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(54) **PROCESS FOR OBTAINING TEREPHTHALIC ACID AND RECOVERING SODIUM HYDROXIDE FROM A SODIUM TEREPHTHALATE SOLUTION PRODUCED FROM REUSED POLYETHYLENE TEREPHTHALATE (PET)**

(57) A process for obtaining terephthalic acid and recovering sodium hydroxide from a sodium terephthalate solution produced from reused PET, which includes the following steps: i) placing a sodium terephthalate solution obtained from a PET hydrolysis process on the anodic side; ii) placing an alcoholic medium on the cathodic side of the electrolytic cell; iii) applying an electric current that circulates through the anodic side and the cathodic side of the electrolytic cell and; iv) recovering terephthalic acid and sodium hydroxide. The method has been designed to not generate the by-products generated in the PET recycling process, and thus have a "green" process. Furthermore, the method described does not require large amounts of water or energy.

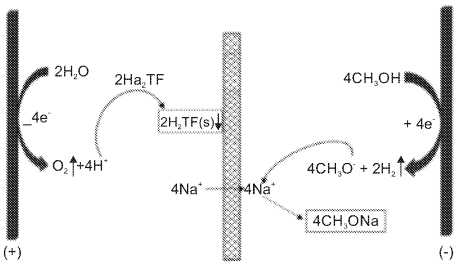

FIG. 1

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Description

FIELD OF THE INVENTION

[0001] The present invention is related to obtaining terephthalic acid, and more particularly it is related to a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET.

BACKGROUND OF THE INVENTION

[0002] In the chemical recycling process of polyethylene terephthalate, better known as PET, by-products are generated, and a large amount of sodium hydroxide (NaOH) is consumed. An example of this is described in document MX271258, in which ethylene glycol and terephthalic acid are generated as products and sodium sulfate as a by-product.

[0003] Terephthalic acid has various applications in the chemical, cosmetic, and even pharmaceutical industries. The most common way to obtain terephthalic acid is from oil. However, this way of obtaining it is not a good long-term viable option, because oil is a non-renewable resource. On the other hand, sodium hydroxide (NaOH) also has many applications in various industries, mainly in the chemical industry. It is known that sodium hydroxide can be obtained electrolytically using ion exchange membranes from aqueous solutions of sodium chloride. However, this industrial process typically only produces solutions of between 20 and 40 % sodium hydroxide and requires more operations to obtain it in a solid state.

[0004] Therefore, there is a need to develop a method that allows recovering the reagents consumed in the PET recycling process, such as sodium hydroxide.

[0005] Among the solutions proposed in the state of the art is document JP2004250544, which refers to a process to recover terephthalic acid from a terephthalic acid polyester by electrolysis. The method uses an alcoholic solution of ethylene glycol, sodium sulfate and water as catholyte for electrolysis. However, this document does not consider the recovery of sodium hydroxide from a sodium terephthalate solution, so it is not suitable as a complement to the PET recycling process.

[0006] Document JP11302208 describes a method for producing terephthalic acid and ethylene glycol from PET. The method involves putting PET in contact with sodium hydroxide in ethylene glycol and subsequently separating the sodium terephthalate produced. However, the method described in document JP11302208 requires reaching high temperatures of between 170 °C to 180 °C, which increases the cost of the process to recycle PET, because a large amount of energy and special equipment are needed for these temperatures to be reached, in addition to the fact that some acid is used to obtain terephthalic acid, for example sulfuric acid, thus obtaining sodium sulfate as a by-product.

[0007] Because of the above, we have sought to eliminate the drawbacks presented by the methods for obtaining terephthalic acid currently used, developing a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET, without using large amounts of water and energy.

OBJECTS OF THE INVENTION

[0008] Considering the defects of the prior art, it is an object of the present invention to provide a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from PET that allows the recovery of the hydroxide of sodium that is normally consumed in the PET recycling process.

[0009] Another object of the present invention is to provide a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET that does not need to use large amounts of water and electricity.

[0010] Finally, another object of the present invention is to provide a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET that does not generate many byproducts.

[0011] These and other goals are achieved through a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET according to the present invention.

SUMMARY OF THE INVENTION

[0012] A method of obtaining terephthalic acid and ethylene glycol and recovering sodium hydroxide consumed in the PET recycling process which does not require large amounts of water or energy has been invented. Thus, the present invention refers to a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET, which comprises the following steps:

i) placing a sodium terephthalate solution obtained from a PET hydrolysis process on the anodic side in an electrolytic

cell;
ii) placing an alcoholic medium on the cathodic side of the electrolytic cell;
iii) applying an electric current that circulates through the anodic side and the cathodic side of the electrolytic cell and;
iv) recovering terephthalic acid and sodium hydroxide.

BRIEF DESCRIPTION OF THE DRAWINGS

[0013] The novel aspects that are considered characteristic of the present invention will be particularly established in the attached claims. However, some embodiments, characteristics, and some objects and advantages thereof will be better understood in the detailed description, when read in relation to the attached drawing, in which:
Figure 1 shows a scheme of the chemical reaction of a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET within the electrolytic cell in accordance with the present invention.

DETAILED DESCRIPTION OF THE INVENTION

[0014] A process has been found to obtain terephthalic acid and recover sodium hydroxide from a solution of sodium terephthalate produced from reused PET, which does not require large amounts of water or energy.
[0015] Thus, one aspect of the present invention is a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET, which comprises the following steps:

i) placing a sodium terephthalate solution obtained from a PET hydrolysis process on the anodic side in an electrolytic cell;
ii) placing an alcoholic medium on the cathodic side of the electrolytic cell;
iii) applying an electric current that circulates through the anodic side and the cathodic side of the electrolytic cell and;
iv) recovering terephthalic acid and sodium hydroxide.

[0016] In a preferred embodiment of the present invention, the alcoholic medium placed on the cathodic side of the electrolytic cell is methanol, ethanol, propanol, butanol, or a mixture thereof. Preferably, sodium hydroxide is added to the alcoholic medium placed on the cathodic side of the electrolytic cell to obtain an alcoholic solution. Preferably, the alcoholic solution with sodium hydroxide has a molarity of 0.25 M to 1 M.
[0017] In another preferred embodiment of the present invention, the electrolytic cell has a membrane or a diaphragm between the anodic side and the cathodic side. Preferably, the electric current applied to the electrolytic cell has a density of between 0.1 to 0.5 Amperes/cm^2. Preferably, the applied electrical current is measured continuously to determine the number of Coulombs passing through the electrodes. Preferably, electric current is applied to the electrolytic cell for a period of 50 to 70 minutes. Preferably, after the period the electrodes are removed. However, a cell could be designed that worked continuously.
[0018] In a preferred embodiment of the present invention, a solution of terephthalic acid is obtained from the anodic side of the electrolytic cell and sodium hydroxide and sodium alkoxide in an alcoholic solution are obtained from the cathodic side of the electrolytic cell. Preferably, the solution from the anodic side, which contains precipitated terephthalic acid, is filtered. Preferably, the water-washed terephthalic acid is oven dried at a temperature of between 125 to 135°C. Preferably, the terephthalic acid is dried in the oven for 50 to 70 minutes. Preferably, the percentage of electronic efficiency of recovery of terephthalic acid relative to sodium terephthalate solution is 50 to 85 %. Preferably, the sodium alkoxide is in an alcoholic solution and in a concentration of between 5 % to 30 % (m/V). In case of alkoxide formation, sodium hydroxide is obtained by adding the stoichiometric amount of water. Solid sodium hydroxide is obtained by evaporation of the corresponding alcohol. It should be noted that the production of terephthalic acid and sodium hydroxide occurs simultaneously.
[0019] For a better understanding, Figure 1 shows a diagram of the chemical reaction of the process to obtain terephthalic acid and recover sodium hydroxide from a solution of sodium terephthalate produced from reused PET within the electrolytic cell according to the present invention. Figure 1 shows the anodic side, that is, the left side of figure 1 and the cathodic side, that is, the right side of figure 1 seen frontally. Initially, in the electrolytic cell, an aqueous solution of sodium terephthalate is introduced on the anodic side of said electrolytic cell and an alcoholic medium with sodium hydroxide is introduced on the cathodic side. Then, a reaction occurs in which, from the oxidation of water at the anode, the protons produced react with the terephthalate ion, terephthalic acid (H_2TF) is precipitated and oxygen (O_2) is released. Simultaneously, on the cathodic side, sodium alkoxide (RONa) and hydrogen gas (H_2) are produced.
[0020] The present invention will be better understood from the following examples, which are presented solely for illustrative purposes to allow a full understanding of the preferred embodiments of the present invention, without implying that there are no other non-illustrated embodiments that can be put into practice based on the detailed description above.

EXAMPLE 1

[0021] A test was conducted to recover terephthalic acid and sodium hydroxide from a sodium terephthalate solution obtained from a PET hydrolysis process according to the present invention.

[0022] In the test, 90 mL of sodium terephthalate solution were added in the anodic side in an electrolytic cell. The sodium terephthalate solution was prepared with 12 g of sodium terephthalate and 100 mL of water. On the cathodic side of the electrolytic cell, 90 mL of a solution of methanol with sodium hydroxide were added. The solution was prepared with 1 g of NaOH and 100 mL of methanol.

[0023] Then, a platinum electrode was placed on the anodic side and a stainless-steel electrode on the cathodic side and the electrolytic cell was connected to a power source. The power source was turned on and the current was controlled and maintained for one hour at an electrical intensity of 0.4 to 0.6 Amperes. The electrolytic cell was connected to an ammeter that continuously measured and recorded the current that passed through the electrodes and in this way the amount of electricity in Coulombs that passed through the electrodes of the electrolytic cell was determined. After one hour the power source was turned off, the electrodes from each side of the electrolytic cell were removed and the solution from the anodic side was filtered, which contained precipitated terephthalic acid, sad precipitate was washed and the terephthalic acid washed, dried in the oven at 130° C for one hour and then weighed. On the cathodic side sodium methoxide was obtained. Adding water to this solution produces sodium hydroxide in methanol. Solid sodium hydroxide is obtained by evaporation of the corresponding alcohol.

[0024] During the test, the temperatures of the two half-cells were measured, as well as the variations in current between the two electrodes of the electrolytic cell, to know the efficiency of the precipitated terephthalic acid obtained.

[0025] Table 1 shows the results obtained:

TABLE 1

Initial current (A)	Time (min)	Terephthalic acid weight (g)	Amount of electricity (Coulombs)	Electronic efficiency (%)
0.4252	60	0.331	1253	30.7
0.5397	60	0.70	1549	52.5
0.530	60	0.784	1411	64.6
0.521	60	0.837	1794	65.7

[0026] As can be seen in the results obtained, the amount of terephthalic acid obtained is good considering the amount of mL of sodium terephthalate solution with which the reaction began. Furthermore, the mL of water used and the electrical energy necessary for the reaction represent a very low cost in relation to the cost at which terephthalic acid and sodium hydroxide can be sold.

EXAMPLE 2

[0027] A test was conducted using different reaction times in a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET, according to the present invention. The purpose of this test is to see if there is variation in the efficiency of the process. From the results obtained, there is no significant difference.

[0028] The experiments were conducted in the same cell and under the same conditions as in Example 1, except that there was variation in the electrolysis duration times. Table 2 shows the results obtained.

TABLE 2

Time (hr)	Charge quantity (Coul)	Weight of the terephthalic acid obtained (g)	Electronic efficiency (%)
1	1753	0.93	68
2	2877	2.13	86
2	2193	1.45	79
3	4860	2.66	64

[0029] As can be seen in the previous table, the electrolysis that lasted the longest was the one in which a greater

amount of terephthalic acid was obtained.

EXAMPLE 3

[0030] A test was conducted using different electrical currents in a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET, according to the present invention. The purpose of this test is to observe variation in the efficiency of obtaining terephthalic acid. The experiments were conducted in the same cell and under the same conditions as in Example 1, except that there was variation in the amount of current during electrolysis. Table 3 shows the results obtained.

TABLE 3

Electric current (A)	Time (hr)	Weight of the terephthalic acid obtained (g)	Electronic efficiency (%)
0.6	1	1.26	68
0.36	1	0.69	62
0.49	1	0.75	63
1.7	1	2.74	52

[0031] According to the above, the process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET has been designed to not generate the by-products generated in the PET recycling process. Furthermore, the described method does not require large amounts of water or energy, and practically does not generate by-products, and it will be evident to any person skilled in the art that the embodiments of a process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET as described above and illustrated in the drawing that accompanies it, are only illustrative but not limiting of the present invention, since numerous considerable changes in its details are possible without departing from the scope of the invention.

[0032] Therefore, the present invention should not be considered restricted except as required by the prior art and by the scope of the appended claims.

Claims

1. A process for obtaining terephthalic acid and recovering sodium hydroxide from a sodium terephthalate solution produced from reused PET **characterized in that** it comprises the following steps: i) placing a sodium terephthalate solution obtained from a PET hydrolysis process on the anodic side; ii) placing an alcoholic medium on the cathodic side of the electrolytic cell; iii) applying an electric current that circulates through the anodic side and the cathodic side of the electrolytic cell and; iv) recovering terephthalic acid and sodium hydroxide.

2. The process for obtaining terephthalic acid and recovering sodium hydroxide from a sodium terephthalate solution produced from reused PET according to claim 1, further **characterized in that** the alcoholic medium placed on the cathodic side of the electrolytic cell is methanol, ethanol, propanol, butanol, or a mixture thereof.

3. The process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET according to claim 1, further **characterized in that** in the alcoholic medium placed on the cathodic side of the cell electrolytic, sodium hydroxide is added to obtain an alcoholic solution.

4. The process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET according to claim 3, further **characterized in that** the alcoholic solution with sodium hydroxide has a molarity of 0.25 M to 1M.

5. The process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET according to claim 1, further **characterized in that** the electrolytic cell has a membrane or a diaphragm between the anodic side and the cathodic side.

6. The process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET in accordance with claim 1, further **characterized in that** the electric current applied to

the electrolytic cell has a density of between 0.1 to 0.5 Amperes/cm^2.

7. The process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET in accordance with claim 1, further **characterized in that**, on the anodic side of the electrolytic cell, a solution of sodium terephthalic acid is obtained, and on the cathodic side of the electrolytic cell, sodium hydroxide and sodium alkoxide in an alcoholic solution are obtained.

8. The process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET in accordance with claim 7, further **characterized in that** the solution on the anodic side, which contains precipitated terephthalic acid, is filtered.

9. The process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET according to claim 7, further **characterized in that** the terephthalic acid washed with water is dried in the oven at a temperature of between 125 to 135 °C.

10. The process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET in accordance with claim 7, further **characterized in that** the percentage of electronic efficiency of the recovery of terephthalic acid is 50 to 85 %.

11. The process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET in accordance with claim 7, further **characterized in that** the sodium alkoxide is in an alcoholic solution and in a concentration between 5 % and 30 % (m/V).

12. The process for obtaining terephthalic acid and recovering sodium hydroxide from a solution of sodium terephthalate produced from reused PET in accordance with claim 7, further **characterized in that** the obtaining of terephthalic acid and sodium hydroxide is produced simultaneously.

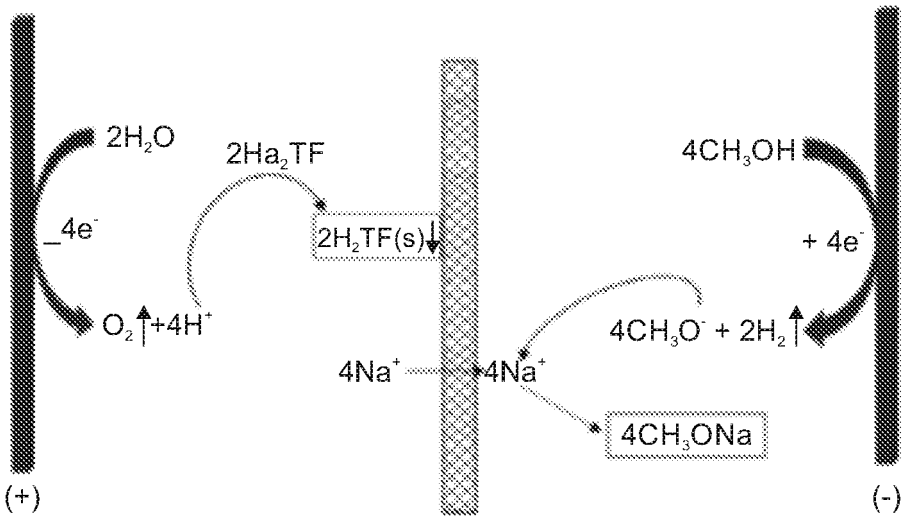
2H2O
2Ha2TF
4CH3OH
_4e-
2H2TF(s)
+ 4e-
O2+4H+
4CH3O- + 2H2
4Na+
4Na+
4CH3ONa
(+)
(-)

FIG. 1

INTERNATIONAL SEARCH REPORT

International application No.
PCT/IB2021/055081

A. CLASSIFICATION OF SUBJECT MATTER

C25B1/16, C25B3/00 (2022.01)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

(CIP) C25B1/16, C25B3/00 (CPC) C07C51/02, C25B1/16, C25B3/00

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

Derwent, Esp@cenet, Patenscope, Google Patent, Google, INAPI

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 6,312,582 B1 (TREDI) 06-11-2001 The whole document	1-12
A	US 4,093,528 (SUNTECH) 06-06-1978 The whole document	
A	US2010038231 (A1) 18-02-2010 The whole document	
A	FR2697839A1 (INST FRANCAIS DU PETROLE) 13-05-1994 The whole document	

[X] Further documents are listed in the continuation of Box C. [X] See patent family annex.

* Special categories of cited documents:

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
12 February 2022 (12.02.2022)	24 February 2022 (24.02.2022)

Name and mailing address of the ISA/	Authorized officer
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INTERNATIONAL SEARCH REPORT

International application No.
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C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP0573042A1 (MITSUI TOATSU CHEMICALS) 08-12-1993 The whole document	
A	US5290404 A (REILLY IND INC) 01-03-1994 The whole document	

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- JP 11302208 B **[0006]**