March 25, 1969 R. C. GLENN 3,434,956

APPARATUS FOR THE ELECTROLYTIC THINNING OF METALLIC SPECIMENS FOR TRANSMISSION ELECTRON MICROSCOPY

Filed June 9, 1965 Sheet 1 of 4

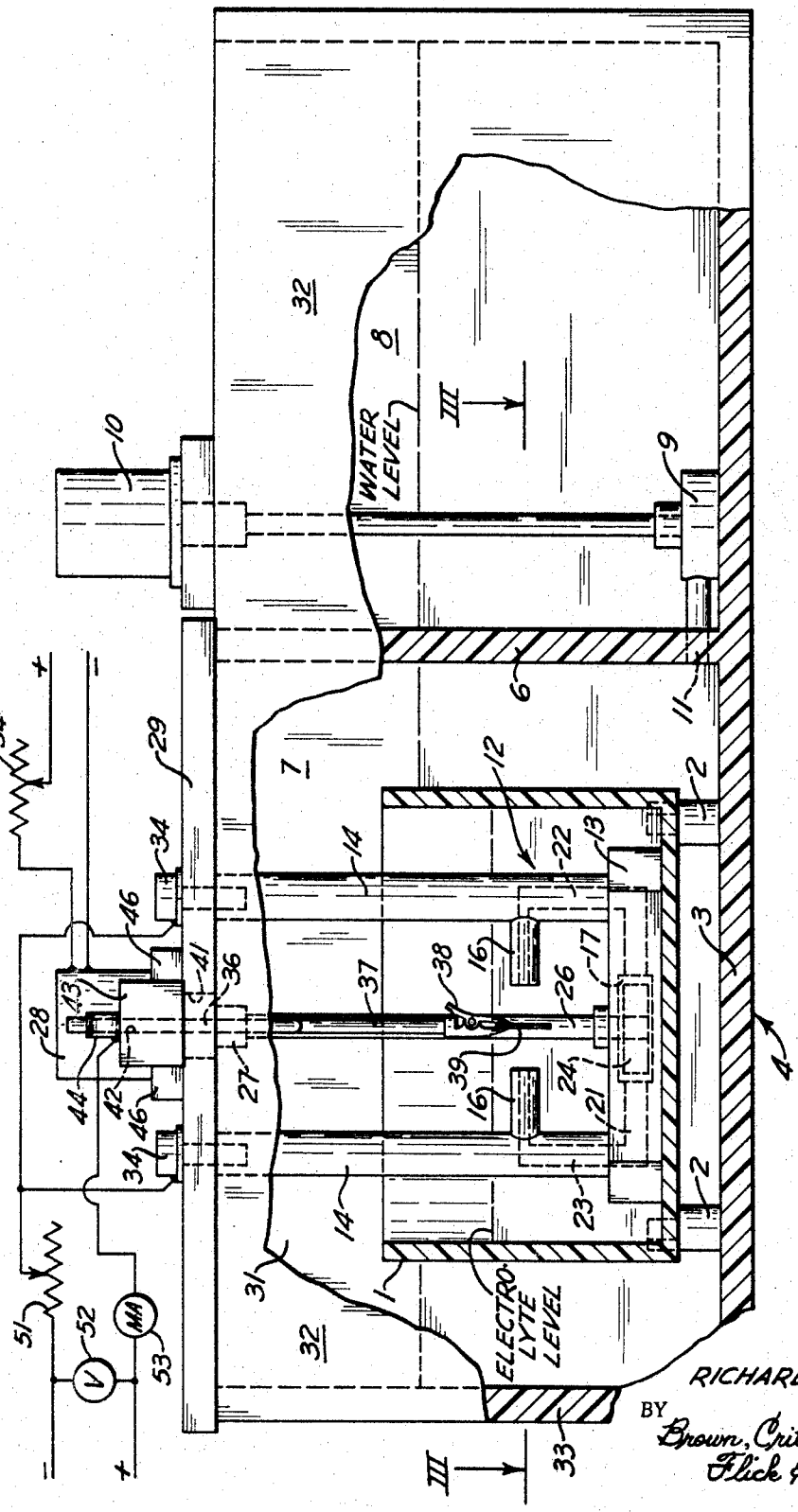


Fig. 1

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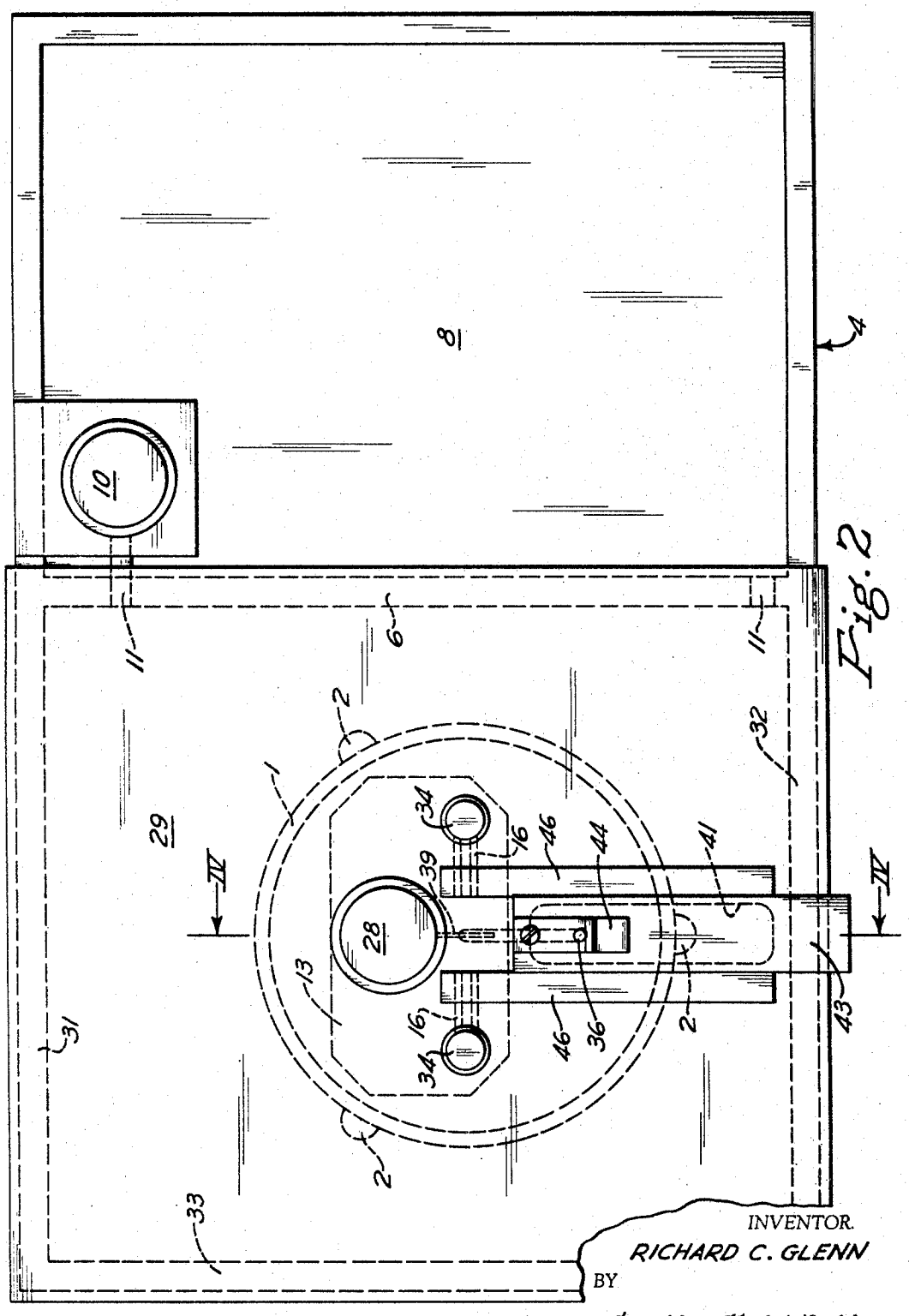


Fig. 2

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Fig. 4

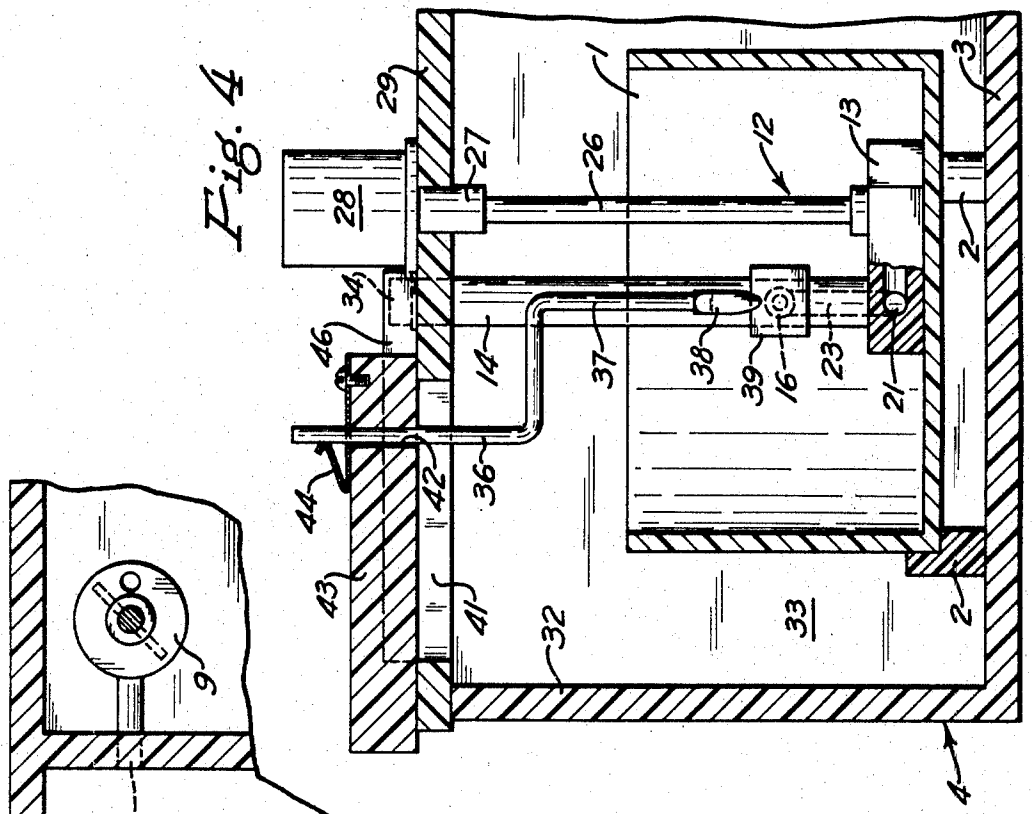
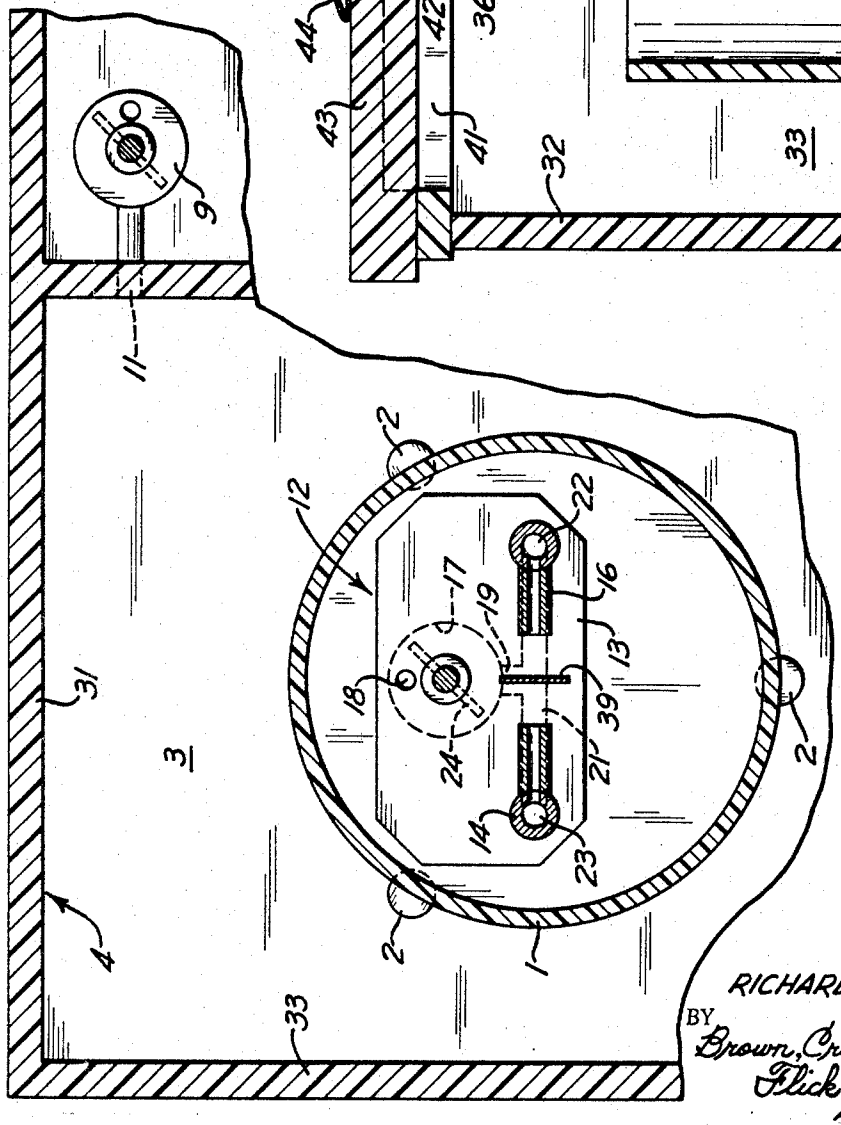


Fig. 3



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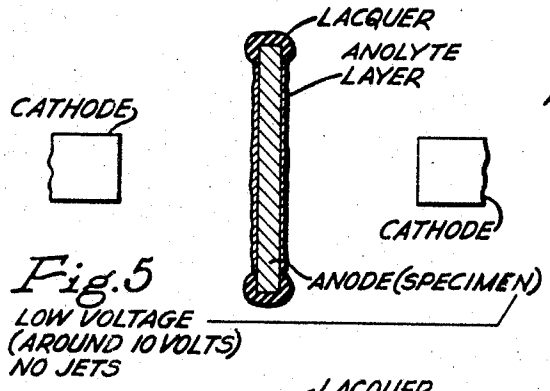


Fig. 5

LOW VOLTAGE (AROUND 10 VOLTS) NO JETS

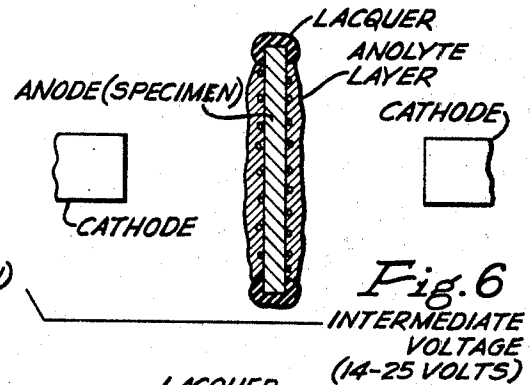


Fig. 6

INTERMEDIATE VOLTAGE (14-25 VOLTS) NO JETS

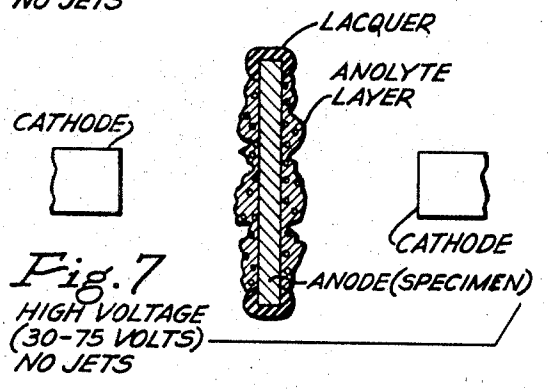


Fig. 7

HIGH VOLTAGE (30-75 VOLTS) NO JETS

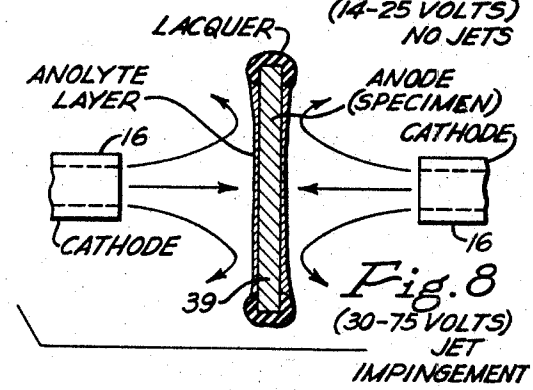


Fig. 8

(30-75 VOLTS) JET IMPINGEMENT

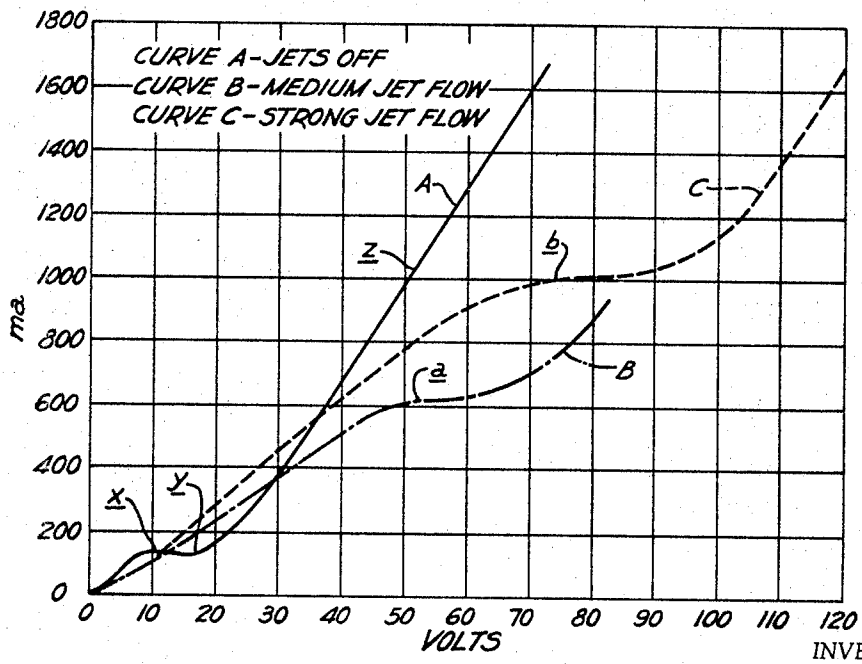


Fig. 9

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3,434,956

APPARATUS FOR THE ELECTROLYTIC THINNING OF METALLIC SPECIMENS FOR TRANSMISSION ELECTRON MICROSCOPY

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Int. Cl. B23p 1/04; C23b 3/06
U.S. Cl. 204—237 8 Claims

ABSTRACT OF THE DISCLOSURE

This invention relates generally to the preparation of metallic specimens for use in transmission electron microscopy and, more particularly, to the final thinning of such specimens by electrolytic reduction to a thickness of less than 0.5 micron over an area of at least 750 square microns. The reduction is effected by supporting the specimen entirely immersed in an anolyte-forming electrolyte and circulating the electrolyte at a predetermined velocity through a pair of jet nozzles disposed on opposite sides of the specimen and by passing an electric current of a predetermined applied voltage through the electrolyte and specimen. At least one of said predetermined parameters is adjusted to form at the electrolyte-specimen interface an anolyte layer of controlled thickness characterized by the electrical resistance of said layer having a value at the lower end of the range over which the resistance increases substantially at the same rate as the applied voltage, while maintaining the applied voltage substantially above the level at which pitting of the specimen would normally occur in the absence of jet stream circulation of the electrolyte.

Metallic specimens suitable for transmission electron microscopy are desirably in the form of exceedingly thin foils that are completely free not only of so-called "artifacts," such as oxide particles formed on the specimen surface during electrolytic reduction, but also of etching and pitting. "Etching," as used herein, refers to the preferential electrolytic attack of the specimen surface caused by grain boundaries, the crystal orientations of different grains, or the presence of different phases, such as precipitates, in the matrix of the specimen. "Pitting," as used herein, refers to the preferential electrolytic attack of the specimen surface caused by the presence, even for short intervals, of gas bubbles on that surface. The term "electropolishing," on the other hand, is used herein to characterize the electrolytic reduction of metallic specimens without etching or pitting and without the creation of artifacts. In addition, an acceptable quality foil should contain areas totalling at least 750 square microns, and preferably at least 15,000 square microns, that are free of the above defects and thin enough (less than 2,000 angstrom units for iron and iron-base alloys) to be suitable for electron transmission microscopy.

Among the electrolytes commonly used for preparing microscopy specimens of the type herein referred to are those of the so-called limiting-current-density types, such as well-known chronic-acetic electrolytes. With these electrolytes, a viscous high resistance layer, often called an "anolyte" layer, begins to form at the anode (specimen) when the applied voltage is increased to a certain point; and further increases in the voltage produce little or no change in the current through the system, reflecting increasing thickness and greater resistance of the anolyte layer, until the voltage reaches the point at which sufficient gas evolution occurs to break up the anolyte layer. The presence and thickness of that layer are known to have considerable effect on the kind of electrolytic reduction

that occurs. Below the voltage at which the anolyte layer begins to form, there is etching of the specimen. At somewhat higher voltages where gas evolution begins, even though characterized by the formation of very small bubbles (sometimes referred to as "micro-bubbles") that do not destroy the anolyte layer, there is pitting of the specimen. In between those two voltage levels, that is, just above the etching range but below the pitting range, is generally the optimum voltage range for electropolishing specimens to thin them for transmission electron microscopy. At very much higher voltages, characterized by rapid gas evolution that disrupts the anolyte layer, there is random pitting of the specimen; and, although there may also be randomly localized areas that are electropolished, their occurrence is so unpredictable that they do not provide a practicable yield of suitable specimens.

Accordingly, it has heretofore been conventional practice in the electropolishing of metal specimens to use relatively low voltages (for example, around 10 volts) to keep within the optimum voltage range mentioned above. A great disadvantage, however, of electropolishing at such relatively low voltage has been the length of time required to reduce a specimen from the usual initial thickness of around 3 mils to the ultimate foil thickness of less than 2,000 angstrom units, for iron or iron-base alloys. Generally 5 hours or more have been required for such reduction.

It is among the objects of the present invention to provide apparatus and procedures for use in the electrolytic thinning of metallic specimens, in which the thinning will provide a high yield of acceptable areas suitable for electron transmission microscopy and in which the thinning is carried out by electropolishing in a fraction of the time formerly required by conventional methods of electropolishing.

Other objects will be apparent from the following description of a preferred embodiment of this invention, in connection with the attached drawings, in which:

FIG. 1 is a somewhat diagrammatic front elevation, partly in section, of apparatus suitable for carrying out the present invention, including a schematic arrangement of certain electrical circuit elements;

FIG. 2 is a plan view of the apparatus of FIG. 1;

FIG. 3 is a horizontal section along the line III—III of FIG. 1;

FIG. 4 is a vertical section along the line IV—IV of FIG. 2;

FIGS. 5, 6, 7 and 8 show diagrammatically various configurations of the anolyte layer surrounding the anode (specimen) under different operating conditions; and

FIG. 9 shows graphically the relationship between current and voltage in the electrolytic reduction of iron and iron-base alloys under different operating conditions.

The present invention is predicated on the discovery that the desirable electropolishing results that are obtained by electrolytic reduction in an anolyte-forming electrolyte under the low voltage conditions described above can also be obtained with the same electrolyte at very much higher voltages, with a consequent saving in the time required, by circulating the electrolyte in jet streams against opposite faces of the specimen at a predetermined velocity determined by the applied voltage to form at the electrolyte-specimen interface an anolyte layer of controlled thickness that will permit electrolytic reduction of the specimen without etching and without pitting.

In accordance with this invention, the specimen to be electropolished is supported in an electrolyte of the anolyte-forming type between and substantially equidistant from two opposed jet nozzles. The electrolyte is circulated through those nozzles at some predetermined velocity to provide subtantially coaxial jet electrolyte

streams that impinge on opposite faces of the specimen. At the same time, an electric current is passed through the electrolyte and specimen by the application of a predetermined voltage. At least one of those parameters, that is, either the velocity of the jet streams or the magnitude of the applied voltage, is adjusted to avoid both etching and pitting of the specimen, while at the same time maintaining the applied voltage substantially above that at which rapid gas evolution, and consequent random and unpredictable pitting, would occur at the surface of the specimen in the absence of jet stream circulation of the electrolyte.

The invention is described herein with reference to the thinning of iron or iron-base alloy specimens, using one of the well-known electrolytes of the anolyte-forming type. One of the more suitable electrolytes of this type consists of one part of anhydrous sodium chromate and five parts of glacial acetic acid. Polishing reactions with this electrolyte are similar to those occurring with standard chromic-acetic acid electrolytes using chromium trioxide as the polishing agent; but the solution here recommended does not appear to be as critically dependent on temperature and voltage as the standard solution. It should be understood, however, that the invention is equally applicable to the thinning of other metallic specimens, and to the use of various other well known anolyte-forming electrolytes of the types generally used for the electrolytic reduction of the particular metals involved.

Referring to the drawings, the electropolishing unit includes a container 1, in the form of a cylindrical dish, which is removably mounted on short notched posts 2 above the bottom 3 of a tank 4. The tank is divided by a partition 6 into two compartments 7 and 8, which are partially filled with water (not shown) to the level indicated below the top of container 1. A pump 9, driven by a motor 10, circulates the water in both compartments through passages 11 in partition 6, for controlling the temperature of the above-described electrolyte (not shown) in container 1. Ice (not shown) may be added as desired to the water in compartment 8. The tank and container may be mode of a suitable plastic, such as Plexiglas.

Removably supported inside container 1 is a pump-cathode assembly 12. It includes a base member 13, which may be made of a suitable plastic, such as Teflon; vertical posts 14 of stainless steel mounted on the base; and stainless steel cathode nozzles 16 supported by the posts. Inside the base is a pump chamber 17, which is provided with an inlet 18 for admitting the electrolyte into the chamber and with an outlet 19 for discharging the electrolyte by way of a conduit 21 and interior passages 22 and 23 to the cathode nozzles 16. These nozzles are mounted so that their outlet orifices are in substantially coaxial opposition just below the level of the electrolyte in container 1. A stainless steel impeller 24 is rotatably mounted in pump chamber 17 and is connected by a shaft 26 and a connector 27 to a D.C. motor 28. This motor is mounted on a lid 29, which may also be of plastic and may be limited to covering compartment 7. For convenience, the lid may be hinged on the back wall 31 of that compartment and additionally supported by front wall 32 and side wall 33 and partition 6. Preferably, the entire pump-cathode assembly 12 also is suspended from lid 29 by thumbscrews 34 threaded into the upper ends of posts 14. A stainless steel specimen holder 36 is also supported by lid 29. The holder is in the form of a thin rod, having an offset lower portion 37, which ends in a spring clip 38 for gripping a metaliic specimen 39. The upper end of the holder passes through a slot 41 in lid 29 and through a hole 42 in a plastic block 43, where the rod is held for vertical and rotary adjustment. A leaf spring 44 mounted on block 43 presses against the side of the rod to hold it in a predetermined position. Block 43 is slidably supported between guide strips 46, for adjusting holder 36 lengthwise of slot 41. When the lid is opened, the entire anode-cathode assembly swings out of the container for easy inspection and cleaning.

The specimen itself should be about 3 mils thick, preferably of rectangular shape and about 7 x 10 mm. in size. It has been found that substantially larger specimens involve higher currents, which provide less uniform current distribution and cause excessive heating and rapid decomposition of the electrolyte. These latter conditions are undesirable, because they reduce control over etching and pitting of the specimen. For convenience, the specimen is held with its longer edges horizontal. Clip 38 and holder 36 are coated with lacquer (not shown) far enough up to insure that the electrolyte will not contact the bare metal; and, in accordance with usual practice, the entire perimeter of the specimen is preferably coated with lacquer (see FIGS. 5–8) for about 1 mm. in from the edges to avoid concentrations of current at the edges. The specimen is then suspended in the electrolyte with its opposite faces between and substantially equidistant from the two opposed jet nozzles 16, the axes of those nozzles being substantially normal to the specimen faces and substantially centered theeron, with the tips of the nozzles about 1½ inches apart.

The specimen 39 (as the anode) and the nozzles 16 (as the cathode) are connected through leaf spring 44 and thumb screw 34, respectively, to a source of direct current (not shown). A suitable voltage regulator, such as a variable resistance 51, is included in this circuit for adjusting the voltage applied to the system. A voltmeter 52 and a milliammeter 53 are also included in the circuit. Pump motor 28 is connected in a separate electrical circuit, and its speed is controlled by an adjustable resistance 54 in series with a source of direct current (not shown). Accordingly, when impeller 24 is rotated, electrolyte will be drawn through pump inlet 18 and discharged through nozzles 16 in substantially coaxial jet streams of adjustable velocity.

In operating the unit, a starting voltage of between 50 and 75 volts is applied across the cathode and anode, and the initial current passing through the system is measured. Pump motor 28 is then turned on and its speed increased until the electrolyte is circulating through the jet nozzles at a velocity that produces a maximum current drop from the initial current reading, reflecting the formation of an anolyte layer about the specimen. This gives the approximately correct electrolyte jet velocity relative to the chosen initial voltage.

Before proceeding further, the specimen should be removed from the electrolyte and visually checked. If it has been subjected to etching, it will have a gray appearance showing the crystal structure. This means that either the voltage is too low or the jet velocity is too high. On the other hand, if the specimen has a blue-haze appearance, it indicates micropitting (i.e., pitting resulting from the formation of adhering microbubbles) and means that either the voltage is too high or the jet velocity is too low. In either case, adjustment of one or both of those parameters may be made, and the specimen again checked visually.

Once suitable operating conditions have been established, substantial electropolishing or thinning of the specimen may proceed. Because the jet streams are substantially coaxial with the center of the specimen, there will be slight preferential electropolishing in the central area as compared to the marginal areas of the specimen. This is desirable, because the electropolishing proceeds until there has been perforation of the specimen at the center, and the areas of least thickness will then be adjacent the center. However, in order to obtain as much thinned area as possible suitable for electron transmission microscopy, the specimen should be attacked preferentially in at least two different central areas, which may be done by shifting it slightly lengthwise back and forth several times (by reciprocating block 43 on lid 29), and

then leaving it centered until it is finally perforated, which will usually occur in two adjacent places that merge into an irregular opening. As a final "touch," after perforation, it is desirable to lower the voltage and reduce the jet flow of the electrolyte, since at higher voltages and jet velocities thin areas may polish away and disappear completely in a few seconds. Depending on the material being thinned, the voltage may be reduced to as low as 10 volts and the jet velocity reduced so that impeller 24 is barely turning, as in the case of the final touch for annealed, high purity iron, or the voltage reduced only to 30 volts with moderate jet flow, as in the case of the final touch for cold-rolled, medium-carbon steel.

After thinning has been completed, the specimen should be removed from the electrolyte, immediately rinsed in acetic acid, and then immersed in methanol while the lacquer is mechanically removed. The specimen should then be rinsed again in clean methanol and placed between two sheets of lens paper to absorb the excess liquid, and finally air dried. It is then ready for mounting as a microscope specimen.

FIGS. 5–8 show approximate configurations of the anolyte layer under the different conditions indicated. FIG. 5 shows the anolyte layer under conditions found in certain conventional electropolishing methods, in which the applied voltage is around 10 volts and there is no jet flow. These conditions correspond to those at point *x* on curve A of FIG. 9, showing the relationship between the current and applied voltage in the electrolyte system. In this curve, the plateau adjacent point *x* reflects the formation of the anolyte layer and its increasing thickness and increasing electrical resistance because as the voltage is increased from about 10 to 20 volts, there is no or very little corresponding increase in the current. It will be noted that in FIG. 5, the anolyte layer is relatively thin and is uniformly distributed over the specimen surface.

FIG. 6 shows the increased thickness of the anolyte layer at slightly higher voltages, again without jet flow of the electrolyte. Because of the higher voltage, gas evolution has begun in the form of microbubbles at the electrolyte-specimen interface, which tend to adhere to the specimen and thereby cause pitting of its surface. The conditions here described are characteristic of point *y* on curve A of FIG. 9.

FIG. 7 is an attempt to approximate the momentary appearance of the anolyte layer under the dynamic conditions at the electrolyte-specimen interface, again without jet flow of the electrolyte, at still higher voltages than those prevailing in FIGS. 5 and 6. Gas evolution has now become very rapid and it continually modifies and disrupts the anolyte layer. The conjectural representation of the shape of that layer as shown in FIG. 7 is based on observed effects on the specimen, when subjected to the indicated conditions; these effects include random pitting of the specimen surface due to the tendency of some gas to adhere to that surface. The disrupted anolyte layer of FIG. 7 would occur, for example, at point *z* on curve A of FIG. 9. It will be noted that because of the continual disruption of the anolyte layer, its electrical resistance no longer increases, and the current varies substantially linearly with the applied voltage as it does in the very low voltage ranges below the plateau in curve A.

FIG. 8 shows the effects on the anolyte layer of jet flow under the same high voltage conditions as in FIG. 7. It will be noted that impingement of the electrolyte on the specimen under conditions of jet flow has thinned the anolyte layer to substantially the same thickness as that shown in FIG. 5, with some preferential thinning in the central area relative to the marginal areas of the specimen. But the layer is symmetrically uniform in thickness. In contrast to the conditions shown in FIGS. 6 and 7, there is in FIG. 8 a complete absence of adhering gas bubbles and, of course, no disruption of the anolyte layer. At around 50 volts with the proper jet velocity, the conditions shown in FIG. 8 will correspond to those at point *a* on curve B of FIG. 9. At 75 volts with a higher jet velocity, the same conditions will correspond to those at point *b* on curve C. In each case, the thickness of the anolyte layer will be such that there will be no etching or pitting of the specimen surface. Nor will that surface be marred by the presence of artifacts.

It has been found that satisfactory electropolishing of iron and iron-base alloys can be obtained at voltages at least as high as 90 volts by increasing the jet velocity of the chromic-acetic electrolyte impinging on opposite faces of the specimen so as to produce and maintain a relatively thin anolyte layer at the electrolyte-specimen interface. The apparent effect of the impinging electrolyte is to wash away the old anolyte layer as a new layer is formed and, as the voltage is increased, to raise the plateau on the current-voltage curve and to decrease the slope of that curve. At voltages higher than 90 volts, the curve tends to flatten out and the plateau thereon to disappear, reflecting the absence of a uniform anolyte layer. It will be apparent from the foregoing description, however, that an anolyte layer can be formed and its thickness controlled at specific voltages over a wide voltage range by proper correlation of the voltage with the velocity of the electrolyte jet streams impinging on opposite faces of the specimen. As a result, it is possible to maintain ideal electropolishing conditions (which are generally recognized as occurring at the beginning of the plateau on the current-voltage curve) at very much higher applied voltages than is possible in the absence of jet flow of the electrolyte.

Although the present invention is directed to the novel and critical relationship between (1) the applied voltage and (2) the jet velocity of the anolyte-forming electrolyte at the specimen surface, it will of course be understood that the temperature of the electrolyte is also an important factor, but such a well-known one that it has not been emphasized herein. When using the chromic-acetic electrolyte identified above as a preferred electrolyte, it is desirable to hold its temperature constant at about 20° C. to obtain the best results. It should be pointed out that the electropolishing unit herein described is designed so that the three variables of voltage, jet velocity, and temperature may be easily adjusted relative to each other to obtain the best electropolishing results in the least time. It is difficult to appreciate the absolute perfection required in electropolishing specimens for transmission electron microscopy until it is realized that they are subjected to magnifications of up to 100,000 diameters. At such magnifications, the pitting of the specimen surface caused by the formation of microbubbles of gas adhering to that surface during its electrolytic reduction will appear either as closely spaced holes or as mottled areas. Accordingly, uniform, nonpreferential surface reduction is an essential prerequisite of any electropolishing process that can expect to meet the exacting standards required of specimens for electron transmission microscopy.

It is among the advantages of the present invention that it permits the above standards to be met with a very substantial saving in time over that required under present conventional practice, which is characterized by relatively low applied voltages and the absence of jet electrolyte impingement on the specimen. For example, the time required to thin a specimen of iron, or iron-base alloy, from 3 mils to less than 2,000 angstrom units is reduced from more than 5 hours required under such conventional practice to 30 minutes or less under conditions of high voltage and controlled electrolytic jet impingement as described herein. This rapid thinning is obtained without creating artifacts on the specimen surface and without pitting or etching of that surface. It is obtained over a preferred voltage range of from 30–90 volts, using the preferred sodium chromate-acetic acid electrolyte herein described. It is applicable to electropolishing not only iron or iron-base alloys but also other

alloys including copper-aluminum, cobalt, and nickel alloys, with comparable results. These results are also obtainable with the same alloys using the standard chromium trioxide-acetic acid electrolyte over the same preferred voltage range.

The invention may be practiced with any one of many other well-known, anolyte-forming electrolytes, such as, for example, a 20 percent mixture of perchloric acid in acetic acid, which is suitable for electropolishing iron and iron-base alloys. In such case, the recommended voltage range would be from about 15 to 20 volts, which is substantially higher than the permissible voltage for electropolishing with the same electrolyte in the absence of electrolytic jet flow, and results in very rapid (5 or 10 minutes) thinning of the specimen from the initial to the final thickness previously mentioned. Similar results are obtained with an anolyte-forming electrolyte consisting of a 20 percent mixture of perchloric acid in ethyl alcohol, which is also suitable for iron and iron-base alloys. The temperature of the electrolytes referred to in this paragraph is preferably kept at about 20° C.

It is even possible to electrolytically thin silver and many of its alloys in accordance with this invention to produce acceptable specimens for electron transmission microscopy, using an anolyte-forming electrolyte containing 5 gms. of potassium cyanide per 100 cc. of water and applying a voltage of not more than 5 volts, while keeping the electrolyte at the relatively low temperature of around 5° C. The electropolishing proceeds very rapidly, taking only about 40 seconds to reduce a specimen from an initial thickness of 3 mils to a thickness suitable for electron transmission microscopy.

The present invention can be practiced with considerable latitude in the dimensions and separation of the jet nozzles. For most purposes, it is preferable to have a ratio of approximately 1 to 4 between the cross sectional area of the jet nozzle orifices and the exposed area of the specimen, but that ratio can vary from as high as 2 to 1 to as low as 1 to 8. Beyond the stated limits, there is a tendency for preferential attack of the specimen and consequent etching or pitting. For example, with a ratio of 1 to 10 between the cross sectional area of jet nozzle orifices and the exposed area of the specimen, the specimen tends to be etched in the central impingement area and to be pitted in the marginal area. The absolute size of the jet orifices is preferably around 0.32 cm. (⅛ of an inch) in diameter, and preferably not more than 0.48 cm. (3/16 of an inch) in diameter. Larger orifices would require larger specimens to maintain the desired area ratio, with the result that the electrolyte tends to be heated locally, changing the conditions of the electrolytic reaction and reducing the probability of obtaining a satisfactory specimen. In addition, the orifices should preferably be separated from each other by at least 1 cm. and not more than 6 cm. Beyond the stated limits, there is the same tendency towards preferential attack noted above where area limits are exceeded. For most purposes, a separation of 3.75 cm., or 1½ inches, works well. It is important that both sides of the specimen be subjected to the same conditions of jet impingement, since any inconsistent attack may produce etching or pitting on one side that will be apparent on the projected image of the specimen. It will be understood of course that the composition of the electrolyte and the composition and size of the specimen will to a large degree determine the optimum size and separation of the jet orifices, the proper applied voltage, and the temperature of the electrolyte, as well as other variables involved in the electrolytic reduction of the specimens.

Generally, optimum electropolishing conditions occur at that point (at x on curve A, at a on curve B, and at b on curve C, in FIG. 9) on the curve $I=f(V)$, at which the value of V is slightly less than its value at the point of inflection, the latter point being where the curvature changes from convex to concave. It will be understood that, in each of curves A, B, and C, the temperature of the electrolyte and the position, size, and composition of the specimen remained substantially constant throughout the range of voltages for which the current is plotted. In addition, for each of curves B and C, the jet flow was kept substantially constant at a predetermined value. (Curve A represents conditions where there was no jet flow.) For curve B, the total jet flow from both nozzles **16** was in the neighborhood of 300 ml. per minute; and for curve C, the total jet flow was in the neighborhood of 487 ml. per minute. In other words, the jet flow is substantially proportional to the applied voltage. In the example of the invention described herein, the jet flow increases at the rate of about 75 ml. per minute for each 10 volt increase in the applied voltage in excess of 10 volts.

According to the provisions of the patent statutes, I have explained the principle of my invention and have illustrated and described what I now consider to represent its best embodiment. However, I desire to have it understood that, within the scope of the appended claims, the invention may be practiced otherwise than as specifically illustrated and described.

I claim:

1. Electropolishing apparatus for electrolytically thinning a metallic specimen to a thickness of less than 0.5 micron for use in transmission electron microscopy, comprising a container holding a liquid electrolyte of the type that forms an anolyte layer at the anode, a pair of electrolyte discharge nozzles supported in the electrolyte in substantially coaxial opposition to each other, means for supporting the specimen totally immersed in the electrolyte substantially midway between the jet nozzles, means for circulating the electrolyte through orifices in the nozzles to cause jet impingement of the electrolyte against opposite faces of the specimen, means for applying a predetermined voltage across the electrolytic system with the specimen as the anode, and means for regulating at least one of the parameters consisting of the applied voltage and the velocity of the jet impingement of electrolyte against the specimen to produce an anolyte layer at the electrolyte-specimen interface having a controlled thickness characterized by its electrical resistances being at the lower end of a range over which the resistance of the layer increases substantially at the same rate as the applied voltage to obtain electrolytic reduction of the specimen without etching and without pitting of its surface while maintaining the applied voltage at a level that is high enough to cause gas evolution at said interface and consequent pitting of the specimen in the absence of jet circulation of the electrolyte.

2. Apparatus according to claim **1**, in which the applied voltage is less than 90 volts, and in which the velocity of the electrolyte through the nozzles is increased until the electric current in said system reaches a minimum value.

3. Apparatus according to claim **1**, in which the ratio of the cross sectional area of the nozzle orifices to the area of the specimen exposed to the electrolyte is at least 1 to 8.

4. Apparatus according to claim **1**, in which the ratio of the cross sectional area of the nozzle orifices to the area of the specimen exposed to the electrolyte is approximately 1 to 4.

5. Apparatus according to claim **1**, in which the spacing of the jet orifices is between 1 centimeter and 6 centimeters.

6. Apparatus according to claim **1**, in which the spacing of the jet orifices is about 3.8 centimeters.

7. The method according to claim **3**, in which the electrolyte is circulated through said orifices at a rate substantially proportional to the applied voltage.

8. Apparatus according to claim **1**, in which the velocity of the electrolyte through the nozzles is initially adjusted

to provide a total flow of less than 600 millilitres per minute and the applied voltage is increased until the electric current reaches a value that does not substantially change as the applied voltage is further increased over a predetermined range, thereafter maintaining the applied voltage at the lower end of said range.

References Cited

UNITED STATES PATENTS

2,828,255	3/1958	Gempe	204—199
2,767,137	10/1956	Evers	204—143

3,293,162	12/1966	Sullivan	204—140.5

OTHER REFERENCES

P. M. Kelly et al.: J. of the Iron and Steel Inst., vol. 192, pp. 246–248, July 1959.

G. R. Booker et al.: J. of the Electrochemical Soc., vol. 109, No. 12, pp. 1167–1171, December 1962.

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