PRECISION ION MILLING OF FIELD-ION SPECIMENS

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Abstract - *The advantages and disadvantages of a precision ion milling system for the fabrication of field-ion specimens have been examined. This technique may be used on a wide variety of materials. As precise control over the ion beam position is possible, the specimen shape can be sculpted to a desired end form. Repeated ion milling and TEM examination is simplified since the same specimen holder is used for both processes. This technique is ideal for positioning a feature of interest at the specimen apex.*

INTRODUCTION

The inability to fabricate high quality field-ion microscopy (FIM) specimens can sometimes be a barrier to successful atom probe investigations. In the vast majority of cases, appropriate needle-shaped specimens may be produced by conventional electrochemical or chemical methods. However, in cases in which the material cannot be fabricated into specimens with these methods[1], ion-milling techniques may be applied.[2,3] Although ion milling is a routine procedure in transmission electron microscopy (TEM) specimen preparation, it has been applied only sparingly to FIM. In addition to fabricating specimens from bulk material, ion milling may also be used to resharpen a blunt or fractured FIM specimen and remove surface contamination. Blunt specimens have also been resharpened in the microscope by reversing the voltage on the specimen in the presence of an image gas.[4-8] Waugh et al.[9] used a secondary ion mass spectrometer (SIMS) equipped with a liquid metal ion source to fabricate and reshape specimens.

In this study, the advantages and disadvantages of a precision ion milling system (PIMS) for fabrication of FIM specimens from metallic, ceramic and semiconducting materials have been evaluated. This instrument differs from conventional ion milling systems in that it removes material from the specimen with a steerable focussed ion beam of micrometer rather than millimeter dimensions.

Precision ion milling offers the advantage of being able to control the position of the beam on the specimen. This ability enables the specimen geometry to be sculpted into a desired end form, and therefore permits the correction of an asymetric end form or allows material to be removed preferentially from the specimen until a desired feature, such as a grain boundary or phase boundary, is in the analyzable volume of the needle. Precision ion milling therefore provides an alternative technique to pulse polishing and micropolishing followed by previewing in the TEM[10-13] or scanning electron microscope[2].

EXPERIMENTAL PROCEDURE

A commercial Gatan model 645 precision ion milling system was used for this investigation. Selected portions of the specimen can be ion milled at a chosen voltage (1, 2, 4, 6, 8, or 10 kV) with a rastered ion beam generated from for example a noble gas, hydrogen, nitrogen or oxygen. Argon is the most commonly used ion beam source. The ion beam diameter is dependent on the voltage used and the objective and condenser lens excitations. The smallest beam diameter that can be obtained is approximately 1.5 μm (FWHM) at 10 kV. The specimen is introduced into the system in a standard TEM holder which has been modified to enable the examination of FIM specimens. The orientation of the ion beam is perpendicular to the needle axis. The specimen is stationary during the PIMS process; however, the specimen holder can be rotated 360° about the specimen axis and thus the specimen can be accurately oriented with respect to the ion beam. In the PIMS, the specimen can be imaged with either secondary ion or secondary electron signals, with a maximum magnification of approximately 1500 times. Since the smallest beam diameter in the PIMS is larger than the near-apex shank diameter for a typical FIM specimen, the specimen apex is poorly resolved. A typical secondary electron image is shown in Fig. 1. The box defines the selected area over which the ion beam is rastered during the milling process. The size of this region can be varied by the operator. The specimen is oriented such that its apex is clearly within this region.

The current during precision ion-milling is determined by the operating voltage, the diameter of the collimating aperture (small - 310 μm, large - 1.1 mm), the probe size (small, medium or large), and the area

over which the beam is scanned. A plot of the current as a function of the operating voltage for various probe size and aperture combinations is shown in Fig. 2. For most of the studies conducted here, an argon ion beam with a medium probe size, a large aperture and an operating voltage of 2 kV or 4 kV was found to be appropriate. These conditions resulted in a current density of a few amperes/m^2, at 2 kV.

To optimize the process, specimens are periodically removed from the PIMS and examined in a TEM. Philips CM30, CM12 and 400TFEG electron microscopes were used for the TEM examination. For the APFIM examination, the specimens were examined either in the ORNL or Oxford atom probes.

A wide range of materials with various initial preparation steps was used. Specimens of type-308 stainless steel, Ni_3Al, and silicon were initially prepared by standard electrochemical or chemical methods. To reduce the ion-milling time required, refractory TiB_2 specimens were initially ion-milled in a conventional Gatan 600 ion-miller with a modified specimen holder. Silicon carbon whiskers were mounted on blunt tungsten needles with a conductive epoxy and ion-milled solely in the PIMS instrument.

RESULTS AND DISCUSSION

In a conventional ion-miller, it is difficult to position a fine needle to ensure that the specimen apex is within the ion beam. In addition, if the whisker is not on the exact axis of the rotating holder, the apex may periodically rotate out of the ion beam during milling. In the PIMS, the specimen can be imaged and the ion beam positioned to ion mill only the apex region. Since the specimen in the PIMS is stationary, it is best to precision ion mill the specimen from several different directions in turn. The advantage of the PIMS is that at each orientation, the beam is refocussed directly onto the whisker itself. A FIM image of a silicon carbide whisker which has been prepared in this manner is shown in Fig. 3. A combination of ion-milling in a conventional system and a PIMS can also be used to fabricate specimens from bulk material. A field-ion micrograph of a TiB_2 specimen produced from bulk stock is shown in Fig. 4.

Tip Sculpting:

The outlines of successive TEM images of an aged type-308 stainless steel weld specimen taken during the PIMS process are shown in Fig. 5. Close examination of Fig. 5 reveals that as little as 20 nm can be successfully and controllably removed from a specimen apex. The initial and final TEM images are shown in Figs. 6(a) and 6(b), respectively. This specimen was precision ion milled almost entirely with the ion beam normal to the orientation shown in the uppermost images of Figs. 6(a) and 6(b). The silicon specimen shown in Fig. 7 was initially elliptical in cross-section. By ion-milling normal to the orientation shown in the uppermost images of Figs. 7(a) and 7(b), a well formed FIM needle was obtained. The FIM image of this silicon specimen is shown in Fig. 7(c). It is clear that by appropriately selecting the direction from which to precision ion mill, the aspect ratio of blade-shaped specimens can be reduced.

The previous examples demonstrate that, as expected, the specimen preferentially narrows normal to the ion beam. However, if the specimen becomes too narrow, the result is catastrophic and the end of the needle becomes filamentary and often curls back on itself, as shown in Fig. 8. In order to prevent filament formation, ion milling should be terminated before the diameter is less than 50 nm.

The PIMS process also results in the simultaneous sharpening of the specimen apex. Therefore, specimens which were fractured in a previous FIM examination can be resharpened. A fractured stainless steel specimen that was successfully resharpened to an apex radius of 25 nm is shown in Fig. 9. This can be an important consideration in cases where there is limited material available for the APFIM investigation.

The arrows in Fig. 5 indicate stages during which milling was performed from the directions marked. This procedure can be used to advantage to sculpt the specimen. In order to reshape a specimen, it should be thoroughly characterized in the TEM first. The widest portion of the blade or the desired microstructural features can be identified and appropriately oriented with respect to the ion beam in the PIMS. Since the same holder and a similar goniometer are used for both TEM and PIMS procedures, this is a trivial, yet important step.

Feature Locating:

In an investigation of the decomposition of the ferrite phase in a duplex type-308 stainless steel weld, the ferrite volume fraction was only 12%. Standard techniques of previewing and pulse-polishing/re-polishing had a low success rate for obtaining ferrite within the analyzable region. Through the use of the PIMS, specimens with ferrite regions within a micrometer of the specimen apex were ion milled such that the austenite/ferrite interface and the ferrite could be analyzed. A composition profile across the interface in Fig. 6 was successfully obtained and the chromium and nickel partitioning to the two phases is shown in Fig. 10. The above technique has also been used to position grain boundaries in Ni_3Al at the specimen apex.

The narrowing of the specimen during the PIMS process limits the distance to which one can controllably mill back to a desired feature. In general, this distance tends to correspond to several initial apex radii, or about 1 μm for an initially blunt specimen. Another difficulty that must be considered is preferential milling. The effect of grain orientation on the ion milling of Ni_3Al is shown in Fig. 8. This effect can be advantageous, as shown in Fig. 11, where the phase of interest, ferrite, was preferentially retained at the specimen apex.

Other Considerations:

The stationary ion-milling process can result in surface ridges similar to those observed in stationary ion-milled TEM specimens. Ridge formation is exacerbated at higher operating voltages, as shown in Fig. 12. The ion-milling process also introduces defects (and argon ions) into the surface layers of the specimen. The thickness of the affected surface layer can be reduced with the use of lower operating voltages. An amorphous surface layer may form during ion milling and often a surface hydrocarbon film results from the TEM examination. The affected surface layers should be field-evaporated from the specimen in the FIM prior to imaging or APFIM analysis.

CONCLUSIONS

Precision ion milling is a very useful tool in the preparation of field-ion microscopy specimens. A variety of materials have been successfully precision ion milled including metallic, intermetallic, ceramic, and semiconducting materials.

The advantages of PIMS include:

- A specimen may be ion milled to selectively remove material so that a specific microstructural feature is within the analyzable region
- The geometry of the specimen apex may be sculpted to a desired shape
- Sequential milling and TEM examination is simplified as the same specimen holder is used

Several aspects of the PIMS process must be considered including:

- If the needle becomes too narrow, an undesirable filament may form
- Preferential ion milling may occur at specific grains, grain boundaries or phase boundaries
- Care must be taken to field-evaporate all surface layers which result from either the ion-milling process or from the TEM examination

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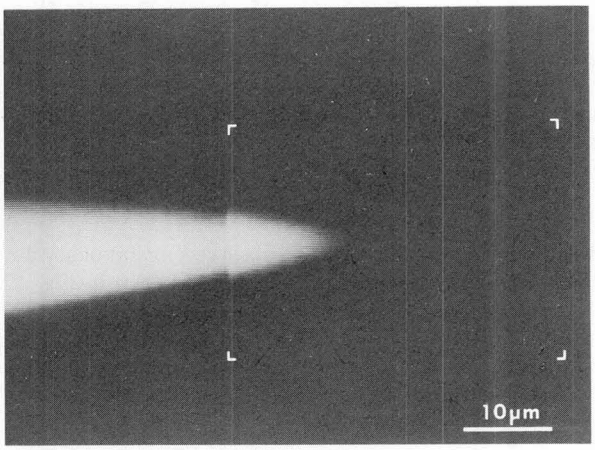

Fig. 1. Secondary electron image of a FIM specimen in the PIMS.

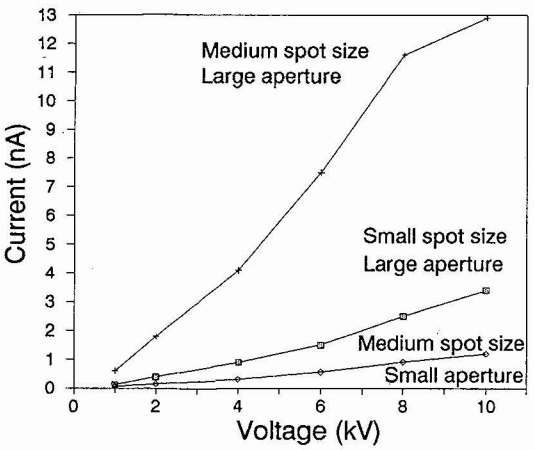

Fig. 2. Ion current as a function of the operating voltage for various probe sizes and apertures.

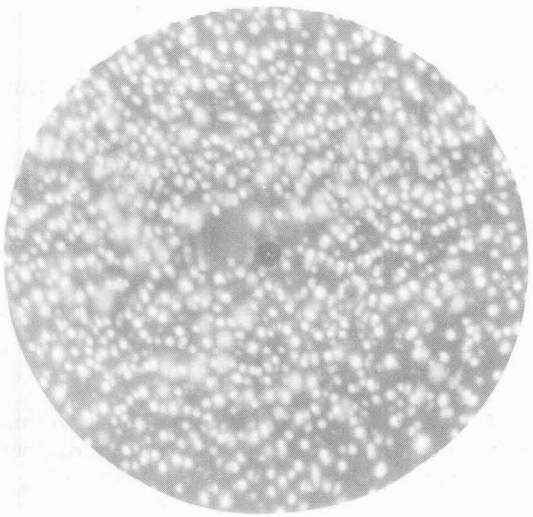

Fig. 3. Neon FIM micrograph of a silicon carbide whisker. Specimen temperature was 40K and BIV was 8 kV.

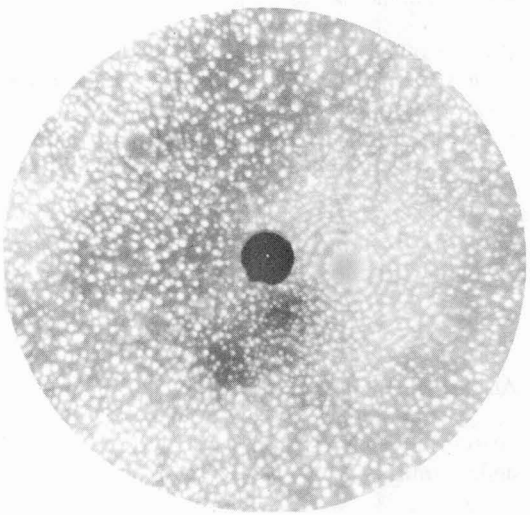

Fig. 4. Neon FIM micrograph of a TiB_2 specimen. Specimen temperature was 50K and BIV was 20 kV.

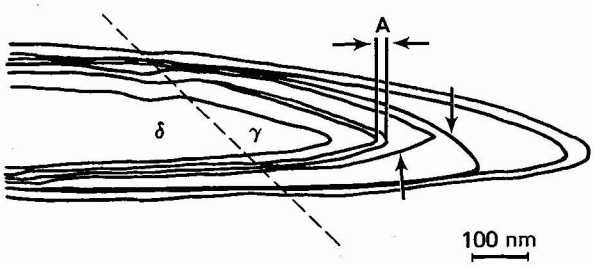

Fig. 5. A sequence of needle shapes observed during precision ion milling a type-308 duplex stainless steel specimen. The step marked 'A' resulted after precision ion milling at 2 kV for 2 min. The specimen outlines marked with arrows were precision ion milled only in the directions arrowed. All other stages were precision ion milled with the beam normal to the page.

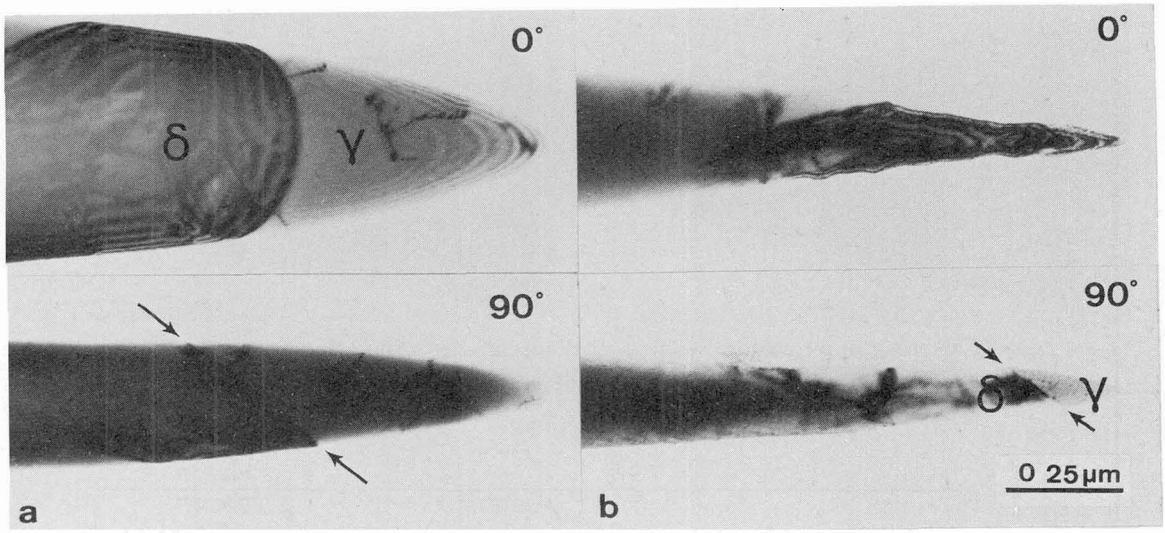

Fig. 6. (a) TEM images, taken from orthogonal directions, of the initial needle shape corresponding to Fig. 5. (b) TEM images of the final needle shape taken at the same orientations as in (a).

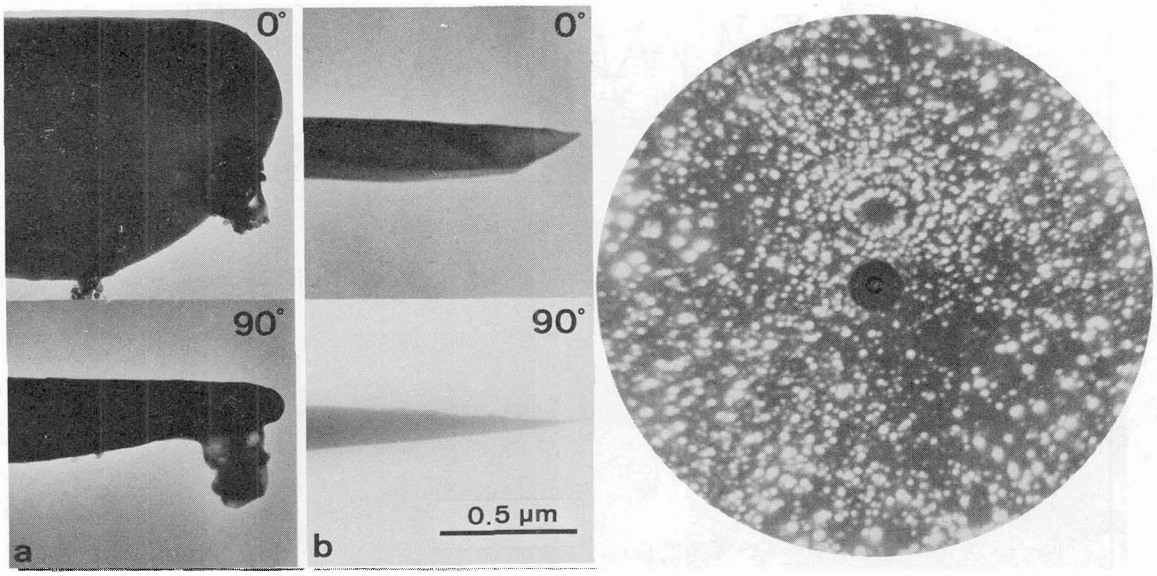

Fig. 7. (a) TEM images, taken from orthogonal directions, of a silicon specimen (a) before and (b) after PIMS. (c) Neon FIM micrograph of the silicon specimen.

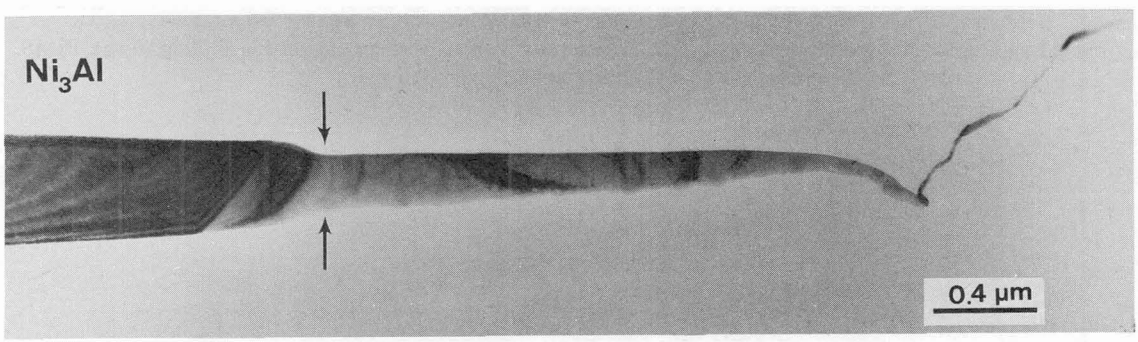

Fig. 8. A filament may form as the result of excessive specimen narrowing.

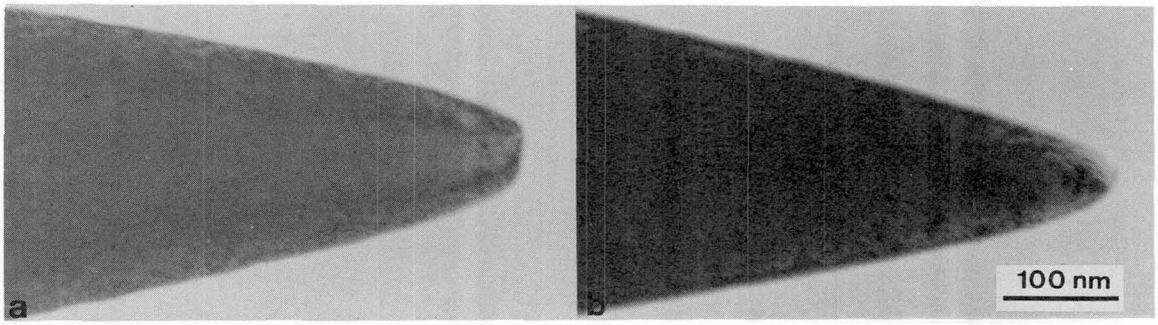

Fig. 9. (a) TEM image of a fractured stainless steel needle after a short duration in the PIMS to remove surface asperities. (b) Same specimen after further precision ion milling.

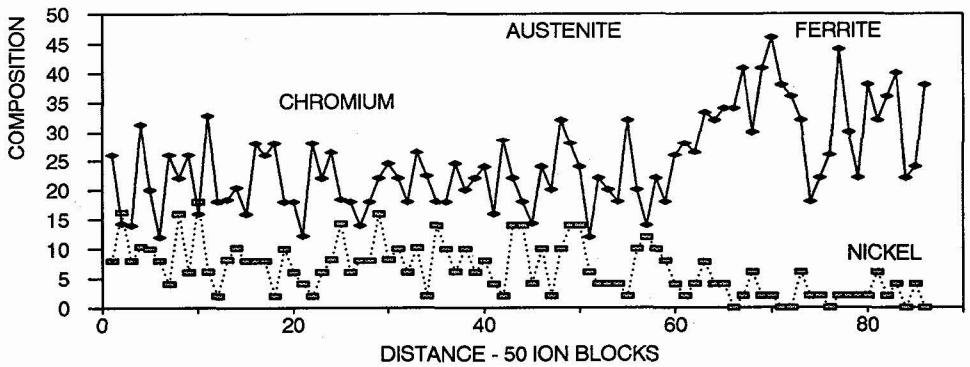

Fig 10. APFIM composition profile of chromium and nickel across the ferrite-austenite boundary shown in Figs. 6(a) and 6(b).

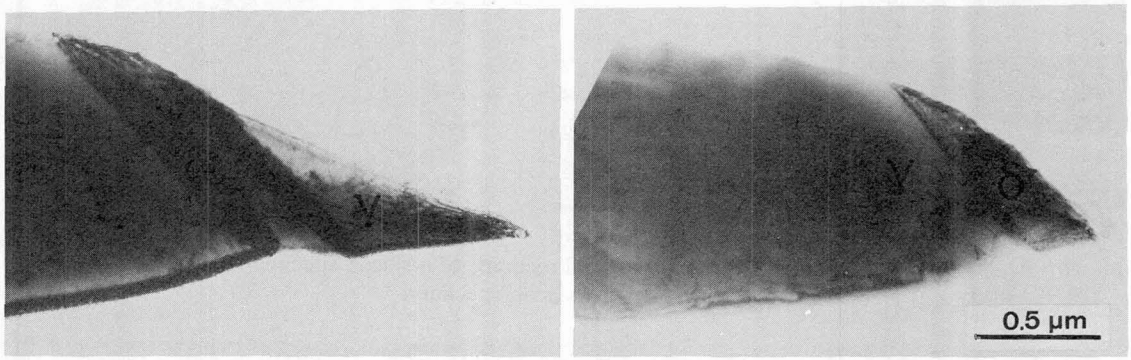

Fig. 11. Duplex phase stainless steel needle at successive stages of precision ion milling in the PIMS. The δ ferrite phase is preferentially retained at the specimen apex.

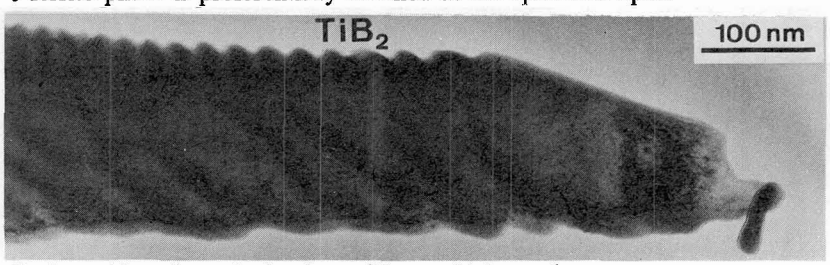

Fig. 12. Surface ridges form during ion milling at higher voltages.