

SPECIFICATION
METHOD AND APPARATUS FOR MANUFACTURING SOLID
ELECTROLYTIC CAPACITOR

Field of the Invention

The present invention relates to a method and apparatus for manufacturing a solid electrolytic capacitor using an electroconductive high polymer as a solid electrolyte.

Background of the Invention

With the recent digitization of electronic appliances, a demand has increased for capacitors reduced in impedance in a high frequency range, reduced in size and increased in capacity to be used in digitized electronic appliances. Conventional capacitors capable of operating in a high-frequency range, provided to meet this demand and presently used include aluminum dry electrolytic capacitors and aluminum or tantalum solid electrolytic capacitors as well as plastic film capacitors, mica capacitors, and multilayer ceramic capacitors.

The aluminum dry electrolytic capacitors above-described have a capacitor element formed by winding an etched anode/cathode aluminum foil with a separator interposed between portions of the aluminum foil. This capacitor element is used in combination with a liquid electrolyte. Aluminum or tantalum solid electrolytic capacitors use a solid electrolyte from the idea of solidifying the electrolyte of the above-described aluminum or tantalum solid

electrolytic capacitors for the purpose of improving the characteristics.

To form a solid electrolyte for such solid electrolytic capacitors, a manganese oxide layer is formed on an anode member by immersing the anode member immersed in a nitrate solution and by thermally decomposing, in a high-temperature furnace at 250 to 350°C, the anode member taken out of the solution. Solid electrolytic capacitors using such a solid electrolyte are free from reduction in capacity by efflux of the electrolyte at a high temperature or by dryup, malfunctioning resulting from solidification in a low temperature region, etc., since its electrolyte is in the solid state, and have improved frequency and temperature characteristics in comparison with electrolytic capacitors using liquid electrolytes.

Solid electrolytic capacitors in which a conductive high polymer formed by polymerizing a polymerizable monomer such as pyrrole or thiophene is used as a solid electrolyte having higher electrical conductivity have recently been put to practical use. There are several methods of polymerizing such a conductive high polymer. In particular, electrolytic polymerization is known as a method for obtaining a solid electrolyte of a high electrical conductivity. This method prepares an electrolytic solution for polymerization by dissolving in a solvent a polymerizable monomer and a supporting electrolyte for developing conductivity. It is known that a high-quality solid electrolyte having a markedly high conductivity can be obtained by this method because

doping with the supporting electrolyte occurs simultaneously with the formation of the conductive high polymer during electrolytic polymerization.

In a process for manufacturing a solid electrolytic capacitor having the above-described conventional electroconductive high polymer as a solid electrolyte, however, since a cathode disposed inside a polymerization bath for electrolytic polymerization is made of metal such as stainless, the surface area of a cathode placed in a polymerization bath is changed with passage of time by melting of a surface portion or deposition of a component of the polymerization solution on the surface during use for electrolytic polymerization over a long time period, resulting in a defective condition, e.g., a reduction in the current flowing through the cathode while a constant voltage is applied thereto. In such a case, it is difficult to maintain the desired stability of production.

Moreover, because the cathode is placed so close to an anode member forming an anode that a gas generated at the cathode can easily be attached to the anode member, there is a problem of difficulty in obtaining a uniform electroconductive high polymer film.

Disclosure of the Invention

In view of the above-described problems of the conventional art, an object of the present invention is to provide a method and apparatus for manufacturing a solid electrolytic capacitor in such a manner that a uniform

electroconductive high polymer film can be formed with stability.

To achieve the above-described object, according to one aspect of the present invention, there is provided a method of manufacturing a solid electrolytic capacitor in which an anode member is immersed in a polymerization solution of pH 5 or less in particular, and a solid electrolyte layer of a conductive polymer is formed on a dielectric oxide film layer on the anode member by performing electrolytic polymerization using a feed electrode as an anode and using as a cathode a carbon member placed in the polymerization solution. The surface of the cathode used in this method is not easily molten even if an acidic polymerization solution of pH 5 or less is used, and the carbon member has a larger surface area in comparison with a metallic member since it is porous. Consequently, the reduction in the current for polymerization during a long period of use due to a deposit on the surface of the cathode is minimized to enable a uniform solid electrolyte layer to be formed with improved stability.

In the above-described method of manufacturing a solid electrolytic capacitor, a conductive feed tape may be adhered to the anode member and electrolytic polymerization may be performed by using the feed tape as an anode. The method using this means has the effect of achieving both an improvement in productivity and stabilization of quality.

According to another aspect of the present invention, there is provided an apparatus for manufacturing a solid electrolytic capacitor, the apparatus having a

polymerization bath filled with polymerization solution, a cathode made of carbon and having at least its portion immersed in the polymerization solution in the polymerization bath, and a feed electrode used as anode through which a current is fed to an anode member immersed in the polymerization solution. This apparatus contributes to the above-described effects of the invention.

In the above-described manufacturing apparatus, a feed tape adhered to the anode member may be used as the feed electrode used as an anode. This arrangement also contributes to the above-described effects of the invention.

In the above-described manufacturing apparatus, the cathode may be formed so as to be reduced in width, the amount of reduction in width being increased along a direction toward the anode member. Also, the cathode may have a slanted surface in its surface facing the anode member, the slanted surface extending along such a direction to one of the opposite ends of the surface facing the anode member that the distance between the slanted surface and the anode member is increased. This arrangement ensures that a gas generated at the cathode can be quickly released to an upper section of the polymerization bath, thereby preventing the gas from being attached to the anode member and making it possible to form a uniform conductive high polymer film with stability.

In this arrangement, the cathode may have at least two surfaces formed as the slanted surface to further improve the effect of quickly releasing the gas generated at the cathode to the upper section of the polymerization bath while

maintaining a certain distance between the cathode and the anode member.

In the above-described manufacturing apparatus, the cathode may be divided to be placed closer to two sides of an upper section of the polymerization bath in the above-described invention. In this arrangement, the divided cathodes are respectively positioned in correspondence with the polymerized film growth directions from the feed tape adhered to a center of the anode member, thereby improving the efficiency with which the anode member is coated with the polymerized film and, hence, the productivity of the solid electrolytic capacitor.

In the above-described manufacturing apparatus, the distance between the anode member and the cathode may be set within the range from 2 to 12 mm to ensure that the gas generated at the cathode is not attached to the anode member, and to thereby achieve a reduction in the electrolytic polymerization time, i.e., an improvement in productivity.

In the above-described manufacturing apparatus of the present invention, the width of the cathode may be made larger than that of the feed tape in the above-described invention, to ensure that the interelectrode distance can be uniformly set through a wide area on the anode member, and to thereby achieve a reduction in the electrolytic polymerization time, i.e., an improvement in productivity.

In the above-described manufacturing apparatus, the specific resistance of the carbon member provided as the cathode may be set to 1000 $\mu\Omega\text{cm}$ or less in the above-described

invention, to reduce the voltage drop across the current path in the cathode from a cathode feed point and to constantly maintain the potential difference between the feed tape and the cathode, thereby achieving the effect of achieving production with improved stability.

In the above-described manufacturing apparatus, a carbon member impregnated with a phenolic solution may be used as the cathode to improve the mechanical strength of the cathode and thereby enable the polymerized film to be formed with improved stability.

Brief Description of the Drawings

FIG. 1 is a cross-sectional view showing a polymerization bath in Embodiment 1 of the present invention;

FIG. 2 is a sectional front view of the polymerization bath shown in FIG. 1;

FIG. 3A is a plan view of an anode member;

FIG. 3B is a cross-sectional view of the anode member shown in FIG. 3A;

FIG. 3C is a cross-sectional view of the anode member on which a polymerized film is formed;

FIG. 4 is a characteristics diagram showing changes with passage of time, of required current for polymerization reaction in Embodiment 1;

FIG. 5 is a cross-sectional view of a polymerization bath in Embodiment 2 of the present invention; and

FIG. 6 is a plan view of an anode member in Embodiment 2.

Description of Preferred Embodiments

Embodiments of the present invention will be described with reference to the accompanying drawings.

FIG. 1 is a sectional side view of a polymerization bath which is an essential portion of an apparatus for manufacturing an anode member for use in a solid electrolytic capacitor. FIG. 2 is a sectional front view of the polymerization bath. In these figures are illustrated cathodes 1 made of carbon, feed electrodes 2, and an anode member 3 made of a valve metal and having a layer of a dielectric oxide film formed on its surface. Each cathode 1 is placed so as to face the upper surface of the anode member 3. A cathode feed point is indicated by 7; the polymerization bath in the form of a channel, by 8; a polymerization solution with which the polymerization bath 8 is filled, by 9; and ends of the cathode, by 10.

FIG. 3A is a plan view of the anode member of the solid electrolytic capacitor, FIG. 3B is a cross-sectional view of the member shown in FIG. 3A, and FIG. 3C shows a semi-finished product. In this figures are illustrated a dielectric oxide layer 3a formed on the anode member 3, insulating tapes 4, and an anode lead portion 5 of the anode member 3, and cathode lead portions 6 of the anode member 3.

(Embodiment 1)

Embodiment 1 of the present invention relating to the attached claims 1 and 3 in particular will be described. Two surfaces of an anode member 3 formed of an aluminum foil having

a thickness of 100 μm were electrochemically roughened and then anodized at an anodizing voltage of 35 V to form a dielectric oxide film layer 3a on the external surface.

Next, as shown in FIGS. 3A and 3B, insulating tapes 4 were adhered to the two surfaces of the anode member 3 on opposite sides to enable an anode lead portion 5 and cathode lead portions 6 to be formed separately from each other. The size of the cathode lead portions 6 was set to 3 x 4 mm.

Thereafter, another dielectric oxide film layer was formed on the external surface of the anode member 3 by an anodizing process to cover defective portions in the sectional surfaces of the aluminum foil and in the dielectric oxide layer initially formed.

Subsequently, an aqueous solution of manganese nitrate was applied to the cathode lead portions 6, and the member with the applied solution was heated at 300°C for 5 minutes in a high-temperature furnace to decompose manganese nitrate. A manganese dioxide layer 13 was thereby formed as a conductor layer on the cathode lead portions 6.

Next, electrolytic polymerization was performed on the anode member 3 prepared as described above while the portion of the anode member 3 immersed in a polymerization solution 9 (an aqueous solution prepared by adding 0.1 mol/liter of propyl phosphate as a PH adjustor to an aqueous solution of 0.2 mol/liter of pyrrole and 0.1 mol/liter of alkylnaphthalenesulfonate) in the polymerization bath 8 shown in FIGS. 1 and 2 was being changed. This electrolytic polymerization was performed by using the polymerization

electrodes, i.e., the feed electrodes 2, as a common positive electrode in contact with the anode member 3, using as four independent cathodes 1 the four carbon members having their portions under the surface of the solution, maintaining the interelectrode distance H at 8 mm, and applying a voltage of 3 V between the feed electrodes 2 and the cathode feed points 7. Table 1 shows the result of measurement of a time required for the above-described polymerization with respect to each of different polymerization solution pH values.

Table 1

Polymerization Solution pH	2.1	3.2	4.0	5.0	5.5	6.2	6.8	7.5
Polymerization Time (min.)	18	21	21	22	30	41	50	63

As is apparent from Table 1, the time required to coat the anode member with the polymer is markedly reduced when the pH is not larger than 5, because the polymerization current contributing to the polymerization reaction can be increased.

In the electrolytic polymerization process performed as described above, polymerization is initiated at the feed electrodes 2, and the polymerization area expands along the cathode lead portions 6 in FIG. 3A until the entire surfaces of the cathode lead portions 6 are coated with polymerized film 14 of the conductive high polymer. That is, during a time period of about 20 minutes from the time when one section of the anode member 3 enters the polymerization bath 8 shown in FIG. 2 to the time when the section moves out of the

polymerization bath 8, polymerized film 14 of the conductive high polymer is formed on the entire surfaces of the cathode lead portions 6.

FIG. 3C shows the anode member 3 on which the above-described polymerized film 14 is formed. A carbon layer 16 is formed on the polymerized film 14 on the anode member 3, and a silver layer is further formed on the carbon layer 16. It is then cut into unit pieces. A desired number of unit pieces thus formed are stacked and packed in a casing to complete a solid electrolytic capacitor.

The above-described polymerization reaction was sustained for one week and the state of the current flowing through the path between the feed electrodes 2 and the carbon cathodes 1 was measured. FIG. 4 shows the result of this measurement in comparison with the conventional art. As shown in FIG. 4, the polymerization current in Embodiment 1 was maintained in a stable state during one week, while the polymerization current in the conventional apparatus started decreasing about 50 hours after the start of the process and was not stable.

In Embodiment 1 of the present invention, as described above, carbon is used as the material of the cathode 1. The amount of reduction with passage of time in surface area of the cathode caused by melting of the cathode surface or deposition of a polymerization solution component is thereby reduced in comparison with the case of use of a metallic material such as stainless steel. Thus, the method of the present invention enables production of a solid electrolytic

capacitor with improved stability by reducing the degree of defectiveness such as a reduction in current when a constant voltage is supplied.

Embodiment 1 of the present invention has been described with respect to the case where pyrrole was used as a polymerizable monomer in a polymerization solution. However, the present invention is not limited to the described case and the same effect can also be achieved by using at least one polymerizable monomer selected from thiophene, aniline and derivatives of these substances.

Embodiment 1 has been described with respect to an arrangement using an aluminum foil as a valve metal by way of example. However, the present invention is not limited to the described arrangement and the same effect can also be achieved by using a foil or a sintered piece of a metal selected from other valve metals, e.g., tantalum, niobium, and titanium.

(Embodiment 2)

Embodiment 2 of the present invention relating to the attached claims 2 and 4 in particular will be described. As Embodiment 2, a process was performed in which polymerization reaction was caused and sustained in the same manner as that in Embodiment 1 except that a feed tape 12 substituted for the feed electrodes used as an anode for electrolytic polymerization in Embodiment 1 was adhered to an anode member 5 and electrolytic polymerization was performed by using this feed tape 12 as an anode. Therefore the portions of the members and the apparatus identical or corresponding to those

in Embodiment 1 are indicated by the same reference numerals, and the detailed description for them will not be repeated. Only different portions will be described with reference to the drawings.

FIG. 5 is a cross-sectional view of a polymerization bath which is an essential portion of an apparatus for manufacturing a solid electrolytic capacitor in Embodiment 2 of the present invention. FIG. 6 is a plan view of the anode member of the solid electrolytic capacitor. Referring to FIG. 6, the feed tape 12 made of stainless steel is adhered to the portion of the anode member 3 so as to cover the entire length of the anode lead portion 5. A current is supplied to the feed tape from feed rollers 15 placed above two ends of the polymerization bath 8. Cathodes 1 are positioned so as to face the upper surface of the feed tape 12. To enable discrimination between the anode member 3 and the feed tape 3 as seen in FIG. 5, the anode member 3 and the feed tape 3 are illustrated in a state of being spaced apart from each other.

In Embodiment 2 of the present invention using the above-described arrangement, electrolytic polymerization is performed by using as an anode the feed tape 12 adhered to the anode member 3 in advance, and there is no physical contact between the feed electrode and the anode member 3 such as that in Embodiment 1. Therefore current feed can be performed with stability without causing any stress in the anode member 3. Also, there is no need to clean or replace the feed

electrode during any long period of use, so that the productivity and production stability can be improved.

(Embodiment 3)

Embodiment 3 of the present invention relating to the attached claims 5, 6, 7, and 8 in particular will be described. The shape of cathodes 1 used in Embodiment 3 is such that the cathode in Embodiment 2 is formed so as to have its lower portion reduced in width with, the amount of reduction in width being increased along the direction toward the anode member 3. Referring to FIG. 5, the cathode 1 is formed so as to have at least two slanted surfaces 11 in its surface facing the anode member 3, the slanted surfaces 11 extending along such directions to the opposite ends of the surface facing the anode member 3 that the distance between each slanted surface and the anode member is increased.

Polymerization reaction was caused and sustained under the same conditions as those in Embodiment 2 except for the condition with respect to the shape of the cathode 1. The shape of the cathode 1 is not limited to that shown in FIG. 5. The lower end of the cathode 1 may be formed into any other shape, e.g., the shape of an equilateral triangle or a right-angled triangle, or the cathode 1 may have any other shape provided that the amount of reduction in width is increased along the direction toward the anode member 3.

In Embodiment 3, the manufacturing apparatus is thus arranged to enable hydrogen gas generated at the cathode 1 during polymerization reaction to quickly move upwardly without staying at the cathode 1, thus enabling prevention

of a phenomenon in which a reduction in polymerization current results from a reduction in the area of the cathode 1 during a substantially long period of use.

The time required to complete polymerization reaction in a case where the distance H between the cathode 1 and the anode member 3 to which the feed tape 12 was adhered was maintained at 8 mm and where the cathode 1 was positioned right above the feed tape 12, and the time required to complete polymerization reaction in another case where the same distance was maintained but the cathode 1 was divided into two portions separately placed along two sides of an upper section of the polymerization bath 8, as set forth in claim 8 were compared. Table 2 shows the result of this comparison. In this embodiment, cathodes having such shapes that the cathode 1 shown in FIG. 5 is cut along a vertical direction at its center were used by being placed in a state of being spaced apart from each other.

Table 2

Cathode Positions	Center	Opposite Sides
Polymerization Time (min.)	30	23

As is apparent from Table 2, there is a substantial difference between the anode member 3 coating times depending on the positions of the anodes 1 while the interelectrode distance is constantly maintained. This is because the growth of the polymerized film spreads in directions from the feed tape 12 to the anode member 3, and because the efficiency

of polymerization is increased if the cathodes 1 are placed in correspondence with the polymerized film growth directions.

(Embodiment 4)

Embodiment 4 of the present invention relating to the attached claim 9 in particular will be described. In Embodiment 4, the distance between the anode member 3 and the lower portion of the cathode 1 in Embodiment 2 is changed. Polymerization reaction was caused and sustained in the same manner as that in Embodiment 2 except for this condition.

Polymerization reaction processing was performed with respect to different distances H: 1, 2, 3, 4, 6, 8, 12, 14, and 16 mm. To form a cathode lead layer, a colloidal carbon suspension was applied to each of anode members 3 on which polymerization processing was performed under the different conditions. This suspension was dried to form a carbon layer. Further, a silver paste was applied to the surface of the carbon layer and dried to form a silver layer as a cathode layer, to which a cathode lead was connected to complete a solid electrolytic capacitor. The equivalent series resistance at 100 kHz of solid electrolytic capacitors thus obtained was measured. Table 3 shows the times required to coat the anode members 3 and the equivalent series resistances of the capacitors.

Table 3

Inter-electrode Distance (mm)	1	2	3	4	6	8	12	14	16
Polymerization	25	25	26	28	29	30	30	40	45

Time (min.)									
ESR (m Ω)	45	35	33	32	30	30	29	28	28

As is apparent from Table 3, a tendency of the resistance of the conductive high polymer to increase with the reduction in the distance H between the anode member 3 and the carbon cathode 1 is recognized. This is because the surrounding solution is alkaline in the vicinity of the cathode 1. An increase in equivalent series resistance of the solid electrolytic capacitor results from such a condition. If the distance H between the anode member 3 and the carbon cathode 1 is unnecessarily increased, it tends to be difficult for polymerization reaction to take place. The optimum distance, determined by considering the productivity, is 12 mm or less.

As described above, in Embodiment 4, it is possible to achieve both an improvement in productivity and stabilization in terms of economy by setting the distance H between the anode member 3 and the cathode 1 in the range from 2 to 12 mm. When the distance H is in this range, the time required to complete polymerization reaction can be effectively reduced while the desired characteristics of the anode member 3 (capacitor element) are maintained.

(Embodiment 5)

Embodiment 5 of the present invention relating to the attached claim 10 in particular will be described. Embodiment 5 includes an experiment in which the width of the cathode 1 in Embodiment 2 was reduced to a value equal to or smaller than that of the feed tape 12. Except for this, polymerization reaction was caused and sustained in the same

manner as that in Embodiment 2, thereby making anode members 3. Table 4 shows the result of measurement of the relationship between the current and time required to complete polymerization reaction in this embodiment.

Table 4

	Cathode Width > Feed Tape Width	Cathode Width = Feed Tape Width	Cathode Width < Feed Tape Width
Polymerization Current (mA)	2053	1860	1508
Polymerization Time (min.)	29	32	48

As is apparent from Table 4, the time required to complete polymerization reaction was increased when the width of the cathode 1 was set to a value equal to or smaller than that of the feed tape 12. This is because reduction in the current contributing to polymerization reaction results from the reduction in the area of the cathode 1, and because the cathode 1 does not extend along the polymerized film growth direction. Consequently, polymerization reaction can be caused and sustained with improved efficiency and stability if the width of the cathode 1 is larger than that of the feed tape 12. (Embodiment 6)

Embodiment 6 of the present invention relating to the attached claim 11 in particular will be described. In Embodiment 6, the specific resistance of the carbon member provided as the cathode 1 in Embodiment 2 was changed by being selected from 500, 1000, 1500, and 2000 $\mu\Omega\text{cm}$. Except for this,

polymerization reaction was caused and sustained in the same manner as that in Embodiment 2, thereby making anode members 3. Table 5 shows the voltage drop between the cathode feed point 7 and the cathode end 10 and the time required to coat each anode member 3.

Table 5

Carbon Cathode Specific Resistance ($\mu\Omega\text{cm}$)	500	1000	1500	2000
Voltage Drop (V)	0.3	0.9	1.8	2.5

As is apparent from Table 5, in the case where the voltage drop is larger than 1 V while the voltage between the cathode feed point 7 and the feed tape 12 is 3 V, the time required to coat the anode member 3 is increased and the productivity is considerably reduced. To solve the problem of such a large voltage drop across the path between the cathode feed point 7 and each cathode end 10 shown in FIG. 2, a method of increasing the number of cathodes 1 by dividing the cathode 1 into short divisions or a method of providing additional feed points may be used. However, these methods are disadvantageous in terms of maintenance and cost.

Therefore the specific resistance of the carbon member forming the cathode is set to 1000 $\mu\Omega\text{cm}$ or less to effectively limit the voltage drop between the cathode feed point 7 and the cathode end 10 even if a reduced number of cathodes 1 each increased in length are reduced, thus making the potential difference between the feed tape 12 and the cathode 1 always constant and ensuring production with improved stability.

(Embodiment 7)

Embodiment 7 of the present invention relating to the attached claim 12 in particular will be described. In Embodiment 7, carbon forming the cathode 1 was baked when the cathode 1 in Embodiment 2 was manufactured, and the carbon member impregnated with a phenolic solution in the final process step was used as the cathode 1. Except for this, polymerization reaction was caused and sustained in the same manner as that in Embodiment 2, thereby making an anode member 3.

In this embodiment, a carbon member impregnated with a phenolic solution is used as the cathode 1 to improve the mechanical strength of the cathode 1 to ensure production with improved stability.

According to the present invention, as described above, a dielectric oxide film layer which is an oxide of a valve layer is formed on the surface of an anode member by anodization, the anode member with the dielectric oxide layer is immersed in a polymerization solution, and electrolytic polymerization is performed by using a feed electrode as an anode and by using as a cathode a carbon member having its portion placed under the surface of the polymerization solution. A layer of a solid electrolyte constituted by a conductive high polymer is thereby formed on the dielectric oxide film layer. This manufacturing method unlike the conventional method using a cathode formed a metal such as stainless steel prevents a reduction with passage of time in the surface area of the cathode caused by melting of the

cathode surface, deposition of a component of the polymerization solution, or the like to solve the problem of a reduction in polymerization current while a constant voltage is applied, and thereby ensures production with improved stability.

CLAIMS

1. A method of manufacturing a solid electrolytic capacitor, wherein:

a dielectric oxide film layer is formed on a surface of an anode member (3) made of a valve metal by anodizing the anode member;

subsequently, a solid electrolyte layer of a conductive polymer is formed on the dielectric oxide film layer, with the anode member (3) immersed in a polymerization solution (8) of pH 5 or less, by performing electrolytic polymerization using a feed electrode (2) as an anode and using as a cathode 1 a carbon member placed in the polymerization solution (8); and

then, a cathode layer is formed on the solid electrolyte layer.

2. The method according to claim 1, wherein a conductive feed tape (12) is adhered to the anode member (3) and electrolytic polymerization is performed by using the feed tape (12) as an anode.

3. An apparatus for manufacturing a solid electrolytic capacitor, comprising:

a polymerization bath (8) filled with polymerization solution (9);

a cathode (1) made of carbon and having at least a portion thereof immersed in the polymerization solution (9) inside the polymerization bath (8); and



a feed electrode (2) used as anode through which a current is fed to an anode member (3) immersed in the polymerization solution (9).

4. An apparatus according to claim 3, wherein the feed electrode used as an anode is a feed tape (12) adhered to the anode member.

5. An apparatus according to claim 3 or 4, wherein the cathode (1) is formed to be reduced in width along a direction toward the anode member.

6. An apparatus according to claim 5, wherein the cathode has slanted surfaces on opposite sides of its surface facing the anode member, the slanted surfaces extending to spread outwards according as the distance between the slanted surfaces and the anode member is increased.

7. An apparatus according to claim 6, wherein the cathode has at least two slanted surfaces.

8. An apparatus according to claim 3 or 4, wherein the cathode is divided to be placed closer to opposite sides of an upper section of the polymerization bath.

9. An apparatus according to claim 3 or 4, wherein the distance between the anode member and the cathode is set within the range from 2 to 12 mm.



10. An apparatus according to claim 4, wherein the cathode has a width larger than that of the feed tape.

11. An apparatus according to claim 3 or 4, wherein specific resistance of a carbon member provided as the cathode is set to 1000 $\mu\Omega\text{cm}$ or less.

12. An apparatus according to claim 3 or 4, wherein a carbon member impregnated with a phenolic solution is used as the cathode.

