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A Simple Technique for the Preparation of Thin Foils in aluminium and its alloys.

by

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1. INTRODUCTION

Transmission electron microscopy using thin metal foils depends largely upon the success of the techniques used for the preparation of the thin foil. Many methods have been developed successfully for a wide range of materials and these have been discussed in detail elsewhere.[1,2] These methods are as diverse as mechanical polishing, ionic bombardment and electrochemical thinning.

The basis of one of the simplest of these methods is to produce a small sample of the metal and to thin this by a combination of mechanical polishing and electrolytic dissolution. Usually, the attack is continued until a small hole or series of holes appears in the specimen and then the material adjacent to the holes then provides a sample sufficiently thin (i.e. approx. $\frac{1}{4}$ 1000 Å) to allow transmission of the electron beam.

The detailed techniques by which this method has been applied may be summarized as follows:

(a) Preparation of discs. The size of discs required is dictated by the dimensions of the specimen holder in the electron microscope and in the present case this is 2.3 m.m. diam. with a maximum depth of 0.3 m.m.

In general these may be prepared by (i) direct punching (ii) spark machining using a hollow tool or (iii) photo-chemical etching. The type of disc preparation used depends upon the amount of damage done to the disc at this stage and the effect of this on the substructure of the final product. To some extent this depends for example upon the mechanical properties of the material and also the amount of damage that can be tolerated.

(b) Preparation of foils. The essential features of this stage are to produce a central region containing the hole or holes whilst preserving a good polished surface on both sides and avoiding excessive preferential attack on second phase particles, grain boundaries and other structural features.

Several methods have been used: (i) Two stage preparation in which a disc is first dimpled on each side using a flowing electrolyte followed by final piercing in a static electrolytic cell the specimen being immersed. This final process is usually monitored by sighting a light source through the specimen.

(ii) Single stage techniques in which both dimpling and final piercing are accomplished together in the same apparatus as a continuous operation. In this case two opposed jets of electrolyte impinge on the specimen faces and the specimen may or may not be immersed.

In the preparation of foils it is clear that a number of parameters are of importance in the electrolytic processes, including for example (a) composition and temperature

of the electrolyte, (b) current density employed and (c) geometrical factors including the flow conditions associated with the electrolyte jets. A wide variety of techniques and electrolytes have been developed and used for a range of metals and alloys and usually, these are highly specific for a single material. Not infrequently, the same material in a different heat treatment or mechanical condition will not successfully respond to a given method.

The purpose of this note is to describe a flexible technique developed for the preparation of thin foils for transmission electron microscopy from a number of different aluminium alloys in widely varying heat treatment conditions and in different states following fatigue stressing.

2. EXPERIMENTAL WORK

The materials under examination were (i) Hiduminium RR58 (ii) Hiduminium 66 and (iii) Commercial purity Aluminium in varying conditions of heat treatment and before and after fatigue at low and high frequencies. The compositions and heat treatments for these materials are given in Table I.

Experiments to produce thin foils in these materials were conducted using several mechanical/electrolytic techniques but little success was achieved. The principal difficulties arising were:-

(a) Modifications to structure arose, particularly in solution treated and quenched material due to the relatively high temperatures of the electrolytes. It is clearly desirable therefore to use a method which prevents any heating of the sample during preparation.

(b) Edge attack during thinning was prevalent in some designs of specimen holder and it was desirable to eliminate this effect.

(c) Most techniques were found to be very sensitive to slight changes in operating conditions such as applied voltage, and current density, and whilst success might be achieved with one specimen, a different sample would show preferential attack. Since a wide range of materials was being examined it was clearly desirable to have less sensitivity to the operating conditions.

(d) The two-stage method of dimpling followed by final piercing was rather slow and the final step in a static cell resulted in problems of temperature monitoring and control and difficulties due to gas evolution at the specimen.

A single-stage jet technique seemed likely to alleviate these effects.

The experimental method developed is as follows:-

(a) A sample is taken from the bulk material (usually a fatigue specimen) by slitting off a disc shaped piece 1-2 m.m. in

thickness, a copious flow of coolant being applied during this process. The disc is then mounted in a simple holder and mechanically polished through the standard range of emery papers to 600 grade, the final thickness being in general 0.25-0.30 m.m. Discs are cut by spark machining and by punching but in the former process it was found impossible to achieve a circular shape and the surface of the disc was frequently severly damaged by stray sparking. These difficulties could without doubt have been overcome by careful tool design, the provision of clean flowing dielectric and by selection of the electrical parameters but it was not thought valuable at this stage to attempt to optimize this process. It was therefore decided to use the simple punching method only. This consists of a flat die with a slightly hollow faced punch mounted in a guide, the punch cutting the periphery of the disc and avoiding contact with the central region of the disc.

(b) The discs were electrolytically thinned and holed using a single stage twin jet process without immersion. The general arrangement of the apparatus is shown in Figure I. The jet unit is gravity fed by a reservoir, the electrolyte passing through a cooling coil immersed in a solid carbon dioxide/methylated spirit mixture in which the temperature can be varied from ambient to -65°C. After passage through the jets the electrolyte is returned to the reservoir by a peristaltic pump.

Figure 2 shows the construction of the jet unit which consists basically of two horizontally opposed jets whose separation can be varied. A T-piece one end sealed with a plane glass window was provided for each jet and the platinium wire cathodes are introduced in the vertical section of the tubes. The flow to each jet can be varied independently by means of a screw pinch-clip. A lamp was placed behind one jet window and viewed through the other window.

The specimen holder, which can be mounted between the jets is made from P.T.F.E. and consists simply of a sheet which has been partially slit and folded so that a hinged double thickness results. The holes for the specimen and clamping bolts were then drilled and a platinium wire anode ring fitted around the specimen hole. The hole exposing the specimen is countersunk to improve the electrolyte flow conditions. 6BA nylon nuts and bolts are used for clamping the hinged holes. The holder is shown diagrammatically in Figure 3. Earlier designs using polyethylene were found to have a short life due to poor sealing around the specimen periphery which also gave rise to edge attack. This arose from deformation of the polyethylene under the clamping loads. P.T.F.E. is better in this respect and one holder has been found to produce more than 150 foils satisfactorily without sign of wear or deterioration. In principle this holder is not unlike that described by Dewey and Lewis[3] for an immersion method but it has the advantage of greater simplicity of construction and operation and considerably lower cost.

The apparatus is easy to set up and in particular the positioning of the specimen holder is not critical, so long as it is approximately midway between the jets and aligned normally. The jet/specimen distance is also not very critical and is determined mainly by the current density required and the viscosity of the electrolyte. In general the distance between jets is about 8 m.m.

Most electrolytes described as suitable for aluminium and its alloys are based on either orthophosphoric acid or perchloric acid. In some cases temperatures up to 70°C are required and clearly this would be unsatisfactory for the solution treated samples during foil preparation. Other electrolytes have their temperatures specified only as for example < 18°C with lower temperatures suggested for minimizing etching effects[1].

Several electrolytes were used and the best results are found with a composition reported by Dewey and Dennis[1] of 40% acetic acid, 30% phosphoric acid, 20% nitric acid, 10% water. Whilst this composition was specified for use with aluminium and Al-Cu-Mg-Si alloys, it was found applicable to all three materials used in all conditions provided that the electrolyte temperature at the jet was in the range -12°C to -5°C. At lower temperatures the electrolyte becomes too viscous for free flow in the jets and at higher temperatures there is danger of etching.

The current density used is 4.2 to 4.6 Amp.cm.$^{-2}$ which is generally rather higher than that quoted for these methods. The voltage applied is dependent upon the geometry of the cell and the temperature of the electrolyte and is in general around 105 to 110 volts.

The flow through the jets is adjusted by the pinch clips until contact with the specimen is just made and no further control is necessary during thinning. The time required for penetration is 3 to 4½ minutes the variation arising from variations of initial specimen thickness. Penetration is seen by viewing the light source through the specimen and holes of < 100μ diameter can readily be detected.

After thorough washing in absolute alcohol and drying the specimen was ready for examination in the electron microscope. All specimens were mounted in a Valdre tilting cartridge and examined in a Siemens Elmiscop I electron microscope operated at 100 kV.

Specimens for examination can be stored successfully for periods up to 2 or 3 days in abolsute alcohol without deterioration but after longer periods of storage some deterioration mainly due to surface contamination is found.

3. RESULTS AND DISCUSSION

The technique described has been found to produce satisfactory thin foils with large central thinned regions. The method is simple, rapid, inexpensive, and gives a very high fraction of successful foils for all the materials in all of the conditions used.

Some features frequently associated with these preparative techniques, such as etching, are absent and no modification of the substructure arises at the low temperatures used except that inherent in the thinning process associated with the relaxation of dislocation arrangements in the thin foil.

Figures 4 to 10 show typical photographs of regions in several materials. The features seen have been tentatively indentified in comparison with work published on similar alloys[4,5]. It will be clear from these photographs that no preferential attack or loss of second phase particles has taken place.

The features which distinguish the technique are the use of fairly intense cooling together with jets which are not immersed. The current density used is also higher than generally used and it is thought probable that polishing is taking place at a current density where pitting associated with bubble formation would normally arise. However, the jets of electrolyte presumably sweep these bubbles away together with the other anodic products. The heating effect of the current is offset by cooling the input to the jets to such an extent that the rise in temperature between the cathode and anode never gets to a level where etching might take place.

No evidence is available on the amount of damage produced by the disc preparation but it is clear that a good spark machining process or photochemical etching method would be preferable to the punching system used in these experiments. Work is currently being undertaken to make use of the photochemical etching method.

4. CONCLUSIONS

1. A single stage electrolytic jet polishing technique has been successfully developed.

2. The method is versatile and has been applied to widely different aluminium alloys in a variety of states.

3. Satisfactory foils with large central thinned areas are produced rapidly and simply.

REFERENCES

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TABLE I

1. Hiduminium R.R 58. - Extruded rod.

   Composition - Cu 2.5 Mg. 1.5, Fe 1.0, Ni 1.2, Ti 0.1 Bal Al.
   Solution treatment - 2½ hours at 530°C - 10 second air cool - water quench.
   Aging - 10 hours at 190°C.

2. Hiduminium 66. (2L65) - Extruded rod.

   Composition - Cu 4.4, Si 0.7, Mn 0.7, Mg 0.7, Bal Al.
   Solution treatment - 2½ hours at 505°C - water quench.
   Aging - 5 hours at 180°C.
   Overaging - 120 hours at 180°C.

3. Commercial purity aluminium - Extruded rod.

   99.0% Al.
   Annealed condition. ½ hour at 300°C.

Fatigue experiments were conducted at 50 Hz and 20 kHz.

FIGURE 1: General arrangement of apparatus.

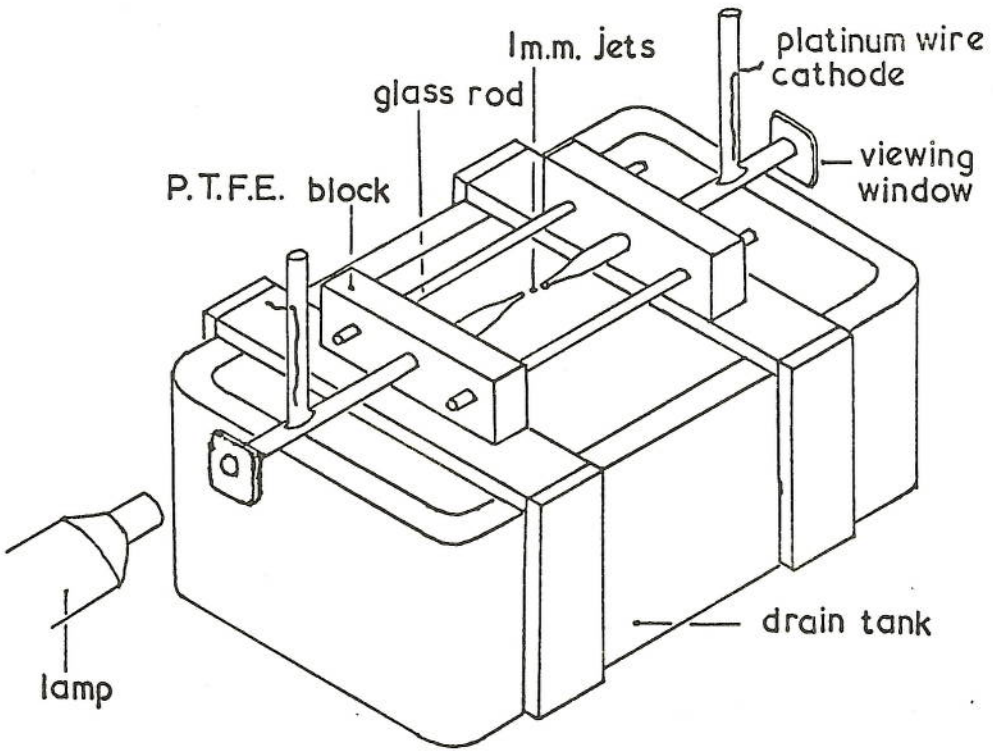

FIGURE 2: The jet unit.

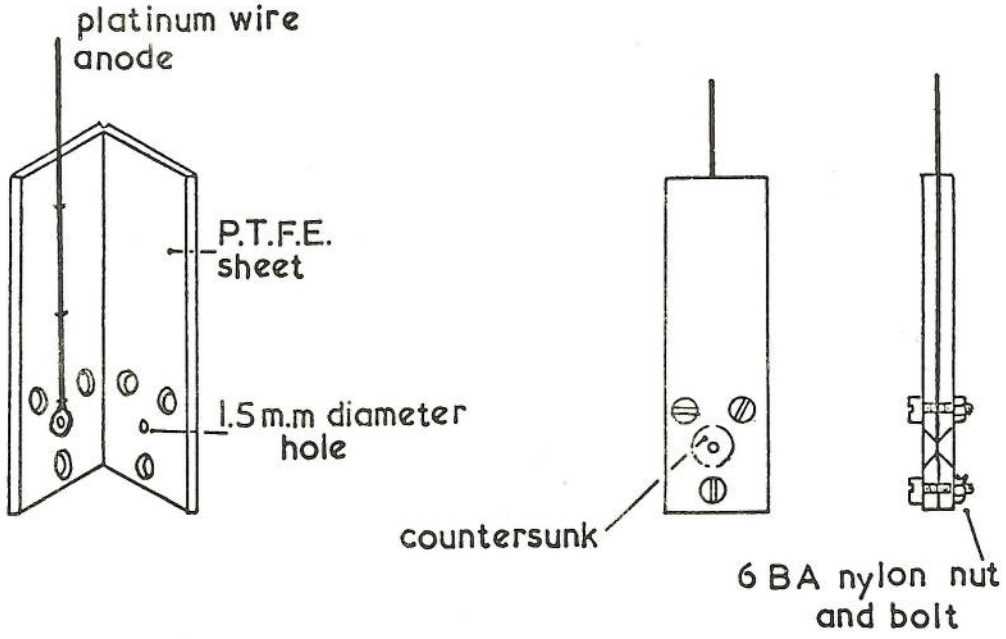

FIGURE 3: The specimen holder.

FIG.4: RR58: Optimum aged. Fatigued at 20kHz in water environment showing bands of dislocations (3.8×10^6 cycles at $\pm$ 12.6 tsi).

FIG.5: Commercial-purity aluminium. Fatigued at 20kHz showing sub-boundaries ($>5 \times 10^8$ at $\pm$ 2 tsi).

FIG.6: L65: Solution treated showing helical dislocations and undissolved second phase particles.

FIG.7: L65: Optimum aged showing θ'' and dislocation-nucleated θ'.

FIG.8: L65: Overaged showing θ", θ' and θ precipitates.

FIG.9: L65: Solution treated. Fatigued at 50 Hz showing high concentration of oriented helical dislocations (1.2×10^6 cycles at ± 10 tsi).

FIG.10: L65: Overaged. Fatigued at 20 kHz showing θ", θ' and θ precipitates (0.97×10^6 cycles at ± 10.5 tsi)