The opinion in support of the decision being
entered today is not binding precedent of the Board.

Paper 22

UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE BOARD OF PATENT APPEALS
AND INTERFERENCES

Ex parte CARL J. GIARDINO,
DAVID B. GRIFFITH, CHUNGFAH HOWARD HO,
JAMES M. HOWELL and MICHELLE HOYT WATKINS

Appeal No. 2004-0495[1]
Application No. 09/503,599[2]

ON BRIEF

Before: SPIEGEL, TIERNEY and POTEATE, Administrative Patent Judges.

SPIEGEL, Administrative Patent Judge.

DECISION ON APPEAL

This is a decision on appeal under 35 U.S.C. § 134 from the examiner's final rejection of claims 1-33, which are all of the claims pending in this application.

I. Introduction

Claims 1 and 16 are illustrative of the subject matter on appeal and read as follows (emphasis added).

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[1] The application on appeal was received at the Board on 10 December 2003.

[2] Application for patent filed 11 February 2000.

1. A process for the continuous production of at least one of bis(3-hydroxypropyl) terephthalate and low molecular weight oligomers of 1,3-propanediol and terephthalic acid, comprising:

(a) **preheating 1,3-propanediol** to a temperature of about 150°C to about 200°C;

(b) **preheating one or more lower dialkyl esters of terephthalic acid** to a temperature of about 150°C to about 220°C;

(c) combining the preheated 1,3-propanediol and catalyst with the preheated terephthalic acid ester to form a mixture having a mole ratio of 1,3-propanediol to terephthalic acid ester of about 1.2:1 to about 2.3:1 and maintaining the temperature of the mixture at about 150°C to about 220°C;

(d) continuously feeding the preheated 1,3-propanediol/terephthalic acid ester mixture through at least one inlet to an ester exchange vessel where the mixture undergoes a continuous transesterification reaction to form a liquid reaction mixture and gaseous reaction products;

(e) continuously heating and mixing the liquid reaction mixture, whereby the temperature of the mixture in a base portion of the ester exchange vessel is maintained at about 215°C to about 250°C and the pressure in a base portion of the ester exchange vessel is maintained at about 800 mm Hg to about 1,000 mm Hg, thereby continuing the transesterification reaction to form liquid reaction products and gaseous reaction products;

(f) continuously separating the gaseous reaction products from the liquid reaction mixture in a separating portion of the ester exchange vessel which is above the base portion; and

(g) continuously removing a stream of liquid reaction products from the base portion of the ester exchange vessel, the stream comprising at least one of bis(3-hydroxypropyl) terephthalate and low molecular weight oligomers of 1,3 propanediol and terephthalic acid.

16. The process according to claim 1, **wherein said at least one inlet is located below the midpoint of the ester exchange vessel**.

According to the specification, the ester exchange vessel is preferably a bubble cap column wherein the liquid reactants are heated to about 215°C to about 250°C (3:32 - 4:2) and the base of the column is maintained at a pressure of about 800 mm Hg to about 1,000 mm Hg, while the pressure at the top of the column is at or near atmospheric pressure (4:26-33). Further according to the specification, a

transesterification catalyst, including organic and inorganic compounds of titanium, lanthanum, zinc and cobalt, is mixed with 1,3-propanediol prior to mixing with the terephthalic acid ester (3:9-28). Still further according to the specification, the preheated 1,3-propanediol/terephthalic acid ester mixture may be injected into the ester exchange vessel at or below about the midpoint of the vessel (4:34 - 5:1). For example,

> a mixture of preheated, catalyzed 1,3-propanediol and preheated dimethylterephthalate is injected into the base of the exchanger, and additional virgin 1,3-propanediol, typically at ambient temperature, is injected at the quarterpoint of the exchange column. In this embodiment, the mixture that is injected into the base of the exchanger has a lower mole ratio of 1,3-propanediol to dimethylterephthalate and a higher catalyst concentration ..., such that the overall mole ratio and catalyst concentration remain within the above-described ranges, after taking into account the additional 1,3-propanediol that is injected at the quarter point. In still another alternative, preheated dimethylterephthalate and preheated catalyzed 1,3-propanediol are injected into the exchanger separately without premixing. For example, the dimethylterephthalate is injected into the base of the exchanger, and the catalyzed 1,3-propanediol is injected at the quarterpoint. [Specification, 5:18-31.]

Gaseous reaction products typically include methanol vapor, acrolein and allyl alcohol, while liquid reaction products include bis(3-hydroxypropyl) terephthalate monomer, low molecular weight oligomers of 1,3-propanediol and terephthalic acid, and less than 9 wt.% unreacted terephthalic acid ester (id., 6:4-23).

We make reference to appellants' Appeal Brief ("Brief," Paper 19, filed 11 March 2003) and the Examiner's Answer ("Answer," Paper 20, mailed 3 April 2003).

The examiner relies on the following references in his rejection.

Armstrong et al. (Armstrong)	3,534,082	13 October 1970
Kurian et al. (Kurian)	5,840,957	24 November 1998

II. Issues

Claims 1-33 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Kurian in view of Armstrong.

III. Grouping of claims

Appellants' Brief fails to include a "Grouping of Claims" statement as required by 37 CFR § 1.192(c)(7). However, appellants argue that claims 16, 17, 20-26 and 28-33 as separately patentable because the preheated 1,3-propanediol and preheated terephthalic acid ester, either as a mixture or separately, are fed to the ester exchange vessel at or below the midpoint of the vessel (Brief, pp. 3 and 9-12) and the examiner has separately addressed claims 16, 17 and 21 (Answer, p. 5). Therefore, since both appellants and the examiner have considered claims 16, 17, 20-26 and 28-33 as a separate group of claims, this decision focuses on claims 1 and 16.

IV. Discussion

A. The prior art

Kurian discloses a process for making bis(3-hydroxypropyl) terephthalate monomer, comprising contacting a C_1-C_4 dialkyl ester of terephthalic acid with 1,3-propanediol in the presence of a lanthanum compound catalyst at a temperature from about 155°C to about 245°C with a mole ratio of 1,3-propanediol to ester of about 1.1:1 to about 2.2:1 at about atmospheric pressure[3] in a reaction vessel in which the transesterification reaction occurs generally within about 0.5 to about 4 hours (c. 1, ll. 49-58; c. 3, ll. 24-35). Kurian's process may be performed as either a batch or

[3] One atmosphere pressure is about 760 mm Hg.

continuous process (c. 3, ll. 38-39). Kurian Example 1 is illustrative.

> A 250 ml flask equipped with a stirrer and distillation column was charged with 58.5 g of dimethyl terephthalate (DMT) and 41 g of 1,3-propanediol ... for a mole ratio of 1,3-propanediol:DMT of 1.8:1. The flask was then purged with nitrogen and the contents of the flask were heated. When the temperature inside the flask reached about 150°C and all of the DMT had melted, the stirrer was started. When the temperature reached 210°C, 30 mg of lanthanum tris(acetylacetonate) hydrate ... was added.
>
> ... Upon addition of the lanthanum tris(acetylacetonate) hydrate, methanol was evolved [i.e., a gaseous reaction product]. The methanol was removed as a liquid condensate by distillation. The total liquid condensate was assumed to be methanol for purposes of this Example because the amount of methanol is more than 99% of the condensate in such a procedure. The temperature was held at 210° C. and the amount of liquid methanol collected was measured every ten minutes until no more methanol was evolved. ...
>
> After evolution of methanol, the resulting monomer, bis(3-hydroxypropyl) terephthalate, was polymerized in the same reaction vessel at a temperature of 250° C. and a pressure of 0.4 mm Hg, using a titanate compound as the polycondensate. ... [c. 4, l. 55 - c. 5, l. 25.]

As noted by the examiner, Kurian "does not teach the preheating of the starting material" (Answer, p. 4, ¶ 2). As noted by appellants, Kurian "does not provide details regarding the reactors used to make his product" (Brief, p. 10, ¶ 3) or "any suggestion whatsoever to feed reactants at or below the midpoint of an ester exchange vessel" (id., p. 11, ¶ 2).

Armstrong discloses continuous production of bis(2-hydroxylethyl) terephthalate by reacting ethylene glycol and dimethyl terephthalate in a vessel and then completing the reaction in a bubble cap column (c. 1, ll. 15-20). As described in Example 1,

> An apparatus is used of the type illustrated in the drawing. Vessel **3** is a stainless steel pot heated by means of electric heating coils **4**, with dimensions such that the liquid content is approximately 40 liters (1.4 cu. ft.). Vessel **7** is a conventional bubble-cap distillation column approximately 30 cm. (12 inches) in diameter with 20 bubble-cap plates and with the liquid level on each plate being about 4.75 cm. (1.875 inches)

> in depth. The liquid volume in the collecting boiler at the bottom of the column is approximately 22 liters (0.77 cu. ft.). Tube **5** feeds the product from vessel **3** onto plate number **14** of column **7** and tube **6** feeds vapors from vessel **3** into the space above plate **15**.
>
> Molten dimethyl terephthalate is fed continuously to vessel **3** through tube **1** at the rate of approximately 60 kg. per hour. At the same time, ethylene glycol and catalyst are fed through tube **2** at the rate of approximately 33 kg. per hour. ... The mole ratio of glycol to dimethyl terephthalate fed to vessel **3** is approximately 1.7. Heat is supplied to heating coils **4** to maintain a temperature of 185° C. The mixture is stirred by the bubbling of boiling methanol released by the exchange reaction. Methanol vapors along with some glycol vapors are led through tube **6** to column **7**. The liquid mixture in vessel **3** overflows into tube **5** through which it is directed to the reaction portion of column **7**.
>
> * * * * * *
>
> The conditions in the column are regulated to give a pressure drop from the bottom to the top of the column of 60 inches of water (112 mm of mercury) The column is heated by coil **12** to maintain the temperature in the calandria (reboiler **13**) at 230±2° C., and partial condensor 9 is adjusted to give an overhead take-off temperature of 70° C. Fresh glycol is fed to the 17th plate at a rate of 5 kg. per hour. ...
>
> * * * * * *
>
> The product from outlet **11** of the ester interchange column is immediately passed into a continuous polymerization system ... . [c. 4, l. 57 - c. 5, l. 60.]

Fresh glycol was added to maintain the desired ratio of ethylene glycol to dimethyl terephthalate, i.e., "hot liquid glycol is supplied continuously in suitable quantity through tube **15** onto a plate located above tube **6**. Such an arrangement helps wash down entrained dimethyl terephthalate from the upper plates and thereby helps prevent plugging of the partial condenser." [See Figure; and, c. 4, ll. 27-35.]

The examiner does not disagree with appellants that Armstrong does not teach or suggest feeding reactants at or below the midpoint of an ester exchange vessel (Brief, p. 11, ¶ 2).

B. The legal standard

For a prima facie case of obviousness to be established, the teachings of the prior art itself must appear to have suggested the claimed subject matter to one of ordinary skill in the art. See In re Rinehart, 531 F.2d 1048, 1051, 189 USPQ 143, 147 (CCPA 1976). The mere fact that the prior art could be modified as proposed by the examiner is not sufficient to establish a prima facie case of obviousness. See In re Fritch, 972 F.2d 1260, 1266, 23 USPQ2d 1780, 1783 (Fed. Cir. 1992). The examiner must explain why the prior art would have suggested to one of ordinary skill in the art the desirability of the modification. Id., 972 F.2d at 1266, 23 USPQ2d at 1783-84.

C. Analysis

According to the examiner, (i) Kurian suggests that neither starting reactant needs to be preheated, whereas Armstrong suggests that the reactants can be preheated (Answer, p. 5, ¶ 1); (ii) preheating starting materials is known in the art to minimize processing time (id., p. 4, ¶ 4); and, (iii) feeding reactants into different levels of a chemical reactor is a routine variable used to control residence time and generation of volatile byproducts in the reactor (id., p. 5, ¶ 3). Further according to the examiner, appellants have not shown any unexpected results due to preheating the starting materials or feeding reactants into the transesterification vessel at different levels (id., pp. 4-6).

First, as pointed out by appellants (Brief, p. 7, last ¶), Armstrong only preheats one starting material, the dimethyl terephthalate, before mixing. Armstrong discloses adding hot glycol to the bubble cap column, if needed to maintain the

glycol/terephthalate ratio. The mere fact that the prior art could be modified as proposed by the examiner is not sufficient to establish a prima facie case of obviousness.

Second, the conclusions of the examiner that (ii) preheating starting materials is known in the art to minimize processing time and (iii) feeding reactants into different levels of a reactor is a routine variable used to control residence time and generation of volatile byproducts in the reactor are unsupported both generally and specifically in regards to the claimed invention. On the contrary, Armstrong suggests feeding reactants in at plate 14 of a 20-plate bubble cap column (see Figure; c. 4, ll. 66-69), i.e., in above the midpoint of the ester exchange vessel. Again, the mere fact that the prior art could be modified as proposed by the examiner is not sufficient to establish a prima facie case of obviousness.

Based on the foregoing, the decision of the examiner to reject claims 1-33 as obvious over Kurian in view of Armstrong is reversed.

V. Miscellaneous

There is art of record which suggests preheating both glycol and terephthalate components prior to reaction. For example, U.S. Patent 2,829,153 to Vodonik, cited in appellants' specification (1:20-23) as disclosing a continuous process for the production of bis(2-hydroxyethylene) terephthalate and made of record in an Information Disclosure Statement (Paper 4, "IDS" filed 13 June 2000), expressly states that

> ethylene glycol may be fed to the column cold, i.e., room temperature or above, but for purposes of economy and optimum operation of the column, it is usually desirable to have its temperature approximate the temperature of the feed plate. Similarly, the temperature of the dimethyl

> terephthalate feed stream may be any reasonable temperature above its melting point, e.g., 145 to 175° C., but here too for the reasons given above, it is usually desirable to have its temperature approximate the temperature on its feed plate. [c. 3, ll. 66-75.]

U.S. Patent 3,167,531 issued to Parker et al. and of record (IDS, Paper 4), states that

> [f]irst, the reactants are prepared for the continuous operation of the process. Dimethyl terephthalate or other suitable lower dialkyl ester of terephthalic acid is heated to the liquid phase. ... Ethylene glycol is supplied from a source and is fed to the ester-interchange zone separately from the molten dimethyl terephthalate being fed to such zone. On the way to the zone the glycol normally will be preheated, preferably to the temperature of 140 to 180° C. While it is preferred to preheat the glycol so that the ester-interchange reaction proceeds in an advantageous manner, it is not essential to do so. [c. 2, ll. 16-32.]

Therefore, upon return of this application, the examiner should consider whether one or more of claims 1-33 are unpatentable over Kurian (5,840,957) in view of Armstrong (3,534,082) (including the newly pointed out disclosure at c. 4, ll. 66-69) and either Vodonik (2,829,153) or Parker (3,167,531). The examiner is reminded to set forth sufficient reasoning for asserting the obviousness or inherent nature of each claimed limitation, i.e., each claim must be individually examined, in order to shift the burden to appellants to come forward with evidence and/or arguments to rebut the examiner's position.

VI. Conclusion

To summarize, the decision of the examiner to reject claims 1-33 under 35 U.S.C. § 103(a) as unpatentable over Kurian in view of Armstrong is reversed.

REVERSED

_____ CAROL A. SPIEGEL Administrative Patent Judge	)
_____ MICHAEL P. TIERNEY Administrative Patent Judge	) BOARD OF PATENT) APPEALS) AND) INTERFERENCES
_____ LINDA R. POTEATE Administrative Patent Judge	)

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