EPMA provides adequate protection for the operator, without increasing the difficulties of instrument maintenance.

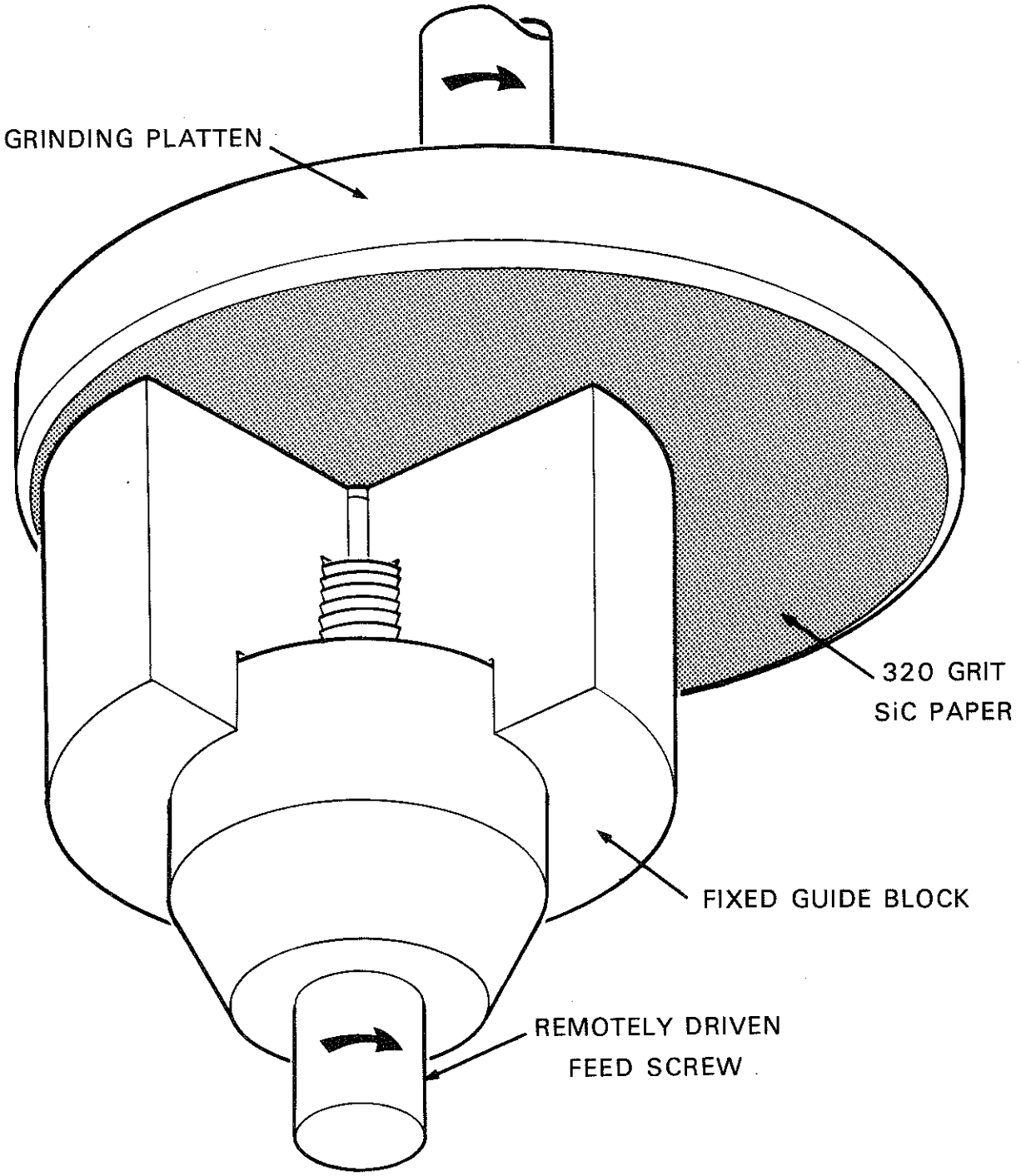
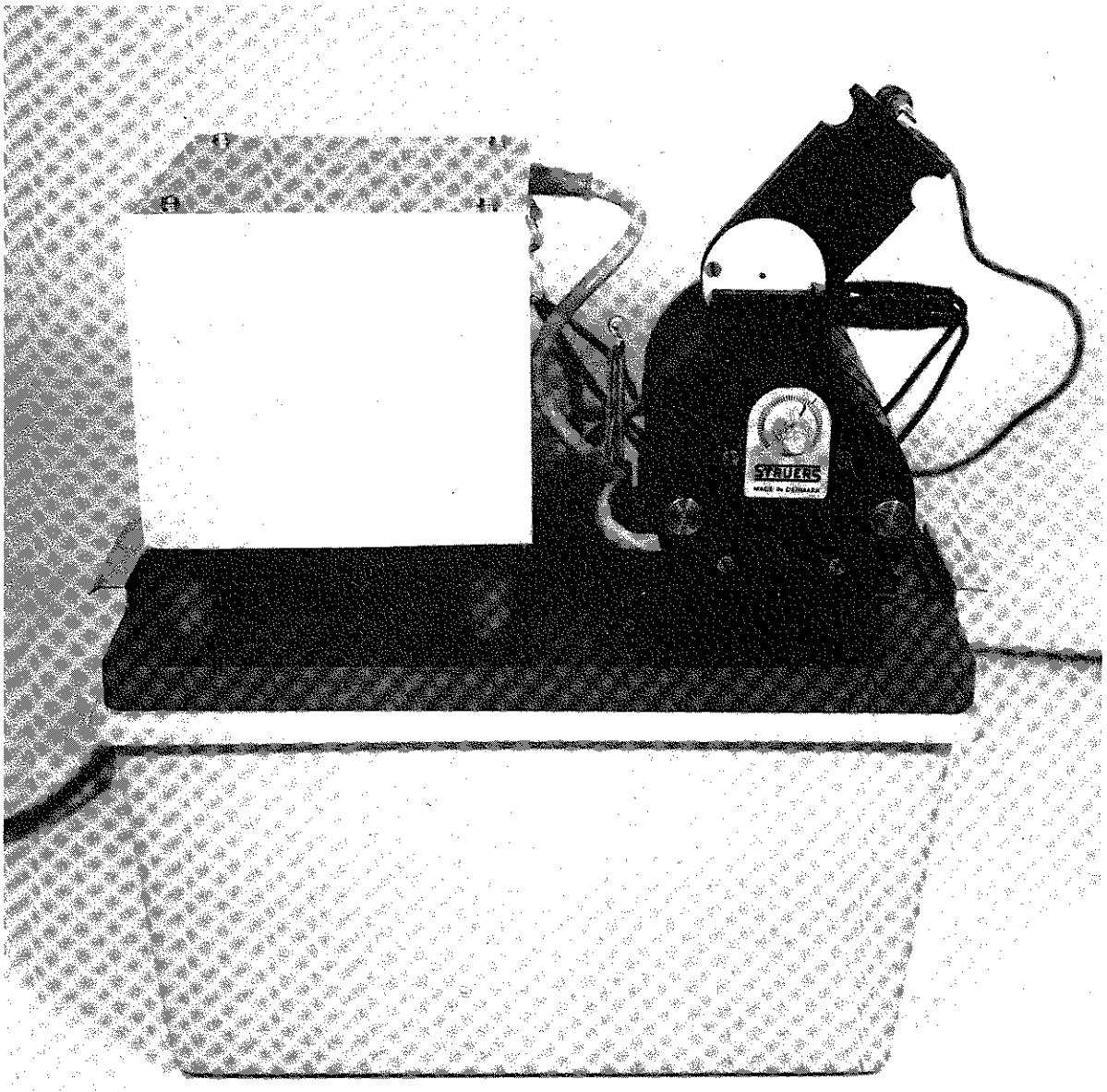


FIG. 1 ESSENTIAL FEATURES OF A PROPOSED DESIGN FOR A REMOTELY OPERATED T.E.M DISC GRINDING MACHINE. THE PLATTEN IS RETRACTABLE TO REPLACE THE GRINDING PAPER AND TO INSERT AND REMOVE THE DISC.



(a)

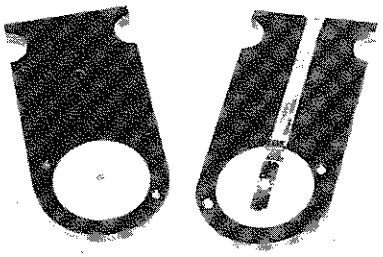
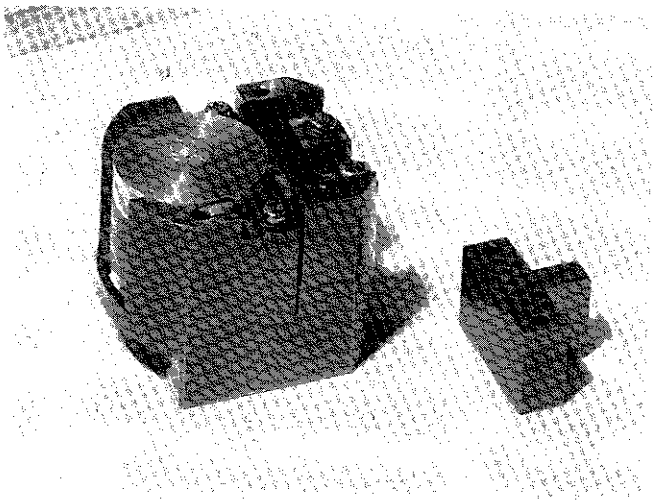
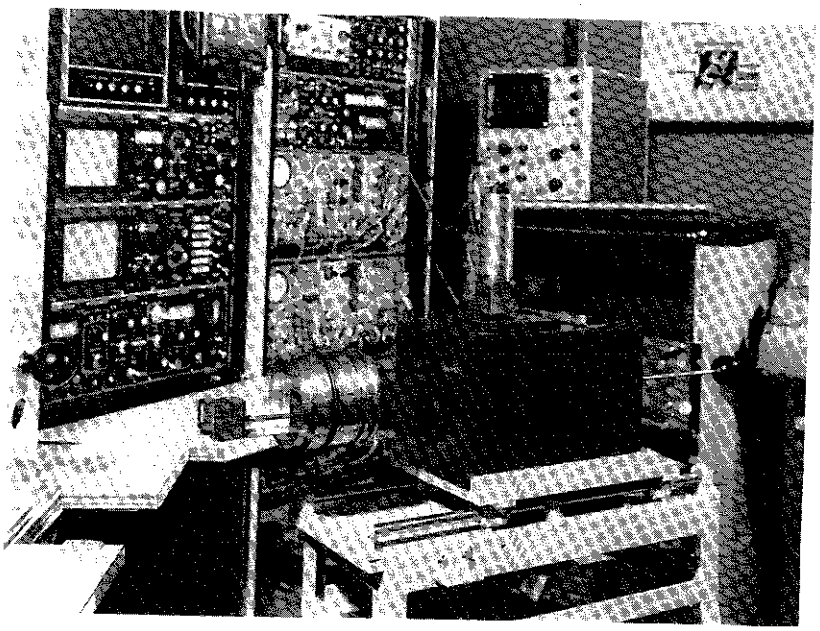


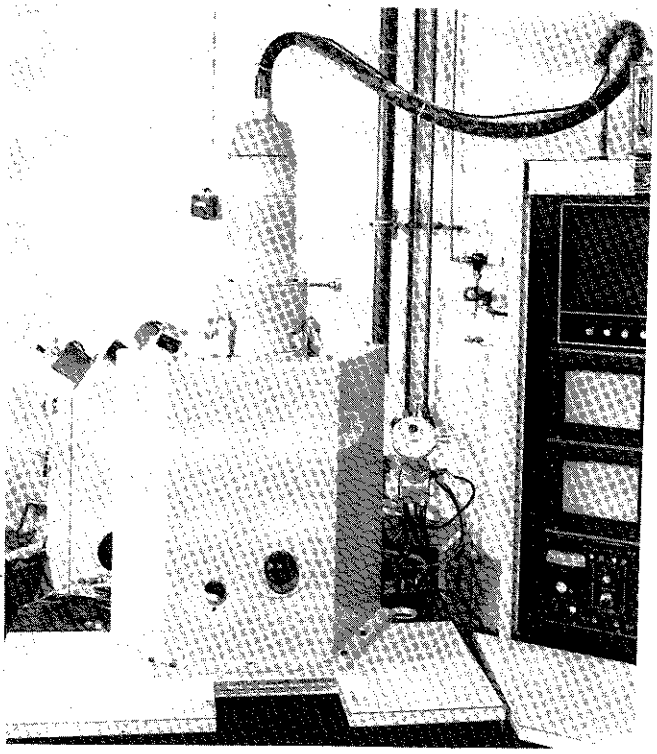
Fig. 2. Commercial Struers 'Tenupol' electropolishing system modified for remote polishing of TEM discs by separating the alarm circuits and pump controls from the in-cell polishing bath.
The split specimen holder is shown at (a). (Dounreay DNPDE)



a. Specimen holder, together with block for analytical standards.



b. Shielded specimen transfer flask and control console.



c. Shielded EPMA column.

FIG 3 EPMA equipment for radioactive specimens at Harwell

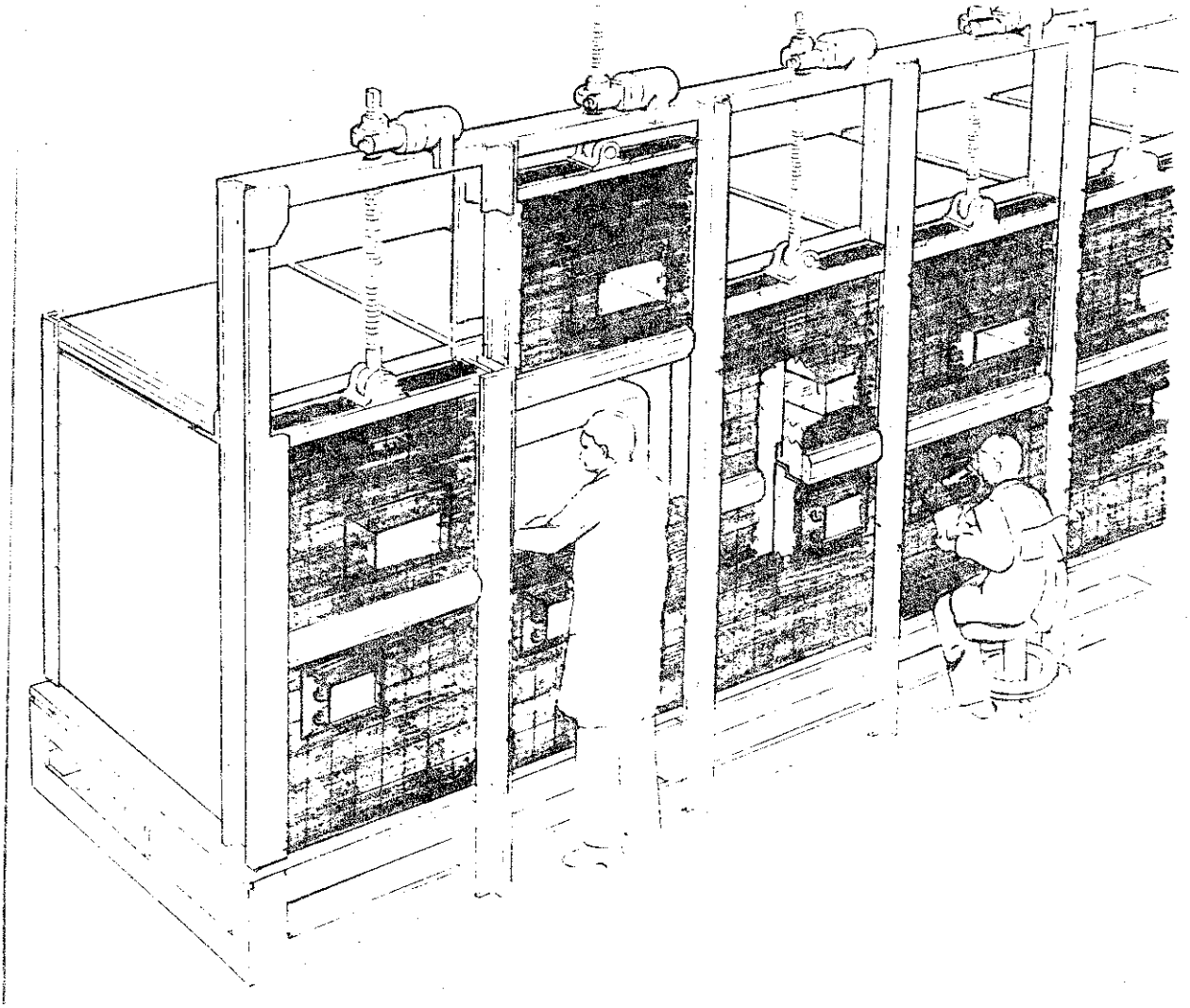


Fig 5 Drawing of proposed electropolishing cells at Harwell

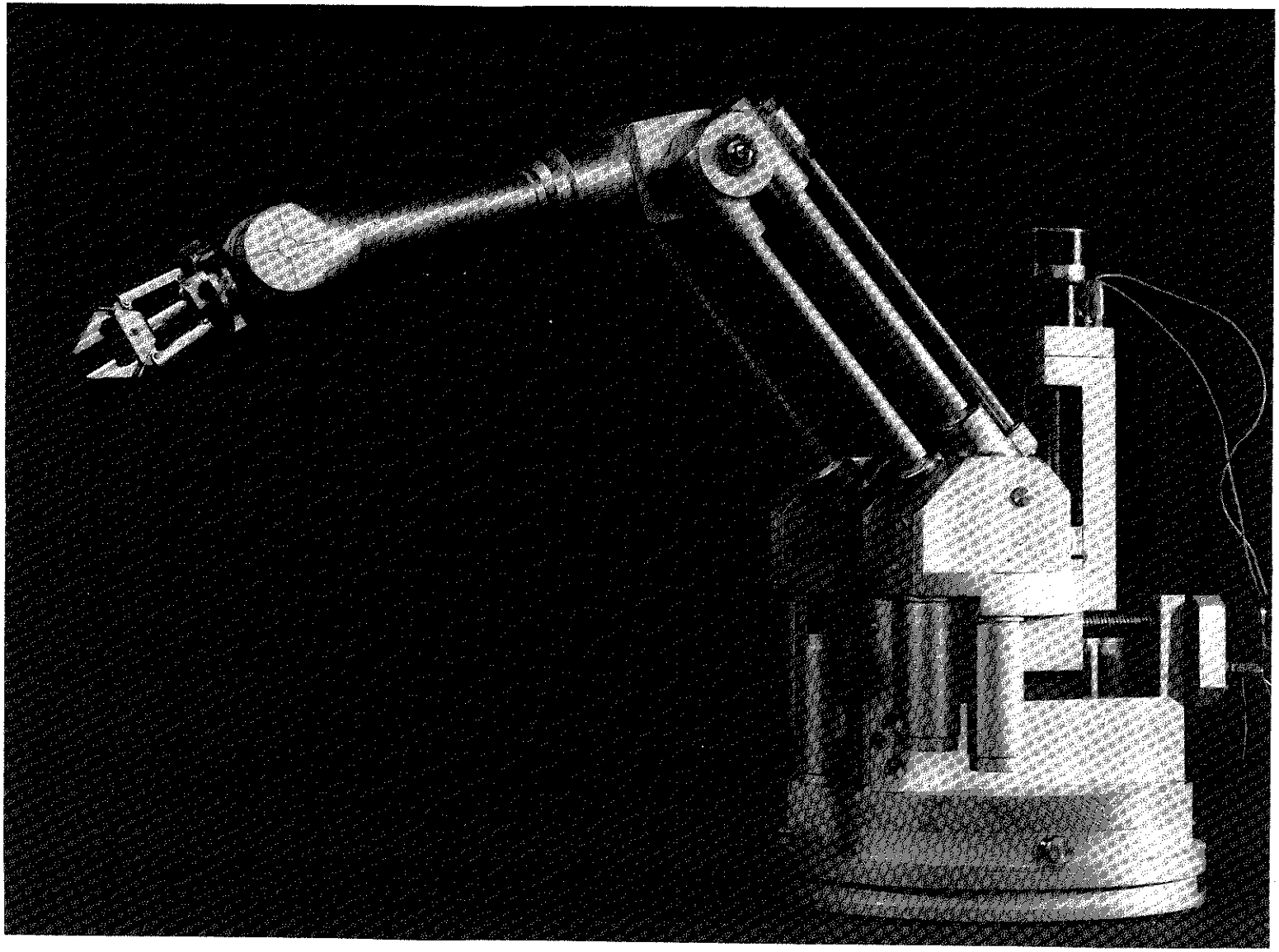


FIG 6a. PHOTOGRAPH OF MICRO-MANIPULATOR DEVELOPED AT HARWELL FOR HANDLING SMALL RADIOACTIVE SPECIMENS

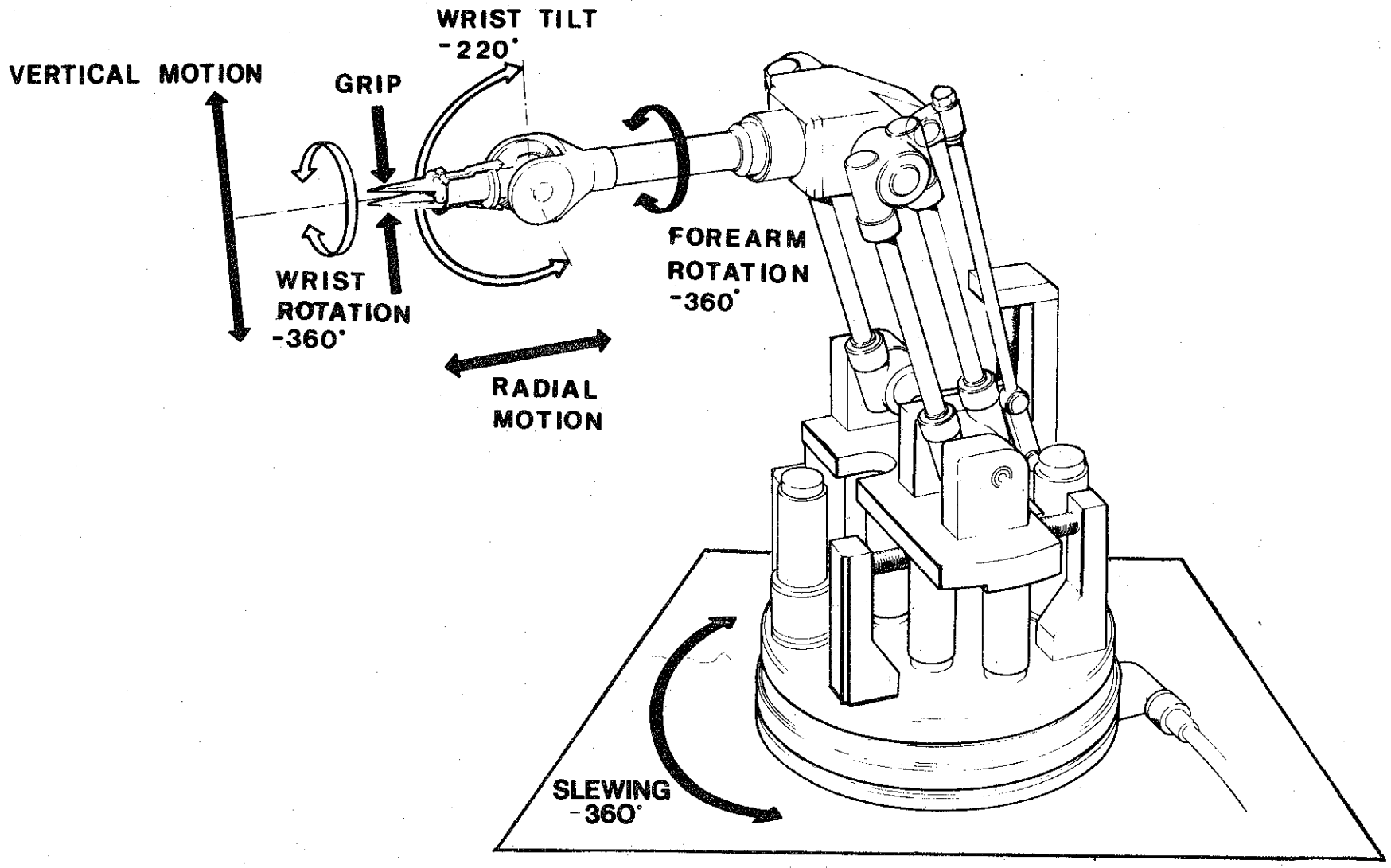


FIG 6b. DRAWING OF MICRO-MANIPULATOR SHOWING MOVEMENTS AVAILABLE

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