

US 20080296151A1

(19) **United States**(12) **Patent Application Publication**
Lee et al.(10) **Pub. No.: US 2008/0296151 A1**(43) **Pub. Date: Dec. 4, 2008**(54) **CONTINUOUS ELECTROLYTIC REFINING
DEVICE FOR METAL URANIUM****Publication Classification**(51) **Int. Cl.**
C25C 7/00 (2006.01)(52) **U.S. Cl.** **204/212**(57) **ABSTRACT**

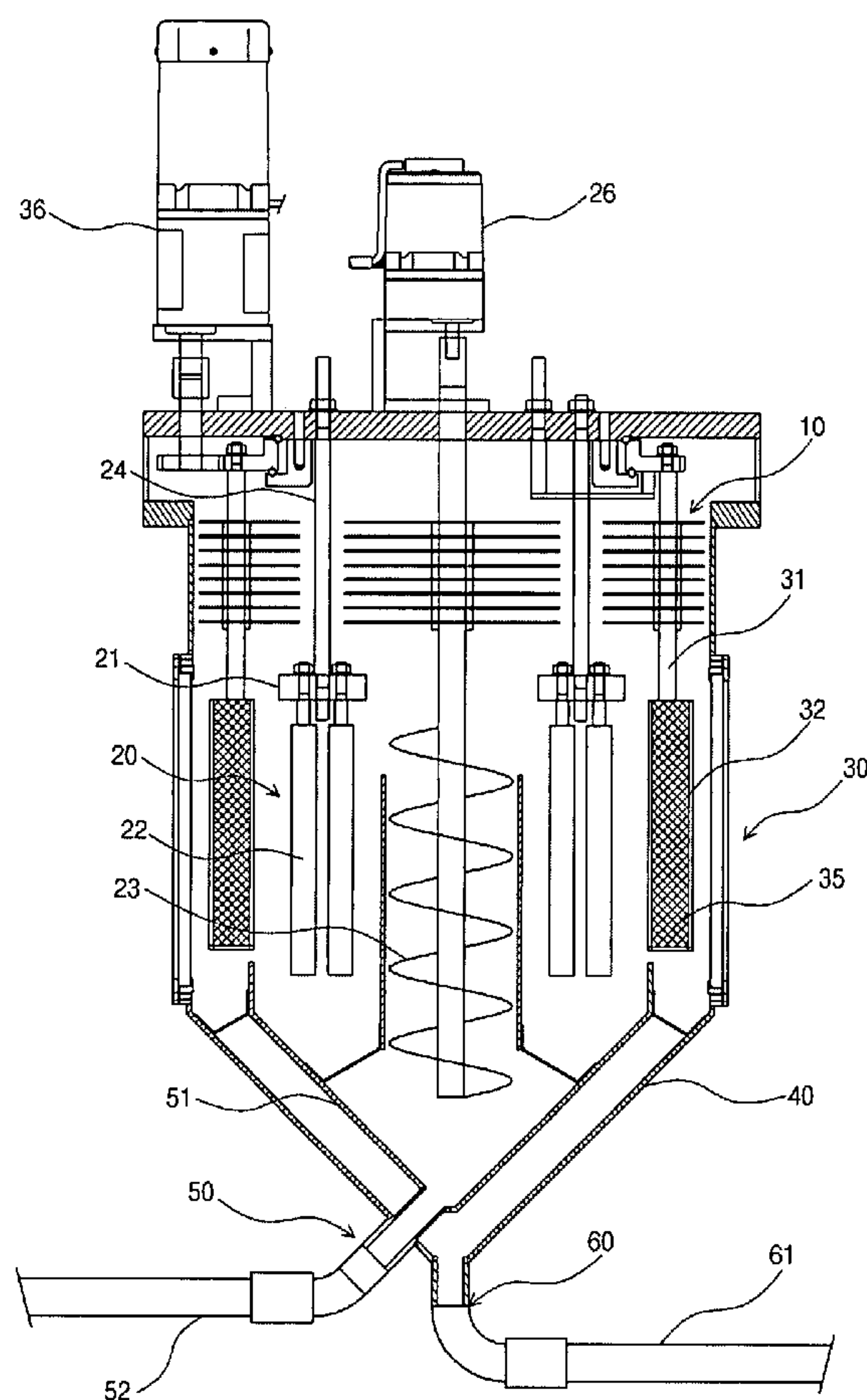
Disclosed herein is a continuous electrolytic refining device for metal uranium, the device comprising a cathode section fixed to the lower side of the heat radiation plate, and having a plurality of graphite cathodes; an anode section encompassing the cathode section to face the cathode section, rotatably fixed to the lower side of the heat radiation plate, and receiving the used nuclear fuel; an electrolytic cell receiving the cathode section and the anode section and filled with electrolytes so as to sink the cathode section and the anode section; an uranium collecting section collecting metal uranium deposited on and detached from the graphite cathode in the lower side of the cathode section inside the electrolytic cell and withdrawing the collected metal uranium to the outside of the electrolytic cell; and a transition metal collecting section coupled with the lower side of the electrolytic cell to withdraw the transition metal particles released from the anode section and collected in the lower side of the electrolytic cell, in order to collect high pure uranium deposits and metal transition elements created in an electrolysis process without stopping an electrolysis process, not including a scrapping process.

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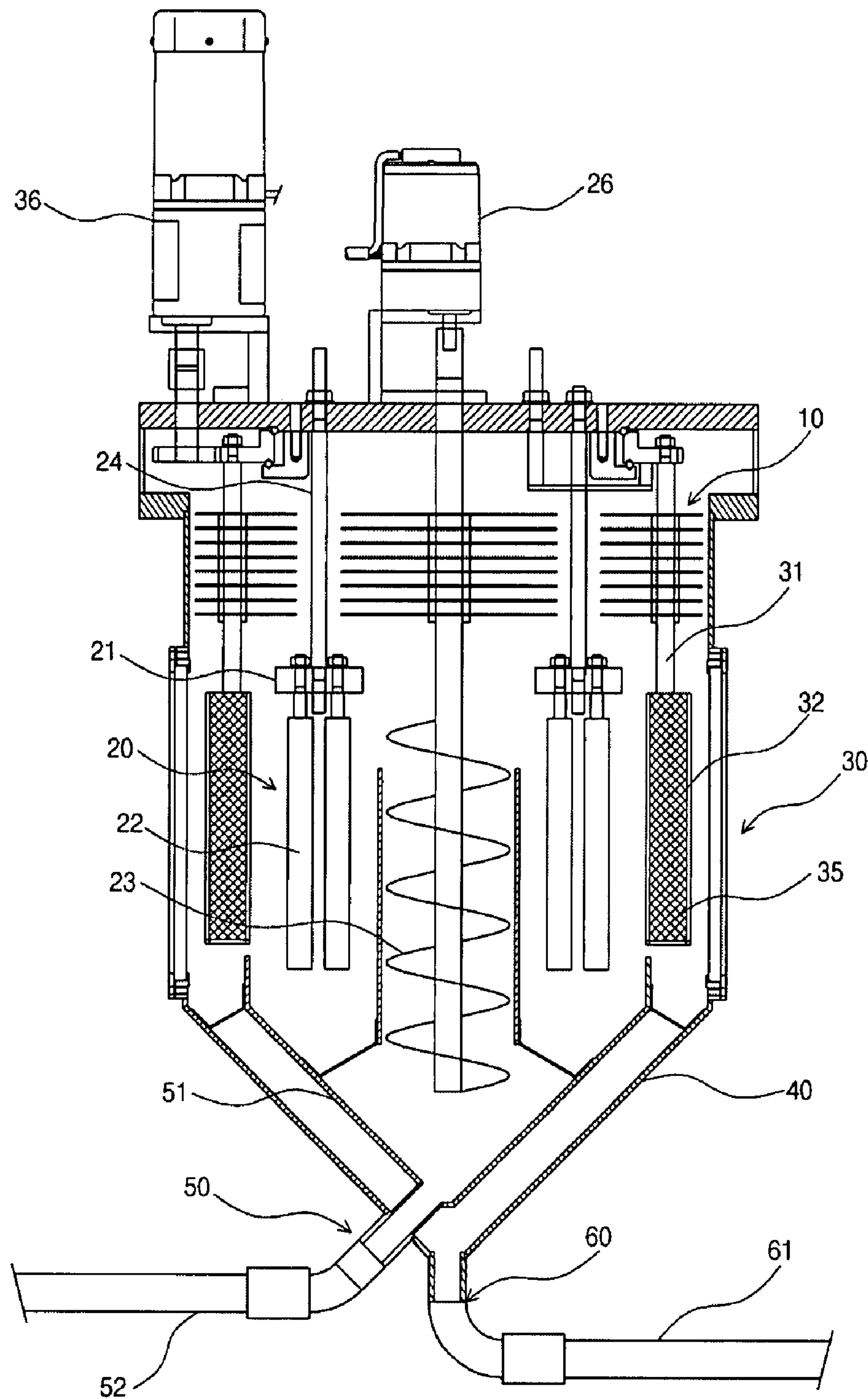
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(21) Appl. No.: **11/889,540**(22) Filed: **Aug. 14, 2007**(30) **Foreign Application Priority Data**

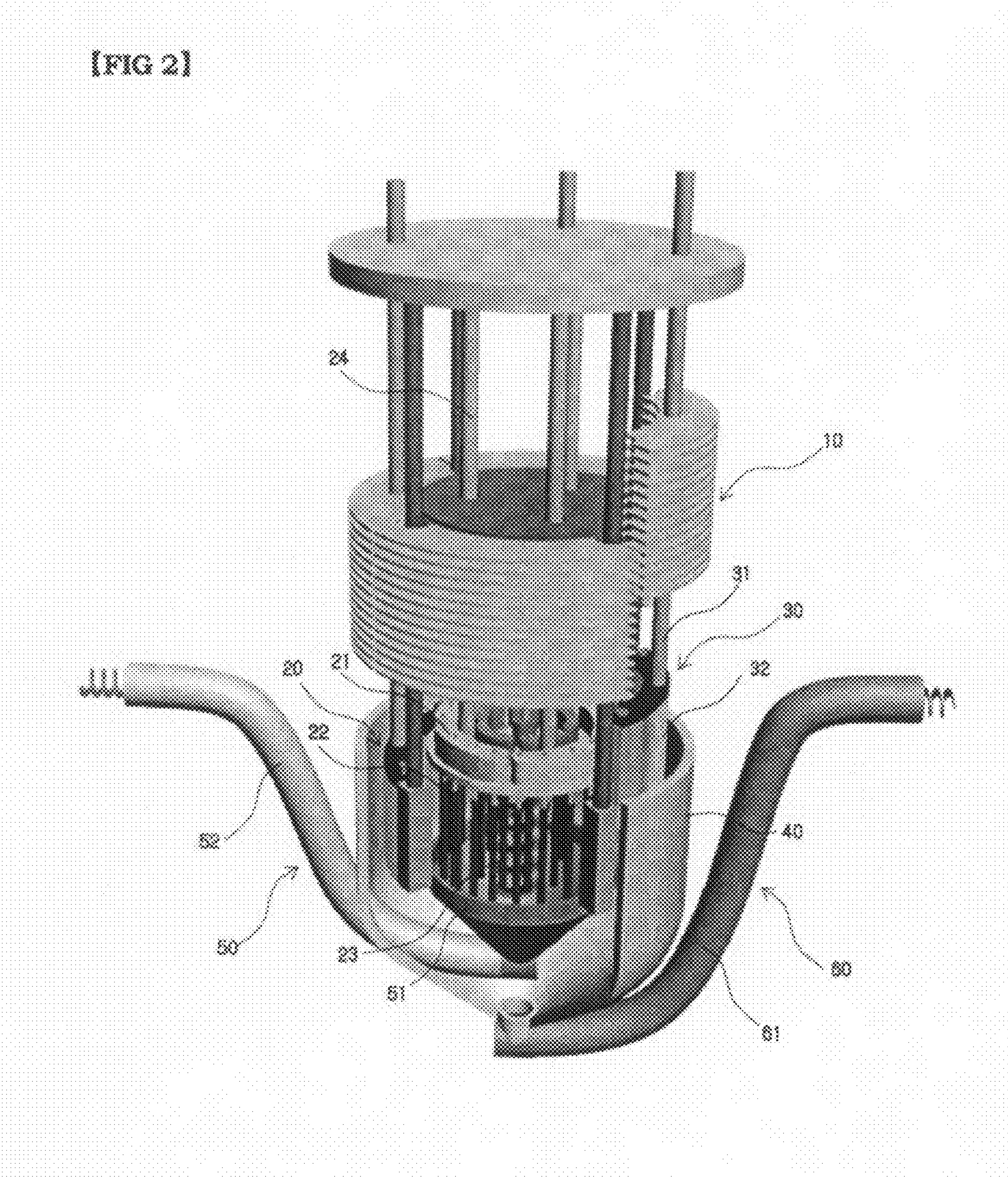
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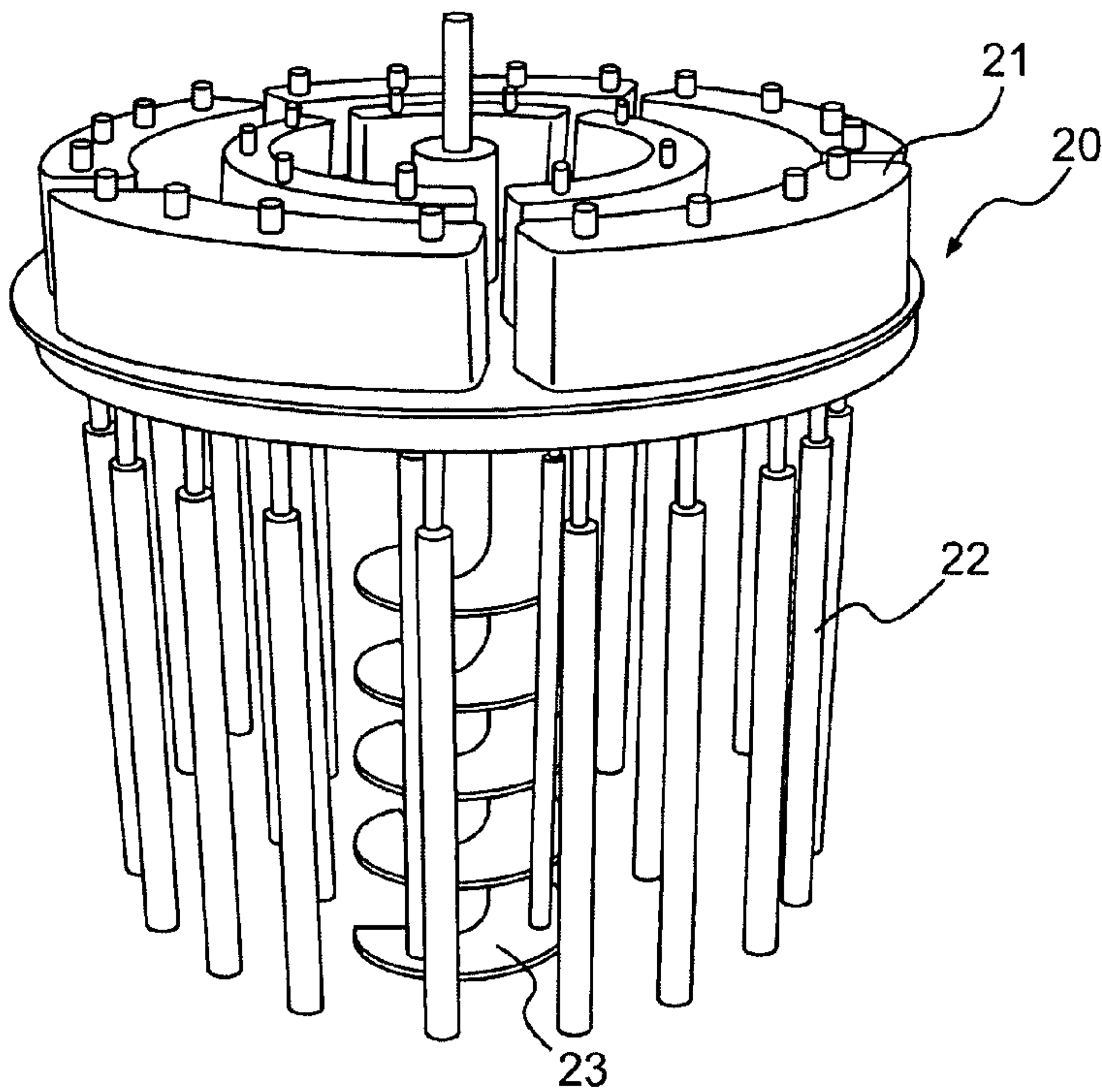


【FIG 1】

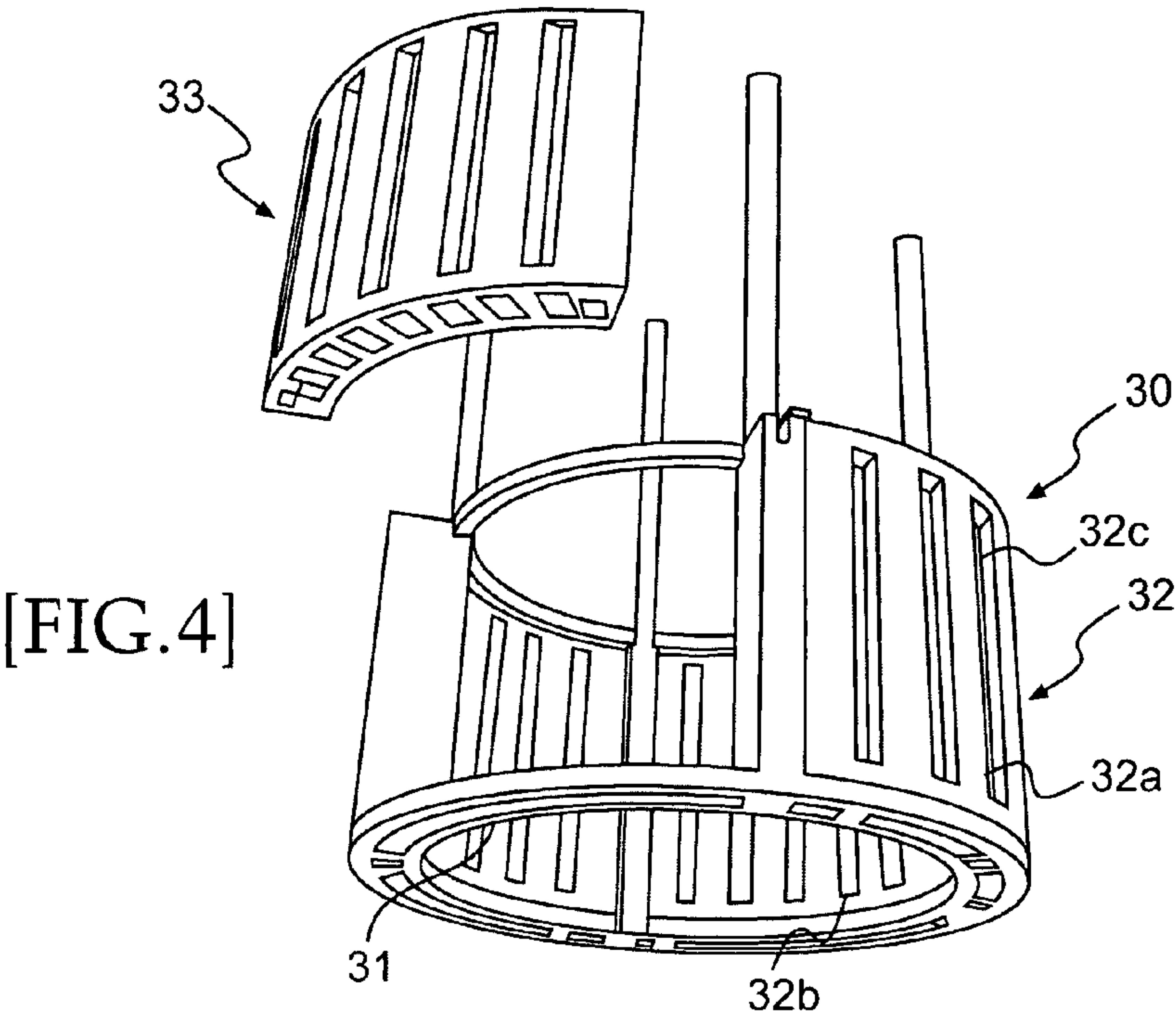


[FIG 2]



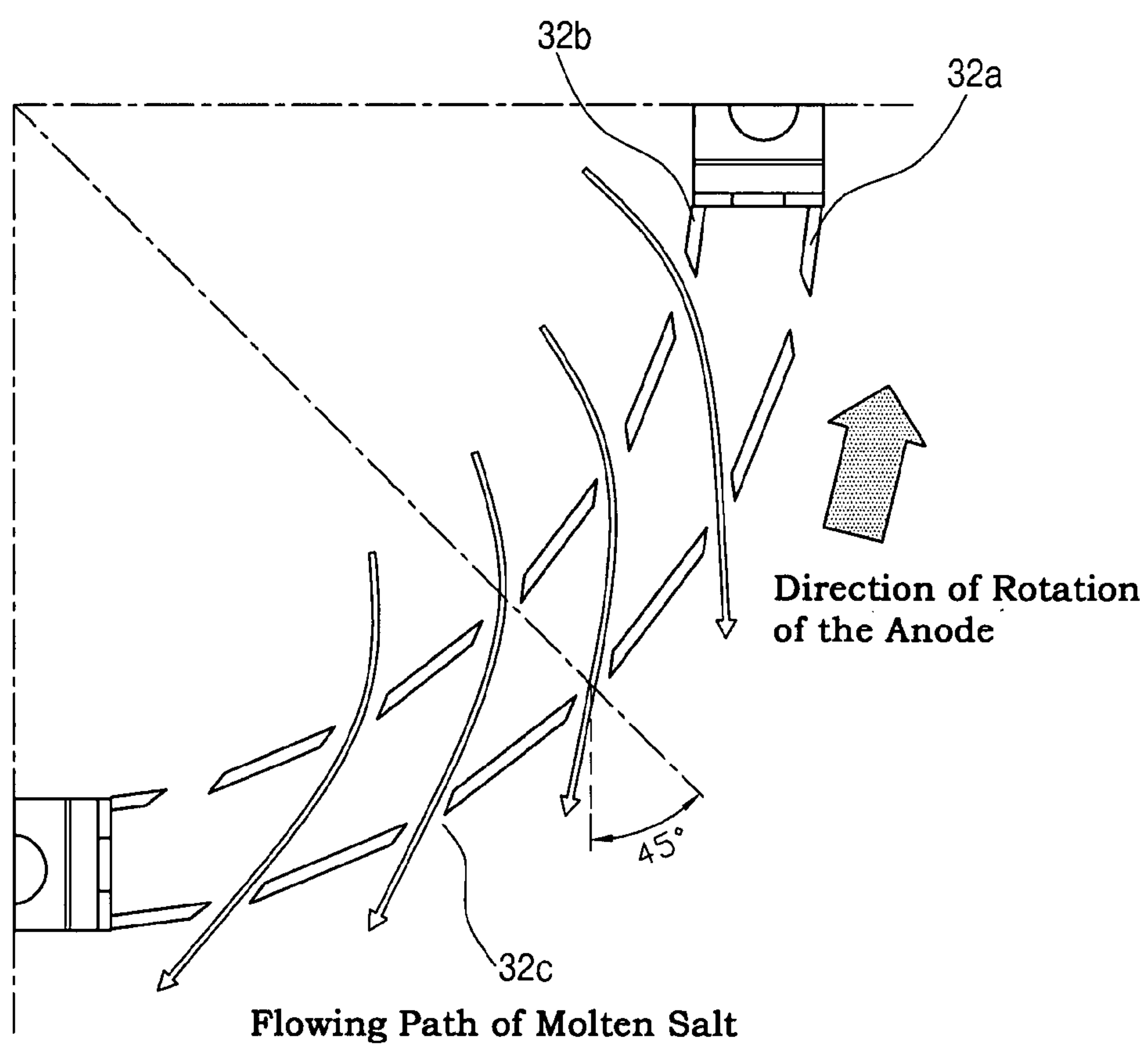


[FIG.3]

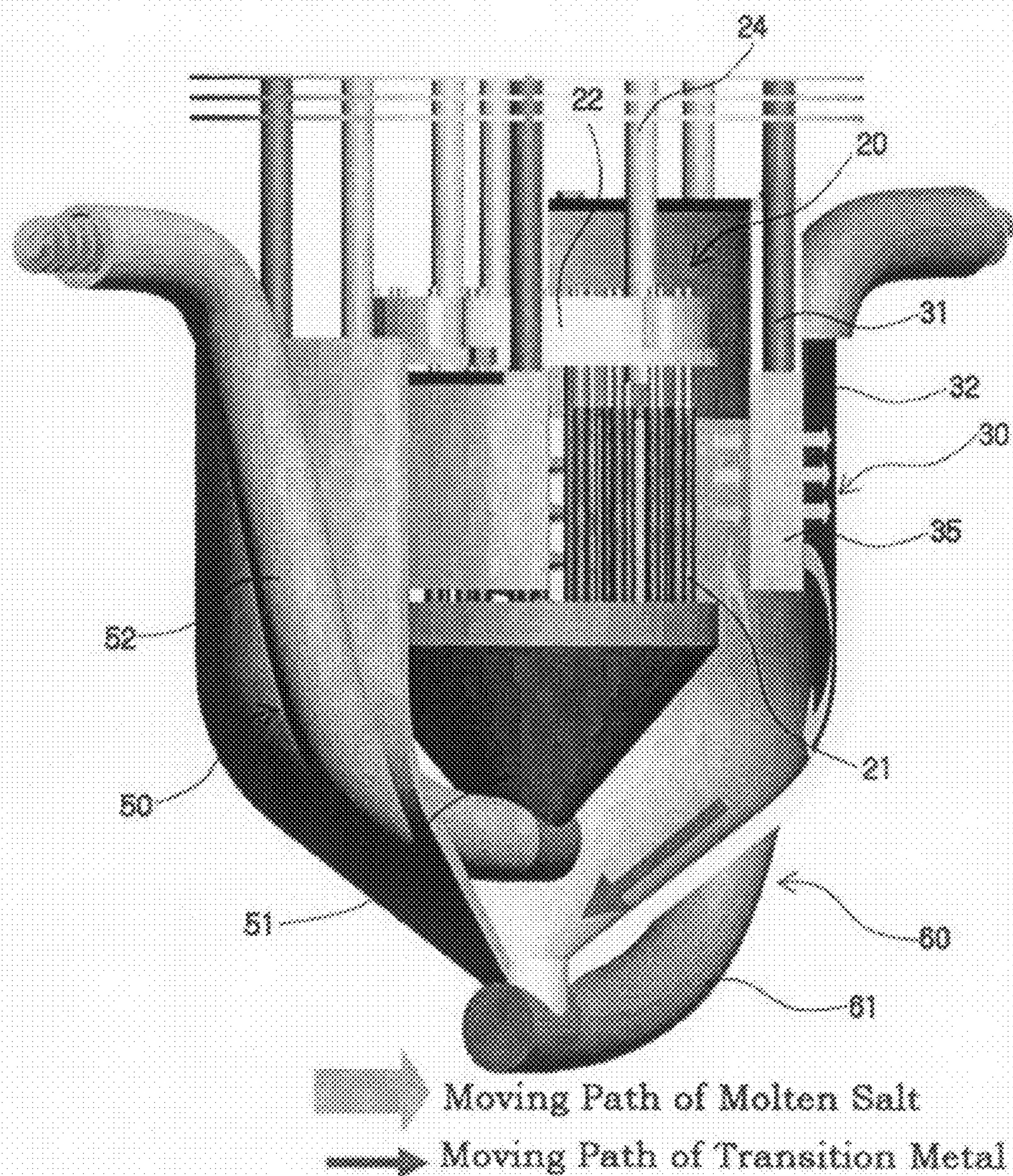


[FIG.4]

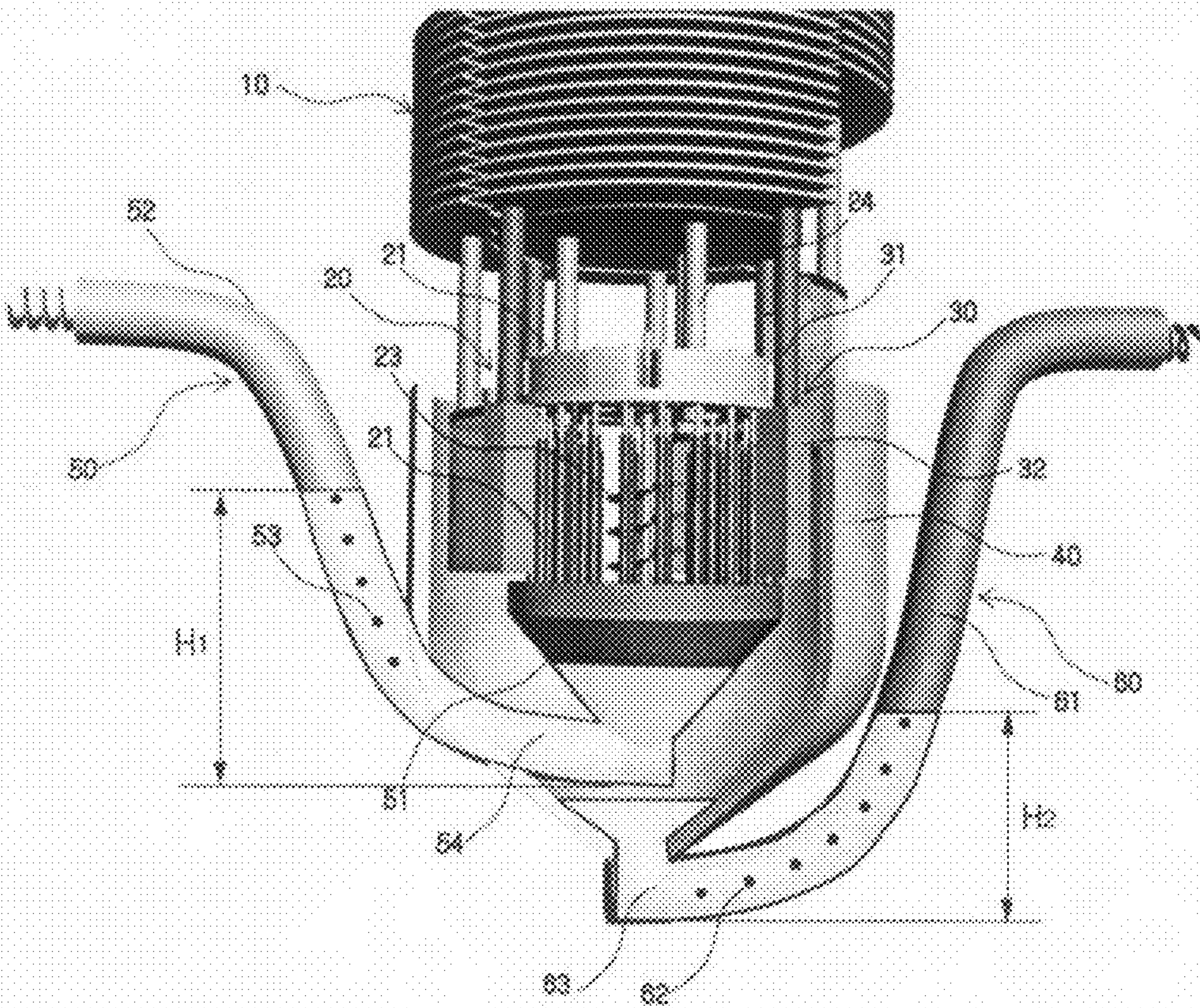
【FIG 5】



[FIG 6]



[FIG 7]



CONTINUOUS ELECTROLYTIC REFINING DEVICE FOR METAL URANIUM

BACKGROUND OF THE INVENTION

[0001] 1. Field of the Invention

[0002] The present invention relates generally to electrolytic refining devices of metal uranium, and more particularly to a continuous electrolytic refining device for metal uranium without stopping an electrolytic process in collecting uranium deposits and metal transition element generated in the electrolytic process.

[0003] 2. Description of the Related Art

[0004] As well-known to the public, the conventional electrolytic refining device of metal uranium used as a nuclear fuel consists of an anode basket including segments of a metal fuel and an iron-based cathode on which pure uranium is deposited after having been used in a melted salt about 500° C. where uranium chloride is molten.

[0005] When refining metal uranium using the conventional electrolytic refining device of metal uranium, if electric power is applied, the uranium chloride in the molten salt is reduced to be deposited on a cathode, and chloride ions separated in the reaction selectively dissolve metal uranium in the anode electrically. It is possible to separate pure metal uranium by reiterating the electrolytic refining process of metal uranium.

[0006] However, the electrolytic refining process of metal uranium using the conventional electrolytic refining device of metal uranium has drawbacks that an electrolytic reaction must be stopped to periodically collect metal uranium deposited on to a cathode, and due to a long operation time to collect electrodeposits, it is impossible to do a continuous operation. Therefore a large quantity of products cannot be obtained in a short time.

[0007] There have been reported electrolytic refining devices to overcome the above drawbacks and separate pure metal uranium at a high speed.

[0008] The U.S. Pat. No. 5,650,053 (Jul. 22, 1997) discloses a refining device where the segments of a metal fuel used in a molten salt in about 500° C. are put in an anode basket of a porous plate, and several anode baskets are placed inside and outside of a cathode which has a tube shape, and if electricity is applied while rotating the anode basket, metal uranium at the anode is melted to be deposited onto the cathode and the deposited metal uranium is scrapped with a ceramic plate attached on the outside of an anode to be collected in a collecting section in the lower part.

[0009] However, the refining device has only a part of the metal uranium deposited on a cathode detached, and the remnant electrodeposits keep sticking on the surface of the cathode, and are gradually changed into a dense tissue which is difficult to detach.

[0010] Accordingly, since it is impossible to detach electrodeposits whose tissue becomes dense with a ceramic plate at an anode, the electrolytic refining operation is stopped after a predetermined time has passed and then electricity is inversely applied to return the electrodeposits to the anode to be stripped. The surface of the cathode becomes clean and the deposition is operated again from the beginning.

[0011] The above stripping process has drawbacks that a lot of electricity is consumed, the deposition efficiency is deteriorated and that the device becomes complicated. Furthermore, there occurs a problem that the electrolysis must be

stopped in order to collect electrodeposits in the lower part and that the entire electrode module should be lifted.

[0012] In addition, the tank for collecting uranium electrodeposits is disposed in the lower part of the anode basket to mix the undissolved transition atom particles generated from the anode with uranium, thus there is a limitation in obtaining high purity uranium electrodeposits.

[0013] The Japanese Patent Publication No. 10-332880 (Dec. 18, 1998) discloses a refining device where metal nuclear fuel components are dissolved in cadmium at 500° C. and are deposited on an iron-based cathode again and the uranium electrodeposits are collected by a mechanical scrapping process and transferred to an individual uranium/salt separator to be treated in order to separate the salt included in the electrodeposits.

[0014] Therefore, the refining device can do a continuous operation without stopping the electrolysis reaction in order to collect uranium electrodeposits, thus increasing the processing speed.

[0015] However, since the electrolysis device still uses an iron-based cathode as well, it has the disadvantage of having a mechanical scrapping process.

[0016] Moreover, it has drawbacks that since a pump is employed in order to transfer electrodeposits, a quantity of salts and cadmium are simultaneously transferred and thus an additional distillation process for collecting uranium electrodeposits should be passed.

[0017] In the meantime, the Japanese Patent Publication No. 10-53889 discloses a refining device where a drum-type cathode whose one part is deposited in a molten salt in order to easily collect uranium deposited on the cathode is rotated to separate uranium electrodeposits with a scraper and argon gas is sprayed to the uranium surface deposited on the surface of the cathode drum to remove the remnant salt.

[0018] However, this refining device also needs to be continuously supplied with argon and is accompanied with a mechanical scrapping process. The problems of the conventional devices are not fundamentally solved.

SUMMARY OF THE INVENTION

[0019] Accordingly, the present invention has been made keeping in mind the above problems occurring in the related art, and an object of the present invention is to provide a continuous electrolytic refining device of metal uranium capable of collecting high purity uranium deposits without stopping the electrolysis process and metal transition atoms generated in the process of the electrolysis process.

[0020] In order to achieve the above technical object, the continuous electrolytic refining device of metal uranium of the present invention comprises: a cathode section fixed to the lower side of the heat radiation plate and having a plurality of graphite cathodes; an anode section encompassing the cathode section to face the cathode section, fixed rotatably to the lower side of the heat radiation plate, and receiving the used nuclear fuel; an electrolytic cell accommodating the cathode section and the anode section and filled with electrolytes so as to sink the cathode section and the anode section; an uranium collecting section collecting metal uranium deposited on and detached from the graphite cathode in the lower side of the cathode section inside the electrolytic cell and withdrawing the collected metal uranium to the outside of the electrolytic cell; and a transition metal collecting section coupled with the lower side of the electrolytic cell to withdraw the transition

metal particles released from the anode section and collected in the lower side of the electrolytic cell.

[0021] The cathode section may further comprise a screw-type agitator arranged in the center of the graphite cathodes.

[0022] The anode section has a cylindrical shape encompassing the circumference of the cathode section and may comprise a cylindrical basket receiving a nuclear fuel between an outer circumferential plate and an inner circumferential plate.

[0023] A plurality of outlets are formed on the outer circumferential plate and on the inner circumferential plate, and the outlet may be extendedly formed in the vertical direction and in parallel along the circumference.

[0024] In addition, the outlet may be formed slantingly with a central line of a circle on a horizontal cross-section of the cylindrical basket, more preferably is formed slantingly at 45° with the central line of a circle on a horizontal cross-section of the cylindrical basket.

[0025] The inner circumference plate may be formed in a mesh shape in the range of 100 to 325 mesh.

[0026] The cylindrical basket may be formed of the combinations of a plurality of circular arc-shaped baskets separated individually along a circumference.

[0027] The uranium collecting section may comprise a collecting basin provided in the lower side of the cathode section inside the electrolytic cell; and the first flexible screw conveyor coupled to the collecting basin through the electrolytic cell to withdraw the uranium collected in the collecting basin.

[0028] A cadmium pool with the predetermined level may be provided in the collecting basin and the transfer pipe of the first flexible screw conveyor.

[0029] The first flexible screw conveyor may be provided with the first level sensor for detecting the level of the cadmium pool in the transfer tube thereof.

[0030] The transition metal collecting section may comprise a second flexible screw conveyor coupled to the lower side of the electrolytic cell to withdraw the different collected transition element.

[0031] A cadmium pool with the predetermined level may be formed in the electrolytic cell and the transfer tube of the second flexible screw conveyor.

[0032] A second level sensor for detecting the level of cadmium pool may be provided in the transfer tube of the second flexible screw conveyor.

BRIEF DESCRIPTION OF THE DRAWINGS

[0033] FIG. 1 is a cross-sectional view showing a continuous electrolytic refining device of metal uranium in accordance with an embodiment of the present invention;

[0034] FIG. 2 is a partial cut-off perspective view stereographically showing the device of a continuous electrolytic refining device of metal uranium shown in FIG. 1;

[0035] FIG. 3 is a perspective view separately showing the cathode section of FIG. 2;

[0036] FIG. 4 is a exploded perspective view separately showing the anode section of FIG. 2;

[0037] FIG. 5 is a partial cross-sectional view showing the shape of an outlet of a cylindrical basket of FIG. 4;

[0038] FIG. 6 is a partial cut-off perspective view showing the flow of a material in the continuous electrolytic refining device of metal uranium of FIG. 1; and

[0039] FIG. 7 is a partial cut-off perspective view showing the level of the cadmium pool in the continuous electrolytic refining device of metal uranium of FIG. 1.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

[0040] Hereinafter, embodiments of a continuous electrolytic refining device according to the present invention will be described in detail with reference to the attached drawings for those having ordinary skill in the arts to which the present invention belongs to easily execute. However, the present invention may be realized in various different shapes and is not limited to the embodiments described herein.

[0041] FIG. 1 is a cross-sectional view showing a continuous electrolytic refining device of metal uranium in accordance with an embodiment of the present invention, and FIG. 2 is a partial cut-off perspective view stereographically showing the continuous electrolytic refining device of metal uranium shown in FIG. 1.

[0042] Referring to FIGS. 1 and 2, the continuous electrolytic refining device for metal uranium in accordance with the present embodiment comprises a cathode section (20) fixed to the lower side of a heat radiation plate (10), an anode section (30) receiving the used nuclear fuel (35) and fixed to the lower side of the heat radiation plate (10) so as to rotate around the cathode section (20), an electrolytic cell (40) receiving the cathode section (20) and the anode section (30) and filled with electrolytes to sink the anode section (30) and the cathode section (20), uranium collecting section (50) collecting and withdrawing metal uranium which was deposited onto the lower side of the cathode section (20) and stripped and a transition metal collecting section (60) withdrawing the transition metal sludge collected in the lower side of the electrolytic cell (40).

[0043] FIG. 3 is a perspective view separately showing the cathode section of FIG. 2.

[0044] Referring to FIG. 3, the cathode section (20) includes a disc fixing plate (21) fixed to the lower heat radiation plate (10) with a distance by a connecting member (24), a plurality of graphite cathodes (22) coupled in a bar shape to the lower side of the fixing plate (21), and a screw agitator (23) fixed rotatably in the center of the fixing plate (21) so as to be positioned in the center of the graphite cathodes (22).

[0045] The graphite cathodes (22) can easily separate the metal uranium deposited on the cathode side in an electrolytic refining process of uranium by weight thereof from the molten salt where a used nuclear fuel is molten by electrolytes.

[0046] In addition, it is preferred that the graphite cathode sections (22) be uniformly arranged along the edge of a circumference of the fixing plate (21) so as to increase the area opposed to the anode section (30) which is formed while encompassing the circumference thereof.

[0047] The screw agitator (23) is formed in a screw shape in order to agitate a molten salt inside the electrolytic cell (40) by the first motor (26) provided in the upper side of the heat radiation plate (10), agitating the molten salts with flowing from lower part to upper part by rotation in the center of the graphite cathodes (22).

[0048] In this arrangement, it is preferred that the screw agitator (23) be constituted to interlock with the anode section (30) which will be described later in order not to raise turbulence when agitating the molten salt.

[0049] FIG. 4 is a disassembled perspective view separately showing the anode section shown in FIG. 2.

[0050] Referring to FIG. 4, the anode section (30) includes an anode frame (31) fixed rotatably on the lower side of the heat radiation plate (10), and a cylindrical basket (32) fixed to the anode frame (31).

[0051] The anode frame (31) is fixed to the lower side of the heat radiation plate (10) to rotate the surroundings of the cathode section (20) by the second motor (36) disposed on the upper side of the heat radiation plate (10).

[0052] The cylindrical basket (32) is formed in a cylinder shape so as to be opposite to the graphite cathode (22) with encompassing the surroundings of the cathode section (20).

[0053] The cylindrical basket (32) comprises an outer circumferential plate (32a) and an inner circumferential plate (32b) which are formed with a distance from each other in order to form a receiving space for receiving the used nuclear fuel (35) inside thereof.

[0054] Moreover, it is preferred that the cylindrical basket (32) be dividedly formed into a plurality of circular arc-shaped baskets (33) along the circumferential direction of the anode frame (31). The present embodiment exemplifies that the cylindrical basket (32) is formed by the combination of the circular arc-shaped baskets (33) of quadrants.

[0055] The arc-shaped baskets (33) are coupled to the anode frame (31) without additional connecting members in order to easily do an individual replacement operation.

[0056] As described above, the circular arc-shaped baskets (33) can individually replace only the arc-shaped baskets (33) where the received nuclear fuel (35) is completely dissolved by electrolytes. Therefore, it is possible to perform a continuous electrolytic refining process without withdrawing the entire anode section (30) to the outside of the electrolytic cell.

[0057] Hereinafter, the cylindrical basket (32) refers to each circular arc-shaped basket (33) constituting itself.

[0058] The cylindrical basket (32) is arranged and elongated along the vertical direction of the outer circumferential plate (32a) and the inner circumferential plate (32b) and a plurality of outlets (32c) are formed in parallel to the circumferential direction.

[0059] FIG. 5 is a partial cross-sectional view showing the shape of an outlet of the cylindrical basket shown in FIG. 4.

[0060] Referring to FIG. 5, the outlet (32c) can be formed on both the outer circumferential plate (32a) and the inner circumferential plate (32b) of the cylindrical basket (32), and it is formed so that the molten salt is flown from the inside to the outside of the anode section (30) when the anode section (30) is rotated.

[0061] Moreover, the outlet (32c) is formed slanted to the central line (L) of the cross-sections of each cylindrical basket (32). In the present embodiment, the outlet (32c) is formed slantingly at 45° from the central lines (L) of the cylindrical basket (32), and furthermore, is formed on the slant in the direction where a pathway through which the molten salt is flown and in the direction opposite to the direction of the rotation of the anode section (30).

[0062] Therefore, in the electrolysis reaction process of nuclear fuel, transition metal sludges less than the predetermined sizes, which are not dissolved by electrolytes to remain in the cylindrical basket (32) are discharged through the outlet (32c) of the outer circumferential plate (32a) by the flow of the molten salt.

[0063] In addition, the inner circumferential plate (32b) of the cylindrical basket (32) may be formed of a mesh less than a preset mesh in order to prevent transition metal sludges from being flown into the anode section (30). At this time, it

is preferable that the inner circumferential plate (32b) be formed of a stainless mesh of approximately 100 to 325 mesh.

[0064] FIG. 6 is a partial cut-off perspective view showing the flow of the material inside a continuous electrolytic refining device for metal uranium shown in FIG. 1.

[0065] Referring to FIG. 6, the molten salt in the electrolytic cell (40) is flown and agitated without turbulence by the anode section (30) and the screw agitator (23) which is interlocked with the anode section (30).

[0066] The anode section (30) is rotated to flow the molten salt towards the cathode section (20) located inside thereof, to the outside in order to improve the dissolution of the nuclear fuel (35) received in the cylindrical basket (32), and discharges the transition metal sludges which were not dissolved and remained inside thereof to the outside through the outlet (32c).

[0067] At this time, the discharged transition metal sludges are collected in the lower side of the electrolytic cell (40) by the difference of weights from the molten salt.

[0068] The screw agitator (23) is circulated to the lower side of the graphite cathodes (22) to move the flown molten salts upwardly, and thus the metal uranium dissolved in the molten salt can be deposited onto the graphite cathode more easily.

[0069] FIG. 7 is a partial cut-off perspective view showing the level of a cadmium pool in the continuous electrolytic refining device for metal uranium shown in FIG. 1.

[0070] Referring to FIG. 7, the metal uranium collecting section (50) comprises a collecting basin (51) formed in the lower portion of the cathode section (20) and the first flexible screw conveyor (52) withdrawing metal uranium collected in the lower part of the collecting basin (51) to the outside of the electrolytic cell.

[0071] The collecting basin (51) is formed in a funnel shape in the lower side of the cathode section (20) to collect inside it the metal uranium electrodeposits which were deposited on the graphite cathodes (22) and stripped in itself.

[0072] The first flexible screw conveyor (52) withdraws continuously the metal uranium collected in the lower collecting basin (51).

[0073] In addition, the transition metal collecting section (60) comprises the second flexible screw conveyor (61) coupled in the lower part of the electrolytic cell (40), the second flexible screw conveyor (61) withdrawing continuously the transition metal sludges collected in the lower part of the electrolytic cell.

[0074] Furthermore, the second flexible screw conveyor (61) can be used for the purpose of recovering transition metal sludges and also utilized as a transfer means at the time of exchanging the molten salt.

[0075] However, the metal uranium electrodeposits withdrawn by the first flexible screw conveyor (52) and the second flexible screw conveyor (61), and the transition metal sludges include a remnant molten salt of approximately 20% to 30%.

[0076] The remnant molten salt is mixed with uranium chloride and transuranic chloride, which has a lower vapor pressure than uranium chloride in general.

[0077] Accordingly, the remnant molten salt increases the density in comparison with uranium chloride in the distillation step to react with metal uranium electrodeposits at a high temperature to form transuranic metal plasma and increases the radioactivity level of the collected uranium. It is regarded as a serious problem when uranium is intermediately stored and is wasted in a low level.

[0078] Therefore, the cadmium pool (54) of the predetermined level (H1) is formed in the collecting basin (51) of the metal uranium collecting section (50) and the transfer tube of the first flexible screw conveyor (52).

[0079] The cadmium forming the cadmium pool (54) has a density of 7 g/cm³ smaller than the density of 19 g/cm³ of uranium, and the solubility in the liquid cadmium of uranium metal is 2.3 wt % at 500° C. Accordingly, the metal uranium electrodeposits detached from the graphite cathode (22) sink to the lower part of the cadmium pool (54). The density of LiCl—KCl molten salt is 1.6 g/cm³ at 500° C. and the density of the salt including 9 wt % UCl₃ is also 1.9 g/cm³ or less, which is very low in comparison with liquid cadmium. In addition, the solubility of a molten salt in comparison with liquid cadmium is extremely small. Therefore, the molten salt included in uranium electrodeposits floats above the liquid cadmium. Likewise, the uranium electrodeposits are separated from the molten salt by the cadmium pool (54).

[0080] Furthermore, a small quantity of cadmium is mixed in the uranium electrodeposits drawn out of the electrolytic cell by the first flexible screw conveyor.

[0081] However, cadmium has a low boiling point in comparison with a molten salt and it is possible to easily remove it in a distillation process in comparison with a salt, and most of transuranium salt is also removed in the step of drawing out uranium electrodeposits by the first flexible screw conveyor.

[0082] Accordingly, it is possible to obtain high pure uranium electrodeposits after cadmium is removed.

[0083] Furthermore, a level sensor (53) capable of measuring the level of cadmium pool (54) is provided in the transfer tube of the first flexible screw conveyor (52).

[0084] The level sensor (53) measures the level of cadmium pool by using the difference of conductivity between molten salt/cadmium/empty space to supplement a required amount, if required, in case that a loss of cadmium occurs in accordance with the continuity number of metal uranium.

[0085] Cadmium can be reused by being separated from the metal uranium electrodeposits in the distillation step.

[0086] In addition, cadmium pool is formed inside the transfer tube of the second flexible screw conveyor (61) of the transition metal collecting section (60) and in the lower part of the electrolytic cell (40), and the second level sensor (62) for measuring the level of cadmium pool (63) is provided in the transfer tube of the second flexible screw conveyor (61).

[0087] Therefore, the transition metals have mostly larger particles than those of liquid cadmium and therefore sink downwardly, and pure transition metals can be obtained by the same manner as uranium.

[0088] Although the preferred embodiments of the present invention have been disclosed for illustrative purposes, those skilled in the art will appreciate that various modifications, additions and substitutions are possible, without departing from the scope and spirit of the invention as disclosed in the accompanying claims.

[0089] As described above, the present invention has an effect of continuously collecting metal uranium through an electrolytic refining process using a graphite cathode and a rotatable cylindrical anode section without a mechanical scrapping process.

[0090] Furthermore, the present invention has an effect of continuously collecting metal uranium electrodeposits which are spontaneously stripped from the graphite cathode and

transition metals which are separated and collected by a rotatable cylindrical anode section using each flexible screw conveyor.

[0091] In addition, the present invention has an effect of removing the remnant molten salt present in uranium electrodeposits by using cadmium pool to prevent transuranium elements from being polluted and to collect high purity uranium.

What is claimed is:

1. A continuous electrolytic refining device of metal uranium, comprising:

a cathode section fixed to the lower side of a heat radiation plate and having a plurality of graphite cathodes;

an anode section encompassing the cathode section to face the cathode section, rotatably fixed to the lower side of the heat radiation plate, and receiving the used nuclear fuel;

an electrolytic cell accommodating the cathode section and the anode section inside, and filled with electrolytes so as to sink the cathode section and the anode section;

an uranium collecting section collecting metal uranium deposited on and detached from the graphite cathode in the lower side of the cathode section inside the electrolytic cell and withdrawing the collected metal uranium to the outside of the electrolytic cell; and

a transition metal collecting section coupled with the lower side of the electrolytic cell to withdraw the transition metal particles released from the anode section and collected in the lower side of the electrolytic cell.

2. The device of claim 1, wherein the cathode section further comprises a screw-type agitator arranged in the center of the graphite cathodes.

3. The device of claim 1, wherein the anode section has a cylindrical shape encompassing the circumference of the cathode section and comprises a cylindrical basket receiving a nuclear fuel between an outer circumferential plate and an inner circumferential plate.

4. The device of claim 3, wherein a plurality of outlets are formed on the outer circumferential plate and an inner circumferential plate, and

wherein the outlet is extendedly formed in the vertical direction and in parallel along the circumference.

5. The device of claim 4, wherein the outlet is formed slantingly to a central line of a circle on a horizontal cross-section of the cylindrical basket.

6. The device of claim 5, wherein the outlet is formed slantingly at 45° with the central line of a circle on a horizontal cross-section of the cylindrical basket.

7. The device of claim 3, wherein the inner circumference plate is formed in a mesh shape in the range of 100 to 325 mesh.

8. The device of claim 3, wherein the cylindrical basket is formed of the combinations of a plurality of circular arc-shaped baskets separated individually along the circumference.

9. The device of claim 1, wherein the uranium collecting section comprises a collecting basin provided in the lower side of the cathode section inside the electrolytic cell; and

the first flexible screw conveyor coupled to the collecting basin through the electrolytic cell to withdraw the uranium collected in the collecting basin.

10. The device of claim **9**, wherein cadmium pool with the predetermined level is provided in the collecting basin and the transfer pipe of the first flexible screw conveyor.

11. The device of claim **10**, wherein the first flexible screw conveyor is provided with the first level sensor for detecting the level of the cadmium pool in the transfer tube thereof.

12. The device of claim **1**, wherein the transition metal collecting section comprises a second flexible screw con-

veyor coupled to the lower side of the electrolytic cell to withdraw the collected different transition element.

13. The device of claim **12**, wherein the cadmium pool with the predetermined level is formed in the electrolytic cell and the transfer tube of the second flexible screw conveyor.

14. The device of claim **13**, wherein a second level sensor for detecting the level of the cadmium pool is provided in the transfer tube of the second flexible screw conveyor.

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