

The opinion in support of the decision being entered today was not written for publication and is not precedent of the Board.

Paper No. 30

UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE BOARD OF PATENT APPEALS
AND INTERFERENCES

Ex parte EXXON CHEMICAL PATENTS INC.

Appeal Nos. 1999-1140 and 1999-1141
Control Nos. 90/004,574 and 90/004,248¹

HEARD: OCTOBER 10, 2000

Before WARREN, OWENS and JEFFREY T. SMITH, *Administrative Patent Judges*.

OWENS, *Administrative Patent Judge*.

DECISION ON APPEAL

This is an appeal from the examiner's final rejection of claims 1-34, which are all of the claims in this reexamination proceeding.

¹ Merged reexamination proceeding for U.S. Patent No. 5,405,922, issued April 11, 1995, based on Application 08/216,520, filed March 22, 1994, which is a continuation of Application 08/065,250, filed May 20, 1993.

THE INVENTION

Appellant claims a continuous gas phase fluidized bed polymerization process which uses a silica-supported metallocene catalyst and condensed mode operation.² Claim 1 is illustrative:

1. A continuous gas phase polymerization process comprising copolymerizing an alpha-olefin monomer with at least one other alpha-olefin comonomer in the presence of a silica-supported metallocene catalyst in a gas phase fluidized bed reactor operating in a condensed mode, said process further comprises a recycle stream passing through a fluidized bed in said reactor wherein the recycle stream comprises a dew point increasing component in an amount greater than 2.0 mole percent.

THE REFERENCES

References relied upon by the examiner

Jenkins, III et al. (Jenkins) 1986	4,588,790	May 13,
Welborn, Jr. (Welborn) 1989	4,808,561	Feb. 28,
Chang (Chang '301) 1990	4,937,301	Jun. 26,
Burkhardt et al. (Burkhardt) 1993	5,240,894	Aug. 31,

(filed May 18,

² "Condensed mode" operation is defined in the patent under reexamination as "the process of purposefully introducing a recycle stream having a liquid and a gas phase into a reactor such that the weight percent of liquid based on the total weight of the recycle stream is greater than about 2.0 weight percent" (col. 1, lines 50-57).

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OPINION

We affirm the aforementioned rejection.

Appellant states that the claims stand or fall together (brief, page 5). We therefore limit our discussion to one claim, i.e., claim 1. See *In re Ochiai*, 71 F.3d 1565, 1566 n.2, 37 USPQ2d 1127, 1129 n.2 (Fed. Cir. 1995); 37 CFR § 1.192(c)(7)(1997).

Welborn discloses a catalyst which is particularly useful for the polymerization of ethylene and copolymerization of ethylene with 1-olefins having 3 or more carbon atoms (col. 1, lines 6-13; col. 2, lines 32-38). The catalyst is made by reacting an alumoxane and a metallocene in the presence of a solid support material which can be silica (col. 3, lines 34-40 and 46-48). The catalyst can be used in a continuous gas

rejection in the merged reexamination final, and refusing to consider the third Muhle declaration and the Floyd declaration (supplemental appendix to brief, tabs 3 and 4, respectively). Appellant acknowledges that petitions of the examiner's decisions on these issues have been denied (brief, pages 8-9; reply brief, page 2). Appellant does not bring to our attention any authority which confers jurisdiction upon the board to render a decision on the propriety of the decisions on these petitions, and we are aware of none. Consequently, we do not address the merits of these issues and do not consider the third Muhle declaration and the Floyd declaration in reaching our decision.

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phase fluidized bed process which includes a recycle stream (col. 8, line 62 - col. 9, line 6). Welborn discloses, in an example, that "[t]he recovered [catalyst] solid was neither soluble nor extractable in hexane" (col. 11, lines 39-40).⁵ Welborn does not disclose condensed mode operation or adding a dew point increasing component to the recycle stream.

Jenkins discloses a continuous gas phase fluidized bed polymerization process which is operated in the condensed mode, which he defines as using a recycle stream below its dew point, at a recycle stream liquid content which can be "quite high" but generally does not exceed about 20 wt% (col. 3, line 60 - col. 4, line 7; col. 5, lines 61-63). The benefit of the condensed mode operation is that "the cooling capacity of the recycle stream is increased by both the vaporization of the condensed liquids entrained in the recycle stream and as a result of the greater temperature differential between the entering recycle stream and the reactor" (col. 3, line 67 - col. 4, line 5). This benefit is small when the liquid content of the recycle stream is below 2 wt% (col. 6, lines

⁵ It reasonably appears that this extraction was carried out at room temperature.

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51-52). The dew point of the recycle stream may be increased to permit a larger increase in heat removal by adding to it a condensible fluid, the disclosed examples of which are saturated hydrocarbons such as butanes, pentanes or hexanes (col. 5, line 64 - col. 6, line 12). The process is not limited to any specific type of polymerization reaction, but is especially advantageous for polymerizations of olefin-type monomers (col. 4, lines 62-66). The polymers which may be made using the process include "copolymers of a major mole percent of ethylene, propylene or butene and a minor mole percent of one or more C2 to C8 alphaolefins" (col. 16, lines 54-57). The only disclosed catalyst is a complex of tetrahydrofuran, magnesium chloride and titanium chloride reduced with diethyl aluminum chloride and tri-n-hexyl aluminum impregnated on triethyl aluminum treated silicon dioxide (col. 18, lines 20-26), which is not a metallocene catalyst.

Thus, Wellborn and Jenkins do not indicate that a supported metallocene catalyst is suitable for use in a condensed mode polymerization process wherein the catalyst is exposed at polymerization conditions to the solvents used to

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increase the dew point of the recycle stream.

Burkhardt states that he believes that the low catalytic activity and reactor wall fouling in polymerization processes which employ a metallocene catalyst are due to several factors, the following two of which are disclosed (col. 2, line 52 - col. 3, line 2):

First, residual solvent remains in the pores of the support material employed at the stage after placement of the catalyst onto the carrier. The residual solvent prevents the catalyst system from securely anchoring itself onto the carrier or into the pores of the carrier. Thus when the supported catalyst is added to the reaction polymerization vessel, the catalyst disassociates from the support, and migrates to the reactor walls where monomer can polymerize therefrom and cause fouling. Secondly, when methyl alumoxane (MAO) is used as cocatalyst in the polymerization at temperatures about or greater than 40°C., the MAO dissolves and extracts the metallocene catalyst from the support and forms a soluble catalyst in the polymerization medium. This soluble catalyst easily deposits polymer onto the reactor walls and/or generates very small particles of low bulk density which are undesirable in a commercial reactor.

Burkhardt teaches that there are minimal or no signs of reactor fouling observed during polymerization when Burkhardt's process is used to make the supported catalyst (col. 3, lines 33-36). This teaching indicates that the catalyst is free of the problem, discussed in the above

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excerpt from Burkhardt, of commercially unacceptable reactor fouling (col. 3, lines 15-16) resulting from dissociation and extraction of the catalyst from the support.

Burkhardt's process involves the following steps: 1) forming a metallocene/alumoxane reaction solution, 2) adding a dehydrated catalyst support, 3) evaporating or heating the resulting slurry to remove liquid solvent, 4) further heating to desolvate the solid supported catalyst and thus remove any residual solvent trapped in the pores prior to use, and 5) optionally prepolymerizing the catalyst with olefinic monomer (col. 4, lines 33-39). The disclosed solvents for forming the solution in the first step include alkanes, cycloalkanes and aromatics (col. 6, lines 6-15). The support, which typically is silica, is thoroughly dried and is added in the second step to the metallocene/alumoxane reaction solution to form a slurry (col. 5, lines 8-55; col. 6, lines 41-45). The slurry is heated under vacuum in the third step until all of the solvent has been removed, and then the solid is heated in the fourth step until all of the residual solvent trapped in the porous support material has been removed (col. 6, lines 55-65). The catalyst so produced can be used for polymerization

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of one or more olefins or alpha olefins containing from about 2 to about 20 carbon atoms to form homopolymers or copolymers (col. 3, lines 57-60).

Burkhardt teaches that the metallocene and alumoxane initially are soluble in alkanes (col. 6, lines 6-15), which are the exemplified condensible fluids which Jenkins adds to increase the dew point of the recycle stream (col. 6, lines 3-12). These teachings, taken alone, indicate that if Burkhardt's catalyst were used in a condensed mode process, the condensible fluids used to increase the dew point would dissolve the metallocene and alumoxane catalyst components. Burkhardt teaches that these dissolved catalyst components would cause reactor fouling (col. 2, line 49 - col. 3, line 2). Burkhardt, however, teaches that "[t]he normally hydrocarbon soluble metallocenes and linear or cyclic alumoxanes are converted to a heterogeneous supported catalyst by depositing the reaction product of metallocene and alumoxane on the thermally or chemically dehydrated supported material" (col. 5, line 65 - col. 6, line 2). This teaching indicates that the metallocenes and alumoxanes, which are normally hydrocarbon soluble in free form, are in a different

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form, i.e., a heterogeneous form in which they are secured in the pores of the catalyst after the solvent has been removed therefrom, after his catalyst preparation process. The disclosures by Burkhardt that 1) dissociation or extraction of metallocene from a catalyst support causes reactor fouling (col. 2, line 52 - col. 3, line 2), 2) metallocenes and alumoxanes are "normally hydrocarbon soluble" (col. 5, line 66), 3) after metallocenes and alumoxanes are converted to a heterogeneous supported catalyst according to Burkhardt's process there are minimal to no signs of reactor fouling when the catalyst is used for polymerization (col. 3, lines 33-36), and 4) the polymerization using Burkhardt's catalyst can be carried out in the presence of hexane in an autoclave (col. 8, lines 15-16; col. 11, lines 25-42), indicate that the "normally hydrocarbon soluble" metallocenes and alumoxanes are rendered sufficiently hydrocarbon insoluble by Burkhardt's catalyst preparation process that Burkhardt's catalyst is suitable for use in the presence of the hydrocarbons which Jenkins uses to increase the dew point of the recycle stream (col. 6, lines 3-12). Thus, the teaching by Burkhardt that the catalyst can be used "in any polyolefin polymerization

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reactor with little modification and without operational process related problems" (col. 7, lines 59-61) would have indicated, to one of ordinary skill in the art, that the catalyst would be suitable for use in Welborn's fluidized bed reactor operated in the condensed mode as suggested by Jenkins.⁶

For a *prima facie* case of obviousness of appellant's claimed process to be established, the applied prior art must have provided one of ordinary skill in the art with both a motivation to carry out appellant's claimed process and a reasonable expectation of success in doing so. See *In re Vaeck*, 947 F.2d 488, 493, 20 USPQ2d 1438, 1442 (Fed. Cir. 1991); *In re O'Farrell*, 853 F.2d 894, 902, 7 USPQ2d 1673, 1680

⁶ Appellant argues (brief, pages 13-14 and 41) that Bailly states (col. 7, lines 31-38) that a catalyst prepolymerization step is preferred because it renders the metallocene less capable of being extracted from the catalyst and producing fine particles of polymer during the polymerization, and that Muñoz-Escalona discloses that as late as the spring of 1998, leaching of metallocene catalysts from their supports during polymerization caused a reactor fouling problem. The catalysts used by Bailly and Muñoz-Escalona, however, were not prepared by Burkhardt's process. Consequently, these references provide no indication as to whether Burkhardt's metallocene is extractable from the catalyst support.

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(Fed. Cir. 1988). One of ordinary skill in the art would have been motivated by the references to use Burkhardt's catalyst in Welborn's fluidized bed reactor operated in the condensed mode in order to obtain the benefits of the enhanced heat transfer resulting from the condensed mode operation disclosed by Jenkins (col. 3, line 67 - col. 4, line 5) and the minimal to no reactor fouling resulting from the particular catalyst preparation method disclosed by Burkhardt (col. 3, lines 33-36). Such a person would have had a reasonable expectation of success in doing so because of Burkhardt's above-discussed teaching that 1) a metallocene catalyst made by the particular technique disclosed therein is capable of being used in any polyolefin polymerization reactor (col. 7, lines 57-61), 2) his catalyst contains "normally hydrocarbon soluble metallocenes and linear or cyclic alumoxanes" which "are converted to a heterogeneous supported catalyst" (col. 5, lines 66-68), 3) his catalyst can be used at polymerization conditions in the presence of hexane (col. 11, lines 25-47), which is among the dew point increasing condensible fluids used by Jenkins (col. 6, lines 3-12), and 4) use of his catalyst results in minimal or no signs of reactor fouling

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(col. 3, lines 33-36). Accordingly, we hold that appellant's claimed invention would have been *prima facie* obvious to one of ordinary skill in the art over the applied prior art.

Appellant argues that Burkhardt "teaches that with a silica-supported metallocene catalyst, the metallocene catalyst can be extracted or solubilized in a suitable hydrocarbon solvent such as isopentane or hexane, particularly when such hydrocarbon solvent is at an elevated temperature (i.e., about 40°C or higher)" (brief, page 29). The portion of Burkhardt (col. 2, line 49 - col. 3, line 2) relied upon by appellant in support of this argument, however, pertains to Burkhardt's discussion of reactor fouling problems which were encountered when using prior catalysts. Burkhardt teaches that his particular catalyst preparation technique results in minimal or no signs of reactor fouling (col. 3, lines 33-36).⁷

Regarding Burkhardt's example in which his catalyst is used in a polymerization process in the presence of hexane (col. 11, lines 25-47), appellant argues that "Burkhardt

⁷ The patent under reexamination states (col. 5, lines 25-37) that Burkhardt's catalyst is among the catalysts which can be used in appellant's process.

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washed and/or prepolymerized the catalyst, both of which are said to reduce the problem in a slurry reactor of unwanted polymerization caused by catalyst that is not fully adsorbed into the pores of the silica" (brief, page 31). Support for this argument is not found in the reference. Regarding prepolymerizing the catalyst, Burkhardt teaches that such prepolymerization "can be employed to strengthen catalyst particles and enhance particle size control of the final polymer formed (col. 7, lines 31-33). Regardless, appellant's claim 1 does not exclude the use of a washed and/or prepolymerized catalyst.

Appellant argues that in Burkhardt's polymerization example (col. 11, lines 25-47), wherein the polymerization was conducted in an autoclave for one hour, there was no opportunity for plugging problems which could arise during many hours of continuous condensed mode operation of a gas phase fluidized bed reactor (brief, page 31). Burkhardt's polymerization example does not provide absolute certainty that his catalyst will perform effectively in a continuous condensed mode gas phase fluidized bed polymerization process. Such absolute certainty, however, is not needed for a *prima*

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facie case of obviousness to be established. Instead, all that is needed is a reasonable expectation of success. See *O'Farrell*, 853 F.2d at 903-04, 7 USPQ2d at 1681. For the reasons given above, the applied references would have provided one of ordinary skill in the art with such a reasonable expectation of success. Furthermore, Jenkins teaches that it is important to minimize the carryover of particles in the recycle stream (col. 5, lines 24-25). Jenkins teaches (col. 7, lines 12-16) that "[t]he amount of solids in the gaseous stream exiting the reactor is typically small, e.g., from about 0.1 to about 0.5 weight percent (based on the total weight of the stream). However, larger amounts on the order of one weight percent or higher, may occur." The reason for minimizing the carryover of solids is: "Since these particles are hot and contain catalyst, they will continue to grow by further reaction with monomer gas as they are carried through the recycle system, potentially causing problems by settling out and agglomerating into a solid mass or sticking to the walls of the recycle line and heat exchanger. This may ultimately lead to plugging of the gas distributor plate,

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recycle lines or heat exchanger, necessitating shutdown" (col. 5, lines 16-23). This teaching, together with Burkhardt's teaching that his catalyst 1) is "capable of operating in any polyolefin polymerization reactor with little modification and without operational process related problems" (col. 7, lines 59-61), 2) contains metallocenes and alumoxanes which are in the form of a heterogeneous supported catalyst rather than the normally hydrocarbon soluble form (col. 5, lines 66-68), and 3) has been observed to produce little or no signs of reactor fouling (col. 3, lines 33-36), would have provided one of ordinary skill in the art with a reasonable expectation that Burkhardt's polymerization in the presence of hexane, which was exemplified in an autoclave, would be effective without an unacceptable degree of fouling in a continuous gas phase fluidized bed system wherein the carryover of solids in the recycle stream is minimized.

Appellant argues that the Speca declaration (supplemental appendix to appeal brief, tab 1) demonstrates that one of ordinary skill in the art would have knowledge of the potential for catalytically active metallocene species to be solubilized off silica supports (brief, page 48). Appellant,

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however, has not established that the evidence in this declaration was known to those of ordinary skill in the art. Thus, this evidence is not useful for supporting appellant's argument (brief, page 48) that one of ordinary skill in the art would have been led away from using, as the catalyst in a condensed mode gas phase fluidized bed olefin polymerization process, a metallocene supported on silica.

Appellant argues that the Speca declaration provides evidence that metallocene catalysts are soluble in hydrocarbons used as dew point increasing components in condensed mode operation, even after being placed on a silica support (brief, pages 25 and 39). Evidence such as this, although not known to those of ordinary skill in the art at the time of appellant's invention, potentially could be used to show that Burkhardt's catalyst cannot function effectively in a continuous condensed mode gas phase fluidized bed olefin polymerization process. The Speca declaration, however, does not establish that such evidence is provided therein. The reasons are, first, it does not established that Burkhardt's catalyst is used in the tests. Burkhardt's catalyst is prepared by a specific procedure which involves forming a

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metallocene/alumoxane reaction product, adding this reaction product to a dehydrated carrier, which typically is silica, and heating the slurry until all of the solvent trapped in the porous support material has been removed (col. 6, lines 24-65). The Speca declaration does not indicate that the silica was dehydrated or that the slurry was heated until all of the solvent trapped in the porous support material had been removed. Second, the declaration does not establish that the tests of the catalyst, wherein either 2.19 grams or 2.0831 grams of the catalyst are stirred in a beaker with 40 cc of hexane at 55°C for 30 minutes, five times, is representative of the conditions to which the catalyst would be exposed in a continuous condensed mode gas phase fluidized bed polymerization process, particularly one in which the recycle stream contains only slightly more than 2 mole% of a dew point increasing component, which is the minimum amount required by appellant's claim 1.

Appellant argues, in reliance upon the second Muhle declaration (supplemental appendix to appeal brief, tab 2), that one of ordinary skill in the art would have expected that use of a silica-supported metallocene catalyst would result in

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reactor system fouling (brief, page 39), and that Muhle was surprised that a silica-supported metallocene catalyst could be used successfully in a condensed mode gas phase fluidized bed olefin polymerization process without significant operability problems (brief, page 47). What Muhle states in his second declaration is that he would have expected that a metallocene catalyst that has been removed from its support and solubilized in the liquid phase of the fluidizing medium will travel throughout the reactor system and cause polymerization on reactor system surfaces, thereby fouling the surfaces. He does not state that one of ordinary skill in the art would have expected that use of a silica-supported metallocene catalyst would result in reactor system fouling or that he was surprised that a silica-supported metallocene catalyst could be used successfully in a condensed mode gas phase fluidized bed olefin polymerization process without significant operability problems.

Appellant argues that condensed mode operations can be carried out only in large, commercial scale gas phase reactors and argue, in reliance upon the second Muhle declaration, that an operator of a commercial unit would have been very

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reluctant to even attempt to practice appellant's claimed process (brief, pages 12-13). Jenkins, however, teaches that it would be very difficult to operate a pilot plant reactor in the condensed mode if the reactor has a fluid bed depth of no more than about five feet (col. 9, lines 52-62). This disclosure indicates that a commercial reactor is not needed to carry out condensed mode operation but, rather, that a pilot plant reactor having a fluid bed depth of more than five feet would be sufficient. Consequently, the applied references at least would have fairly suggested, to one of ordinary skill in the art, carrying out appellant's claimed polymerization process in a pilot scale system having a fluid bed depth greater than five feet.

For the above reasons we conclude, based upon the preponderance of the evidence, that the claimed invention would have been obvious to one of ordinary skill in the art within the meaning of 35 U.S.C. § 103.

DECISION

The rejection of claims 1-34 under 35 U.S.C. § 103 over

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Welborn in view of Jenkins, Burkhardt, Chang '301, Chang '593
and ECN, is affirmed.

No time period for taking any subsequent action in
connection with this appeal may be extended under 37 CFR
§ 1.136(a).

AFFIRMED

	CHARLES F. WARREN)	
	Administrative Patent Judge)	
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	TERRY J. OWENS)	BOARD OF
PATENT	Administrative Patent Judge)	APPEALS AND
)	INTERFERENCES
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