(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau

(43) International Publication Date
14 April 2011 (14.04.2011)

PCT

(10) International Publication Number
WO 2011/043515 A1

(51) International Patent Classification:
*B01J 19/00* (2006.01) *B01J 19/24* (2006.01)
*C08J 11/04* (2006.01)

(21) International Application Number:
PCT/KR2009/007667

(22) International Filing Date:
22 December 2009 (22.12.2009)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
10-2009-0096032 9 October 2009 (09.10.2009) KR

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(81) Designated States *(unless otherwise indicated, for every kind of national protection available)*: AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PE, PG, PH, PL, PT, RO, RS, RU, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

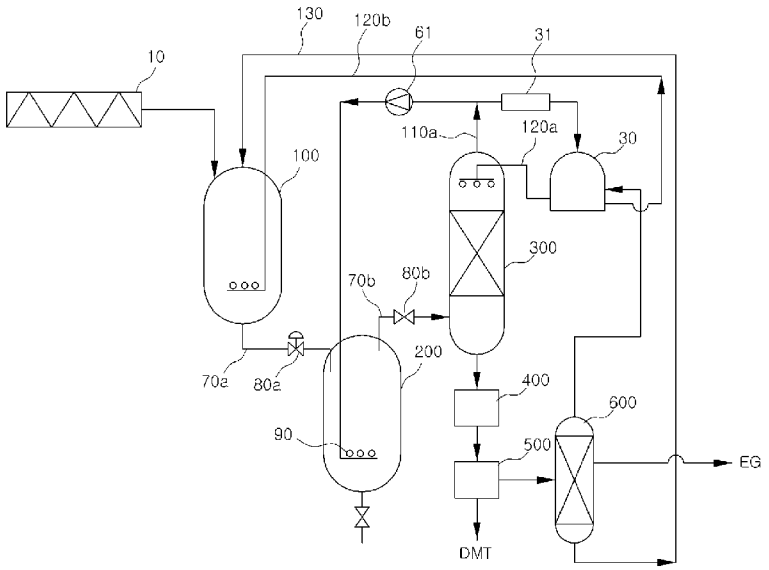
(84) Designated States *(unless otherwise indicated, for every kind of regional protection available)*: ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

**Published**:

— *with international search report (Art. 21(3))*

(54) **Title**: FEEDSTOCK RECYCLING PROCESS FROM POLYESTER WASTES AND APPARATUS USING THE SAME

[Fig. 1]




(57) **Abstract**: The present invention relates to a process for recycling feedstock from polyester wastes and an apparatus using the same, and more particularly, to a process for recycling dimethyl terephthalate and ethylene glycol or feedstock for polyester from polyethyleneterephthalate (PET) and an apparatus using the same. More particularly, the present invention relates to a novel continuous recycling process for improving a process efficiency and energy saving effect through recycling of produced methanol vapor and crystallization of dimethyl terephthalate.

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# Description

# Title of Invention: FEEDSTOCK RECYCLING PROCESS FROM POLYESTER WASTES AND APPARATUS USING THE SAME

## Technical Field

[1] The present invention relates to a process for recycling feedstock from polyester wastes and an apparatus using the same, and more particularly, to a process for recycling dimethyl terephthalate (hereinafter, referred to as DMT) and ethylene glycol (hereinafter, referred to as EG) or feedstock for polyester from polyester wastes (bottles, magnetic films, beer bottles, oligomer sludges and so on) mainly containing polyethylene terephthalate (hereinafter, referred to as PET) and an apparatus using the same.

## Background Art

[2] Nowadays, as plastic products are flooded with development of plastic industry, disposal of wastes becomes a severe environmental problem.

[3] The disposal of the plastic wastes largely employs landfill, incineration and recycle, and it is preferred to recycle the plastic wastes since the landfill and the incineration cause a problem such as environmental pollution. In general, the recycle can be divided into material recycle and chemical recycle. First, in the material recycle, generated plastic wastes are collected and sorted and then physically recycled. However, the material recycle has a problem that the recycled plastic has low quality though it is economic in recycle cost.

[4] In the chemical recycle, plastic wastes are depolymerized and monomers or polymerization feedstock for the plastic are obtained. In one example, there is recycle of PET wastes of plastic wastes and a conventional process includes the following processes:

[5] a depolymerization process of putting polyester wastes into ethylene glycol including depolymerizing catalyst in a glycolysis reactor to depolymerize the polyester wastes into bis-hydroxyl terephthalate (BHET) and oligomer, and putting the partially glycolisized product into a transesterification reactor, producing crude DMT and EG through a transesterification of the partially glycolisized product in methanol (MeOH) in the presence of a transesterification catalyst and obtaining final products of DMT and EG through purification of the crude DMT and EG using distillation and crystallization process with recycle of the methanol to the transesterification reactor.

[6] With respect to the transesterification reactor in aforementioned process, US Patent No. 5,051,528 discloses that super-heated methanol gas are passed through oligomers of ethylene glycol and terephthalic acid or dimethyl terephthalate at a low pressure and

gaseous DMT, EG and methanol are recovered overhead to thereby be able to treat more contaminated PET. That is, BHET and oligomers are obtained by the partial glycolysis and DMT and EG are obtained by methanolysis of the BHET and oligomers. However, the depolymerization into BHET by the partial glycolysis takes a long reaction time since this is a reversible reaction.

[7] Also, the obtained BHET and oligomer is depolymerized again by gaseous methanol to form DMT and EG which should be discharged in gaseous phase. Therefore, high pressure methanol cannot be used and the reaction speed is thus slow.

[8] Various methods for PET methanolysis are disclosed in patent documents. Operation is conducted in a batch and continuous conditions, and a major problem of the continuous process is that it is difficult to feed solid polyester wastes to a methanolysis reactor operating at high pressure. Due to the aforementioned reason, the methanolysis is often conducted in the batch process despite of various disadvantages of the batch process. A high pressure batch process is known that the depolymerization of the polyester is generated fast as a concentration of the methanol in the oligomer solution is maintained high but the reaction speed is slowed with lapse of time due to reaction equilibrium limit by increase in concentrations of produced DMT and EG, and thus the polyester cannot be completely depolymerized into DMT and EG and about 15% remains in the form of oligomer.

[9] On the contrary, a low pressure continuous depolymerizing process is not limited by the reaction equilibrium since DMT and EG are removed through the top of the reactor, but has a disadvantage that the reaction speed is slow since the concentration of the methanol in the reaction solution is maintained low.

[10] Korean patent No. 0837781 which is a prior application of the present applicant discloses a method in which the glycolysis reaction and the methanolysis reaction are conducted simultaneously in a single reactor and a single high pressure reactor thus acts as the polyester dissolving tank, the glycolysis reactor and the methanolysis reactor. The aforementioned process could increase the reaction speed notably and obtain effect of decreasing a usage amount of the feedstock methanol due to increase in reaction efficiency.

[11] However, in the methanolysis reactor and methanol recovery process in which the subsequent process is conducted, it is inevitable to conduct a process, which requires a complex apparatus and consumes large energy, of liquefying the methanol vapor from the methanolysis reactor, injecting the methanol or the feedstock from a methanol reservoir by a high pressure pump and vaporizing the methanol by a vaporizer.

[12] Therefore, the aforementioned process has disadvantages of a complex apparatus and consuming relatively large energy since liquefaction and vaporization are repeated as the methanol collected in a methanol recovery tower is liquefied to be stored in the

methanol reservoir and then vaporized again through the vaporizer to be fed into the methanolysis reactor.

## Disclosure of Invention

## Technical Problem

[13] Embodiments of the present invention are directed to providing a process of recycling feedstock from polyester wastes such as bottles, magnetic films, oligomer sludges and so on, which can simplify the apparatus, save energy and obtain high purity monomer DMT.

[14] More specifically, an embodiment of the present invention is first directed to providing a process, in which methanol is separated in a rectification column (300) from a product produced in a methanolysis reactor, a second reactor (200) and recycled to the second reactor (200) without condensing or re-vaporization, thereby notably saving the enormous energy consumed by repeated vaporization and liquefaction of recycled methanol and simplifying the apparatus.

[15] Second, another embodiment of the present invention is directed to provide a process which directly liquefy DMT and EG but vaporizes only the methanol in the solution by directly contacting a gaseous reaction product discharged through the top of the second reactor (200) with the liquid pool at the bottom of the rectification column (300) e.g. through dipping, and liquid in the tray or column section of the column which flows downward from the methanol reservoir (30), thereby enabling the recycle of high purity methanol to the second reactor (200) and using it in the methanolysis reaction.

[16] Third, further another embodiment of the present invention is directed to provide a process which can regulate a feed amount of the methanol in mixing the methanol separated from gaseous reaction product discharged through the top of the second reactor (200) and a feedstock methanol fed from the methanol reservoir (30) and recycling the mixture to the second reactor (200).

[17] Fourth, further another embodiment of the present invention is directed to provide a process in which the methanol fed to the second reactor (200) is provided by the methanol separated in the rectification column (300) and thus there is no need for existing methanol vaporizer, heater and high pressure pump and the rectification column functions their roles.

[18] Fifth, yet another embodiment of the present invention is directed to provide a process which vaporizes the methanol alone by controlling a temperature in the lower portion of the rectification column (300) to facilitate the subsequent crystallization. Sixth, yet another embodiment of the present invention is directed to provide a process which can freely regulate reaction and rectification conditions so that some of the methanol is stored in the methanol reservoir using a back-pressure regulator (31) when

the amount of the methanol discharged through the top of the rectification column (300) is excessive, thereby improving productivity.

## Solution to Problem

[19] The advantages, features and aspects of the invention will become apparent from the following description of the embodiments with reference to the accompanying drawings, which is set forth hereinafter.

[20] Unless defined otherwise, all technical and scientific terms used herein have the same meanings as commonly understood by one of ordinary skill in the art to which the present invention belongs. In other instances, well known functions and structures have not been described in detail in order not to unnecessarily obscure the present invention.

[21] Fig. 1 is a schematic diagram showing an overall process of an apparatus for recycling feedstock, DMT and EG, from polyester wastes, and the apparatus in accordance with the present invention includes a feedstock feeder (10); a first reactor (100) for conducting glycolysis reaction and methanolysis reaction in a single reactor using the feedstock fed from the feedstock feeder (10); a second reactor (200) for subjecting product of the first reactor (100) to methanolysis reaction; a rectification column (300) for separating methanol from gas discharged through a top of the second reactor (200) and recycling the separated gaseous methanol to the second reactor (200); and a methanol reservoir (30) for supplying the methanol to the rectification column (300).

[22] Each element will be described in detail.

[23] The feedstock feeder (10) of the present invention is a feedstock supplier and can employ an extruder, a screw feeder or an air lock valve, but not limited thereto.

[24] The first reactor (100) of the present invention is a glycolysis/methanolysis reactor in which glycolysis reaction and methanolysis reaction take place at the same time, requires no separate dissolving tank as DMT and oligomer are produced therein, and can increase the depolymerization speed notably.

[25] The second reactor (200) subjects the reactant transferred from the first reactor (100) to methanolysis with the methanol supplied from a bubble sparger (90) at a lower portion of the second reactor and discharges gaseous reaction product simultaneously with completion of the depolymerization, thereby capable of treating low-grade polyester wastes containing many impurities.

[26] Also, the rectification column (300) contacts the product of the second reactor (200) directly to liquid pool at the bottom of the rectification column (300) and liquid in the tray or column section of the column which flows downward from the methanol reservoir (30), to vaporize and separate the methanol alone and also recycles a mixture of the separated methanol and feedstock methanol supplied from the methanol

reservoir (30) to the upper portion of the rectification column (300) and vaporized to the second reactor (200).

[27] Further, the methanol reservoir (30) for supplying the methanol to the rectification column (300) also supplies the methanol to the first reactor (100) upon starting of the operation.

[28] The apparatus for recycling feedstock from polyester wastes in accordance with the present invention may further include a back-pressure regulator (31) to control the pressure of the rectifier as well as the methanolysis reactor (200) and the vapor methanol from the back pressure regulator are condensed and stored in the methanol reservoir (30); a crystallization tank (400) for crystallizing DMT from the liquid in the rectification column (300); a solid-liquid separator (500) for separating the crystallized DMT; a distillation column (600) for separating the methanol from the EG-rich liquid methanol in an upper portion thereof to store the separated methanol in the methanol reservoir (30) and separating oligomer from the EG-rich liquid methanol in a lower portion thereof to recycle the separated oligomer to the first reactor (100).

[29] Further, the apparatus of the present invention may further includes, between the feedstock feeder (10) and the first reactor (100), a scrap middle reservoir for weighing the feedstock supplied from the feedstock feeder and supplying the weighed feedstock to the reactor.

[30] Further, a transfer pipe (70a) for transfer to the second reactor (200) is further provided with a sensor for sensing a liquid level in the second reactor (200), and a level controller senses the liquid level using the sensor and regulates the liquid level to automatically open a valve (80a) when the liquid level is lowered.

[31] The pressure in the second reactor (200) and the rectification column (300) can be regulated using the back-pressure regulator (31) or other pressure regulating valve by manipulating an outflow of some of the methanol at the top of the rectification column.

[32] Further, it is preferred that a condenser for condensing the methanol is further provided in an upper portion of the methanol reservoir (30) to collect the some methanol to the methanol reservoir (30) when the amount of the methanol discharged through the top of the rectification column (300) is excessive.

[33] Next, the present invention provides a process for recycling DMT and EG, feedstock of polyester from PET through partial glycolysis and methanolysis reactions and purification.

[34] Specifically, a process for recycling DMT and EG, polyester feedstock from polyester wastes in accordance with the present invention includes: putting feedstock and a mixture of predetermined amount of EG or oligomers from the distillation column (600) and depolymerizing catalyst in a first reactor (100) and depolymerizing the feedstock by increasing a pressure in the reactor with continuous supply of

methanol; transferring reactant from the first reactor (100) by opening a transfer valve (80a) of a transfer pipe (70a) connected to a second reactor (200) when the depolymerization is completed; transferring gaseous reaction product produced in the second reactor (200) to a rectification column (300) and separating pure methanol in an upper portion of the column and liquefying the rest methanol, DMT and EG in a lower portion of the column; recycling the methanol separated in the upper portion of the rectification column (300) to the first reactor (100) and storing some of the methanol in a methanol reservoir (30); transferring the liquid flowed out from the lower portion of the rectification column (300) to a crystallization tank (400) to crystallize DMT; transferring product from the crystallization tank (400) to a solid-liquid separator (500) to separate DMT crystals and EG-rich liquid methanol; transferring the EG-rich liquid methanol to a distillation column (600) to separate EG, oligomer and methanol and storing the methanol in the methanol reservoir (30) and recycling the oligomer to the first reactor (100).

[35] The first reactor (100) and the second reactor (200) are connected by a transfer pipe (70a) so that the reactant is transferred by a pressure difference. More specifically, when a liquid level in the second reactor (200) is lowered a transfer valve (80a) of the transfer pipe (70a) is automatically opened and the reactant is transferred from the first reactor (100) to the second reactor (200) by the pressure difference.

[36] Also, the first reactor (100) is operated at a temperature of 200 to 300 ℃ and a reaction pressure of an atmospheric pressure to 50 bar and the second reactor (200) is operated at a temperature of 200 to 300 ℃ and a reaction pressure of an atmospheric pressure to 10 bar so that the reactant is automatically transferred.

[37] Furthermore, the EG separated in the distillation column (600) can be used as the feedstock which can be put in the first reactor (100).

[38] More specifically, the feedstock used in the process of the present invention is polyester wastes and is fed to the first reactor (100) by a feedstock feeder (10) such as an extruder, a screw feeder and an air lock valve. The polyester wastes can be fed in a solid form from a powder with a suitable size to a flake or a melt form. The first reactor (100) can be operated in continuous, semi-continuous or batch type, and, by using the reactor, the polyester having a large molecular weight can be depolymerized to BHET, MHET or oligomer with considerably reduced molecular weight through glycolysis reaction.

[39] Also, since methanolysis reaction is conducted parallel with the glycolysis reaction, a considerable amount of DMT is produced in the first reactor (100). In this process, since EG produced by methanolysis can be used again in the glycolysis, the two reactions show synergy with each other. Further, the EG produced as such is used to dissolve newly supplied PET feedstock and thus a feed amount of EG required for the

glycolysis can be considerably reduced or the EG are not required to be fed.

[40] When EG including the catalyst and oligomer is fed to the first reactor (100), the glycolysis and methanolysis reactions are generated simultaneously to reduce the polyester wastes considerably and thus conversion into DMT and EG in the second reactor (200), the subsequent process is generated easily and quickly. The pressure in the first reactor (100) can be regulated by adjusting an amount of methanol fed from the methanol reservoir (30) to the first reactor (100) through the transfer pipe (120b).

[41] The first reactor (100) and the second reactor (200) are connected by a transfer pipe (70a) so that the reactant is transferred by a pressure difference. More specifically, when a liquid level in the second reactor (200) is lowered a transfer valve (80a) of the transfer pipe (70a) is automatically opened and the reactant is transferred from the first reactor (100) to the second reactor (200) by the pressure difference.

[42] To the second reactor (200) of the present invention, the methanol is fed to the lower portion of the reactor through a bubble sparger (90). The methanolysis reaction is generated while the methanol fed to the lower portion and the reactant transferred from the first reactor (100) contacts directly with each other, to thereby discharge gaseous reaction product through the top of the second reactor (200). In this process, the methanol functions not only as the reaction material but also as a carrier to discharge the reaction product together with the methanol vapor itself.

[43] The reaction product produced in the second reactor (200) is transferred to the rectification column (300) while the transfer valve (80b) of the transfer pipe (70b) is opened, and contacts with liquid, i.e. a solution of DMT, EG and methanol, at the bottom of the rectification column (300). Therefore, the methanols alone is separated at the top of the column and the methanol, DMT and EG are liquefied in the lower portion of the column, which makes the subsequent crystallization process advantageous. The separation of the methanol is achieved as downwardly flowing liquid and upwardly flowing gaseous methanol come in contact with each other in a tray or packing while refluxing with supply of feedstock methanol. With the aforementioned process, it is possible to improve an efficiency of separation of the methanol from the other products.

[44] The methanol vapor is recovered at the top of the rectification column (300) and most of the recovered methanol vapor is recycled to a low pressure continuous methanolysis reactor, the second reactor (200), through a compressor (61) or a blower.

[45] The pressure in the second reactor (200) and the rectification column (300) can be regulated with the back-pressure regulator (31) or other pressure regulating valve by adjusting an outflow of some of the methanol vapor recovered at the top of the rectification column (300). Also, when the amount of the methanol discharged through the top of the rectification column (300) is excessive, some methanol vapor can be flowed

out by the back-pressure regulator (31) and can be condensed and stored in the methanol reservoir (30).

[46] Therefore, the present invention allows saving of larger energy than existing processes which vaporize the feedstock methanol supplied from the methanol reservoir (30) and feeds the vaporized methanol to the depolymerization reactor. Further, DMT and EG are concentrated in the lower portion of the rectification column (300) as the methanol is recovered at the top of the rectification column (300), and this aids the subsequent crystallization process.

[47] The DMT and EG flowed out from the rectification column (300) are transferred to the crystallization tank (400) and subjected to crystallization process. DMT is crystallized by concentration or cooling and then moved to the solid-liquid separator (400). In the solid-liquid separator, DMT crystals and EG-rich liquid methanol are separated using a centrifuge or a filter. DMT obtained through the above process has advantages that a recovery efficiency of DMT is considerably high and it can be obtained in high purity.

[48] The EG-rich liquid methanol is a liquid containing excessive amounts of EG and methanol and also contains a small amount of oligomer.

[49] Successively, the EG-rich liquid methanol is transferred to the distillation column (600) and is subjected to a subsequent separation process to be separated into EG, oligomer and methanol. At this time, the distillation column (600) can be operated in continuous type or batch type. The methanol separated in the distillation column (600) is stored in the methanol reservoir (30), oligomer can be recycled to the first reactor (100) through a transfer pipe (130), and EG is stored in an EG reservoir.

## Advantageous Effects of Invention

[50] In accordance with the present invention, it is possible to separate the gaseous methanol in the rectification column (300) by contacting the methanolysis product from the methanolysis reactor (200) directly with the liquid pool at the bottom of the column to liquefy DMT and EG and vaporizing the feedstock methanol as a reflux in the rectification column (300) from the methanol reservoir. Also, as the separated gaseous methanol in the rectification column is recycled to the second reactor (200) through the circulating compressor (61) or the blower and comes in direct contact with a polyester solution in the inside of the reactor through the bubble sparger (90) in the lower portion of the second reactor (200) to generate the methanolysis reaction, it is possible to notably save the cost for energy caused by repeated vaporization and liquefaction in the existing methanol recycle process. Further, as it is not necessary to use a methanol vaporizer, a heater and a high pressure pump for the existing methanol recycle process, it is possible to simplify the apparatus and notably save the cost for the

apparatus.

[51] Furthermore, it is possible to freely regulate reaction and rectification conditions by regulating the pressure in the second reactor (200) and the rectification column (300) with manipulation of outflow of the gas from the upper portion of the rectification column (300), and it is also possible to obtain a high recovery efficiency of the monomer and high purity monomer DMT with introduction of the crystallization tank (400) and the solid-liquid separator (500).

## Brief Description of Drawings

[52] Fig. 1 is a schematic diagram showing an apparatus for recycling feedstock from polyester wastes.

[53]

[54] [Detailed Description of Main Elements]

[55] 10: feedstock feeder 30: methanol reservoir

[56] 31: back-pressure regulator 61: circulating compressor

[57] 90: bubble sparger 100: first reactor

[58] 200: second reactor 300: rectification column

[59] 400: crystallization tank 500: solid-liquid separator

[60] 600: distillation column

[61] 70a, 70b, 110a, 120a, 120b, 130: transfer pipe

[62] 80a, 80b: transfer valve

## Best Mode for Carrying out the Invention

[63] Herein after, apparatus and process for recycling feedstock from polyester wastes will be described.

[64] First, feedstock of polyester wastes is put in a feedstock feeder (10) and then fed to a first reactor (100). The feedstock feeder (10), a feedstock supplier, can employ an extruder, a screw feeder or an air lock valve, but not limited thereto. The polyester wastes can be fed in a solid form from a power with a suitable size to a flake or a melt form. The first reactor (100) can be operated in continuous, semi-continuous or batch type. A predetermined amount of EG and methanol are injected to the first reactor (100) to generate depolymerization reaction, and if necessary, oligomer separated in a distillation column (600) can be put in the first reactor (100) to participate in the reaction. After increasing a temperature in the reactor to a desired temperature, the feedstock methanol is fed from the methanol reservoir (30) to the first reactor (100) through a transfer pipe (120b) so that a pressure in the reactor is reached to a desired reaction pressure. At this time, the methanol is continuously fed so that the pressure in the reactor is maintained at a constant pressure. As glycolysis reaction and methanolysis reaction are generated simultaneously in this first reactor (100) at a high

pressure, a molecular weight of the polyester wastes is considerably reduced and thus the polyester wastes becomes to a state of a solution of DMT and oligomer.

[65] In a continuous second reactor (200) at a low pressure, reactant transferred from the first reactor (100) by the transfer pipe (70a) and the transfer valve (80a) reacts with methanol fed through a bubble sparger (90) from the rectification column (300) in a lower portion of the second reactor (200) to generate methanolysis reaction. Reaction product is discharged in a gaseous phase by a transfer pipe (70b) and a transfer valve (80b) connected to the top of the reactor. Therefore, a predetermined amount of the reactant is transferred from the first reactor (100) when a liquid level in the second reactor (200) is lowered. At this time, the transfer is made automatically by a pressure difference. It is preferred that a valve is provided in a lower portion of the transfer pipe (70a) to control the level of the reaction liquid in an inside of the second reactor (200) and the controller can be connected to the valve (80a) of the transfer pipe so that the valve is automatically operated by the controller.

[66] Into the first reactor (100) which transfers the reactant to the second reactor (200), when the first reactor (100) is operated in a batch type, the solid polyester wastes are put and oligomer and EG solution and methanol are fed, and thus new batch type reaction is started by repeating the process as described above.

[67] That is, the polyester wastes is converted to DMT and oligomer in the first reactor (100) through a high pressure reaction, and gaseous DMT, EG, oligomer and methanol are continuously discharged from the second reactor (200) by the methanolysis. At this time, when the liquid level in the second reactor (200) is lower, the reactant in the first reactor (100) is transferred again to the second reactor (200) to meet the desired level and new batch process is started in the first reactor (100).

[68] Meanwhile, the gaseous reaction product discharged from the second reactor (200) is transferred to a rectification column (300) by the transfer pipe (70b) and comes in direct contact with liquid pool in a lower portion of the rectification column (300) to liquefy DMT and EG and vaporize methanol. As the methanol and other products are separated by such direct contact, it is possible to save a large energy.

[69] Also, feedstock methanol is injected to the upper portion of the rectification column (300) to reflux it and this liquid flow and gaseous flow of the reaction product come in contact with each other in a tray or a packing section to separate the methanol alone through the top of the column. The separated methanol vapor is injected to the second reactor (200) using a circulating compressor (61) or a blower connected to the rectification column (300). After that, the methanol comes in contact with polyester solution in the inside of the second reactor (200) through the bubble sparger (90) to generate the methanolysis reaction.

[70] Also, when the amount of the methanol discharged through the top of the recti-

fication column (300) is excessive, some of the methanol gas is flowed out using a back-pressure regulator (31) or other pressure regulating valve to regulate the pressure of the second reactor (200) and the rectification column (300) and stored in the methanol reservoir(30)after condensation.

[71] On the contrary, when the amount of the methanol fed to the second reactor (200) should be increased, the methanol in the methanol reservoir (30) is fed more to the upper portion of the rectification column (300) and vaporized to be fed to the second reactor (200). A concentration of DMT at the bottom of the rectification column (300) is regulated by adjusting a temperature in the lower portion of the rectification column. The temperature can be controlled by regulating an injection amount (refluxing amount) of the feedstock methanol fed from the methanol reservoir (30) to the upper portion of the rectification column (300) through the transfer pipe (120a), and heating amount in the lower portion of the rectification column (300) is determined in consideration of a recycling or circulating flow rate of the methanol gas.

[72] The DMT and EG rich methanol solution liquefied in the lower portion of the rectification column (300) is transferred to the crystallization tank (400). DMT is crystallized by evaporative cooling and DMT and EG-rich liquid methanol are separated in the solid-liquid separator (500) placed next to the crystallization tank (400). Successively, EG-rich liquid methanol is transferred to the distillation column (600) and is separated into EG, methanol and some oligomer. The separated methanol is stored in the methanol reservoir (30) and EG is collected. Furthermore, the oligomer which is obtained at the bottom of the rectification column (600) is fed to the first reactor (100) and depolymerized again.

[73] While the present invention has been described with respect to the specific embodiments, it will be apparent to those skilled in the art that various changes and modifications may be made without departing from the spirit and scope of the invention as defined in the following claims.

# Claims

[Claim 1] A feedstock recycling apparatus from polyester wastes, which produces dimethyl terephthalate and ethylene glycol, feedstock for polyester from polyester wastes, comprising:
a feedstock feeder (10);
a first reactor (100) for conducting glycolysis reaction and methanolysis reaction in a single reactor using the feedstock fed from the feedstock feeder (10);
a second reactor (200) for subjecting product of the first reactor (100) to methanolysis reaction;
a rectification column (300) for separating methanol from gas discharged through a top of the second reactor (200) and recycling the separated gaseous methanol to the second reactor (200); and
a methanol reservoir (30) for supplying the methanol to the rectification column (300).

[Claim 2] The apparatus of claim 1, wherein the rectification column (300) contacts product of the second reactor (200) directly to liquid pool at bottom of the rectification column (300) to vaporize and separate the methanol alone.

[Claim 3] The apparatus of claim 2, wherein the rectification column (300) recycles a mixture of the methanol separated from the product in the second reactor (200) and methanol fed from the methanol reservoir (30) to an upper portion of the rectification column (300) to be refluxed and vaporized, to the second reactor (200).

[Claim 4] The apparatus of claim 2, further comprising a back-pressure regulator (31) to control the pressure of the methanolysis reactor (200) and the rectification column (300).

[Claim 5] The apparatus of one of claims 1 to 4, further comprising a crystallization tank (400) for crystallizing dimethyl terephthalate from the liquid in the rectification column (300).

[Claim 6] The apparatus of claim 5, further comprising a solid-liquid separator (500) for separating the crystallized dimethyl terephthalate.

[Claim 7] The apparatus of claim 6, further comprising a distillation column (600) connected to the solid-liquid separator (500) and separating the methanol from the ethylene glycol-rich liquid methanol in an upper portion thereof to store the separated methanol in the methanol reservoir (30) and separating oligomer from the ethylene glycol-rich

liquid methanol in a lower portion thereof to recycle the separated oligomer to the first reactor (100).

[Claim 8] The apparatus of claim 1, further comprising, between the feedstock feeder (10) and the first reactor (100), a scrap middle reservoir for weighing the feedstock supplied from the feedstock feeder and supplying the weighed feedstock to the reactor.

[Claim 9] The apparatus of claim 1, further comprising a sensor for sensing a liquid level in an inside of the second reactor (200) in a transfer pipe (70a) for transfer to the second reactor (200).

[Claim 10] The apparatus of claim 9, further comprising a valve (80a) connected to the sensor in the inside of the second reactor (200) and automatically opened when the liquid level sensed by the sensor is lowered.

[Claim 11] The apparatus of claim 1, wherein the second reactor (200) includes a bubble sparger (90) for feeding vaporized methanol from the rectification column (300).

[Claim 12] A feedstock recycling apparatus from polyester wastes, which produces dimethyl terephthalate and ethylene glycol, feedstock for polyester from polyester wastes, comprising:

putting feedstock and a mixture of predetermined amount of ethylene glycol and depolymerizing catalyst in a first reactor (100) and depolymerizing the feedstock by increasing a pressure in the reactor with continuous supply of methanol;

transferring reactant from the first reactor (100) by opening a transfer valve (80a) of a transfer pipe (70a) connected to a second reactor when the depolymerization is completed;

transferring gaseous reaction product produced in the second reactor (200) to a rectification column (300) and separating pure methanol in an upper portion of the column and liquefying the rest methanol, dimethyl terephthalate and ethylene glycol in a lower portion of the column;

recycling the methanol separated in the upper portion of the rectification column (300) to the second reactor (200) and storing some of the methanol in a methanol reservoir (30);

transferring the liquid flowed out from the bottom of the rectification column (300) to a crystallization tank (400) to crystallize dimethyl terephthalate;

transferring product from the crystallization tank (400) to a solid-liquid separator (500) to separate dimethyl terephthalate crystals and ethylene

glycol-rich liquid methanol; and

transferring the ethylene glycol-rich liquid methanol to a distillation column (600) to separate ethylene glycol, oligomer and methanol and storing the methanol in the methanol reservoir (30) and recycling the oligomer to the first reactor (100).

[Claim 13] The process of claim 12, wherein the first reactor (100) and the second reactor (200) are connected by a transfer pipe (70a) so that the reactant is transferred by a pressure difference.

[Claim 14] The process of claim 12, wherein the first reactor (100) is operated at a temperature of 200 to 300 ℃ and a reaction pressure of an atmospheric pressure to 50 bar and the second reactor (200) is operated at a temperature of 200 to 300 ℃ and a reaction pressure of an atmospheric pressure to 10 bar.

[Claim 15] The process of claim 12, wherein when a liquid level in the second reactor (200) is lowered, a transfer valve (80a) of the transfer pipe (70a) is automatically opened and the reactant is transferred from the first reactor (100) to the second reactor (200) by a pressure difference.

[Claim 16] The process of claim 12, wherein the ethylene glycol separated in the distillation column (600) can be used as the feedstock which can be put in the first reactor (100).

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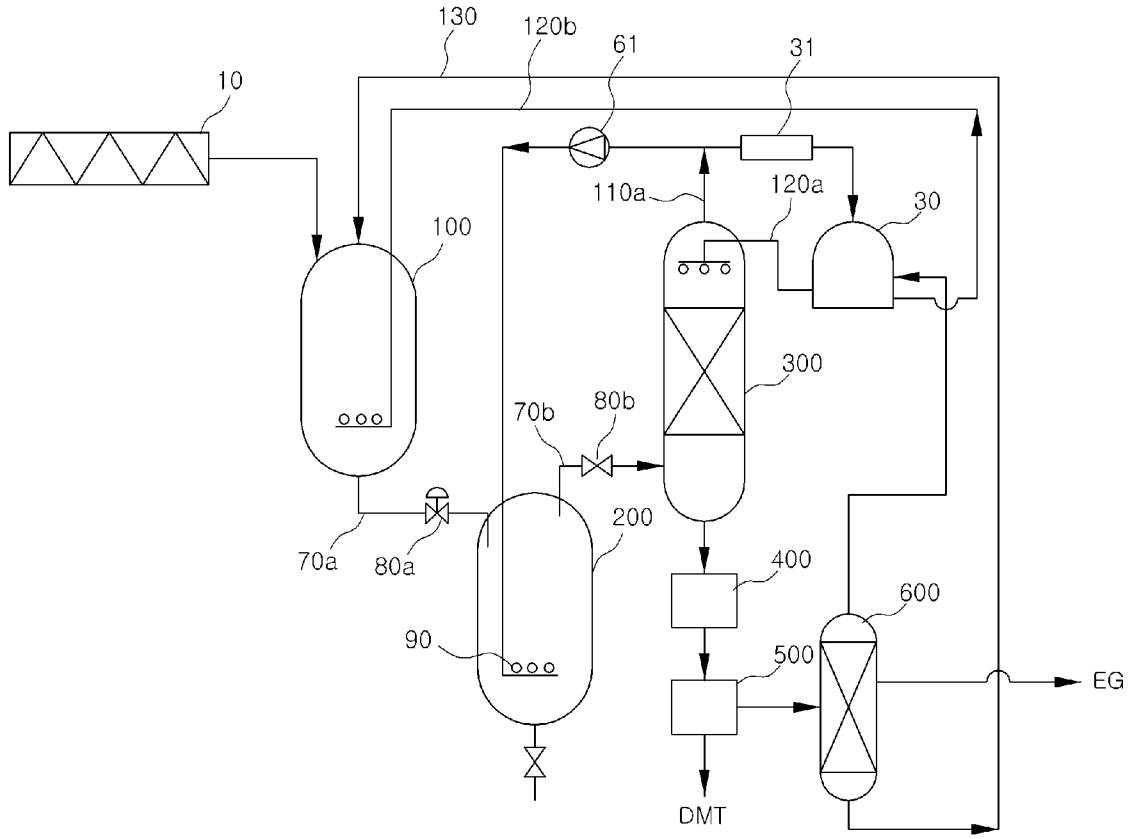
[Fig. 1]



**INTERNATIONAL SEARCH REPORT**

International application No.
**PCT/KR2009/007667**

**A. CLASSIFICATION OF SUBJECT MATTER**

***B01J 19/00(2006.01)i, C08J 11/04(2006.01)i, B01J 19/24(2006.01)i***

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)

B01J 19/00; C08F 6/00; C07C 27/02; C07C 67/60; C08J 11/24; B01D 9/02; B01J 3/00; C08C 67/48

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

Korean utility models and applications for utility models
Japanese utility models and applications for utility models

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

eKOMPASS(KIPO internal) & Keywords: polyester, PET, terephthalate, DMT, waste, recycle

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
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☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
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"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents,such combination being obvious to a person skilled in the art
"&" document member of the same patent family

| Date of the actual completion of the international search | Date of mailing of the international search report |
|---|---|
| 14 OCTOBER 2010 (14.10.2010) | **19 OCTOBER 2010 (19.10.2010)** |


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**INTERNATIONAL SEARCH REPORT**

Information on patent family members

International application No.

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