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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.
11/241,321	09/30/2005	Robert A. Dagle	14756-e

EXAMINER
KEYS, ROSALYND ANN

ART UNIT	PAPER NUMBER
1621	

MAIL DATE	DELIVERY MODE
01-30-2013	PAPER

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UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

Ex parte ROBERT A. DAGLE, YONG WANG,
EDDIE G. BAKER, and JIANLI HU

Appeal 2011-011644
Application 11/241,321
Technology Center 1600

Before TONI R. SCHEINER, DEMETRA J. MILLS, and LORA M. GREEN, *Administrative Patent Judges*.

SCHEINER, *Administrative Patent Judge*.

DECISION ON APPEAL

This is an appeal under 35 U.S.C. § 134 from the final rejection of claims 28-43, 46-49, 51, and 52 directed to a process for producing dimethyl ether. The claims have been rejected as obvious. We have jurisdiction under 35 U.S.C. § 6(b).

We affirm.

STATEMENT OF THE CASE

Claims 28-43, 46-49, 51, and 52 are pending and on appeal. Claims 1-27, 44, 45, and 50 have been canceled (App. Br. 2).

Appellants do not provide separate arguments for the claims with the exception of 30. Accordingly, we select independent claim 28 and claim 30 as representative for purposes of deciding this appeal, and the remaining claims will stand or fall accordingly. 37 C.F.R. § 41.37(c)(1)(vii). Claims 28 and 30 are as follows:

> 28. A process for producing dimethyl ether comprising:
> reacting syngas in contact with a hybrid catalyst in a microchannel reactor at a temperature from about 200º C to about 400º C with a contact time of from about 25 milliseconds to less than about 1 second; and converting more than about 60% of CO in the syngas to dimethyl ether and methanol.
>
> 30. The process of claim 28, wherein there is no liquid holdup inside the microchannel reactor.

The Examiner relies on the following evidence:

Grady et al.	US 5,821,111	Oct. 13, 1998
Wegeng et al.	US 5,811,062	Sep. 22, 1998
Shikada et al.	US 6,562,306 B1	May 13, 2003
Tonkovich et al.	US 6,616,909 B1	Sep. 9, 2003

Appellants rely on the following additional evidence:

Declaration of Dr. Anna Lee Tonkovich, submitted under the provisions of 37 C.F.R. § 1.132, dated December 27, 2010 ("Decl.").

The claims stand rejected under 35 U.S.C. § 103(a) as unpatentable over Shikada, Tonkovich, Grady, and Wegeng.

ISSUES

The Examiner finds that the Shikada patent discloses a method of manufacturing dimethyl ether (DME) from syngas (a mixture of carbon

monoxide and hydrogen, or carbon monoxide, hydrogen, and carbon dioxide) in a thermal reaction using a hybrid methanol-synthesis, methanol-dehydration catalyst, wherein the carbon monoxide conversion rate is 35% or more. The Examiner concedes that the Shikada patent's reaction proceeds in a slurry-bed reactor, rather than a microchannel reactor. However, the Examiner finds that the Tonkovich patent teaches that performing the reaction in a microchannel reactor provides an "enhanced production rate of thermal chemical reactions . . . as well as suppress[es] the formation of undesirable byproducts" (*id.* at 5-6) in "reactions includ[ing] dehydration, methanol synthesis and water gas shift" (*id.* at 6), "which are the types of reactions that take place in the process taught by Shikada" (*id.* at 9).

The Examiner concludes that it would have been obvious for one of ordinary skill in the art to use the Tonkovich patent's microchannel reactor to carry out "the thermal chemical reaction of Shikada . . . as this would allow one to produce the dimethyl ether . . . at an enhanced production rate, while suppressing undesirable byproducts by using short contact times" (Ans. 6) of "about 25 milliseconds to less than about 1 second" (*id.* at 5).

Appellants contend that the Shikada patent teaches that "the reaction must occur in the slurry phase" (App. Br. 4), and "at the time of the invention . . . it would not have been known if the slurry phase process of Shikada could be conducted in a microchannel" (*id.* at 4-5). Appellants contend that "there is no reason to have believed that, even if conducted in a microchannel at the claimed contact times, Shikada's slurry-phase process would have succeeded to obtain the claimed levels of conversion" (*id.* at 5).

In addition, Appellants contend that "the reaction of syngas to DME in a microchannel [unexpectedly] results in superior selectivity" (App. Br. 5), "as compared with a conventional slurry bed process" (*id.*).

The issues raised by this appeal are as follows:

Does the preponderance of the evidence support the Examiner's conclusion that it would have been obvious to produce dimethyl ether in a microchannel reactor, given the teachings of Shikada and Tonkovich?

If so, have Appellants provided evidence of unexpected results that outweighs the evidence supporting the prima facie case of obviousness?

FINDINGS OF FACT

1. The Shikada patent discloses a method of manufacturing dimethyl ether from syngas (carbon monoxide and hydrogen) in a slurry bed reactor, using a variety of hybrid catalysts, including a methanol-synthesis, methanol-dehydration catalyst (Shikada, col. 7, ll. 61-62; col. 9, ll. 6-35). The catalysts are in particulate form, and suspended in a solvent for reaction with the syngas (*see e.g.*, col. 2, l. 34 - col. 3, l. 48).

2. The Shikada patent further teaches that "dimethyl ether synthesis is a significantly exothermic reaction" (Shikada, col. 53, ll. 33-34).

3. The Tonkovich patent teaches that "the intrinsic kinetics of a thermal chemical reaction can be much faster than the heat transfer rate between the reaction vessel and the thermal sink, source or environment, [thus] the actual rate of product production . . . is slower than the intrinsic rate" (Tonkovich, col. 1, ll. 29-34), i.e., "the rate at which products could theoretically be formed at the catalyst surface" (*id.* at col. 1, ll. 34-36). The Tonkovich patent teaches that "[l]imited production rates may result from longer residence time which is typically seconds to minutes in conventional

thermal chemical reaction vessels" (*id.* at col. 1, ll. 37-39). Moreover, "[i]n the case of exothermic reactions, low rates of heat removal may promote undesired side reactions, or cause thermal hot spots or thermal runaway in the reactor" (*id.* at col. 5, ll. 25-27).

4. The Tonkovich patent discloses a microchannel reactor with multiple reaction chambers, multiple porous catalyst inserts and multiple heat exchangers (Tonkovich, col. 10, ll. 8-51; Figs. 6, 7) that, unlike conventional reactors, "permit[s] realization of theoretical or near theoretical reaction kinetics" for "catalytic thermal chemical reactions" (*id.* at col. 3, ll. 5-8). The heat exchange capacity of the microchannel reactor is "such that, at steady state, the catalyst is maintained within a temperature range that reduces the formation of at least one undesirable chemical reaction product" (*id.* at col. 4, ll. 15-18). In addition, the heat exchange capacity of the microchannel reactor permits a contact time of less than about 0.3 seconds, thereby reducing the formation of undesirable chemical products that "can result from secondary reactions or slow parallel reactions" (*id.* at col. 4, ll. 18-23).

5. The Tonkovich patent teaches that the microchannel reactor is suitable for methanol synthesis reactions, dehydration reactions, etc., using hybrid catalysts (Tonkovich, col. 11, ll. 12-30). These are the same types of reactions and catalysts involved in the Shikada patent's DME synthesis, but in this case, the hybrid catalyst is not in the form of particles suspended in a slurry. Rather, in the Tonkovich patent, "a preferred catalyst has a porous support, a solution deposited interfacial layer thereon, and a catalyst material on the interfacial layer" (Tonkovich, col. 5, l. 66 - col. 6, l. 1; *compare* FF1).

6. According to the Declaration of Dr. Anna Tonkovich, the Examples and Table 2 of the Specification "show improved results for the synthesis of DME including substantially improved selectivity to the desired product (as shown as lower selectivity to the undesired carbon dioxide) as compared to a conventional slurry process" (Decl. ¶ 5). Dr. Tonkovich declares that "this is a surprising and superior result for the [claimed process . . . as compared to a slurry process such as in the [Shikada] '306 patent" (*id.*). Dr. Tonkovich further declares that she "do[es] not believe that the [Tonkovich] '909 patent predicts substantially improved selectivity as compared to a slurry phase process because a slurry process is well known as a means for limiting reaction isotherms and should be quite isothermal" (*id.*).

7. Table 2 of the Specification is as follows:

Table 2

Performance Comparison of Syngas Conversion to DME in Different Reactor Configurations

reactor configurations	Air Products slurry	microchannel
reaction conditions		
T, °C	250	280
P, atm	52	38
GHSV, h^{-1}	4500	10 238
H_2/CO	0.7	2
performance conversion	37	80
CO_2 selectivity	32	22
specific activity, gCO/g.h	1.19	3.52
liquid yield, g/(g,h)	0.79	2.25

(Specification, Table 2.)

8. Wegeng teaches that a "microchannel reactor is preferably used for reactions that do not require materials or solid that would clog the

microchannels and that do not produce material or solids that would clog the microchannels" (Wegeng, col. 10, ll. 31-34).

DISCUSSION

We agree with the Examiner's rationale and conclusion that it would have been obvious to produce dimethyl ether in a microchannel reactor, given the teachings of the prior art.

We are not persuaded otherwise by Appellants' contention that the Shikada patent's production of dimethyl ether "must occur in the slurry phase" (App. Br. 4), and "there is no suggestion in any of the cited references that Shikada's slurry phase process would have had a reasonable expectation of success when carried out at short contact times in a microchannel reactor" (*id.* at 5). As discussed above, the Tonkovich patent teaches that the microchannel reactor is suitable for methanol synthesis and dehydration, the same types of exothermic reactions, using the same types of hybrid catalysts, as in the Shikada patent's reactions, but on catalyst-coated porous solid supports, rather than in a slurry phase (FFs 1, 4, 5).

To the extent Appellants contend that "there is no reason to have believed that, even if conducted in a microchannel at the claimed contact times, Shikada's slurry-phase process would have succeeded to obtain the claimed levels of conversion" (App. Br. 5), we disagree. The Tonkovich patent teaches that the use of the microchannel reactor allows production at near theoretical reaction kinetics (FF4), and Appellants have not explained why one would not have expected optimal conversion of carbon monoxide under such conditions.

Appellants further contend that "[n]one of the cited references describe[s] a technique for carrying out a slurry phase process in a

microchannel" (App. Br. 5). Appellants contend that Wegeng "teach[es] away from reactions such as slurry phase processes that would contain solid particles that could clog a microchannel" (*id.*).

This argument is not persuasive. Wegeng teaches that microchannel reactors are appropriate for reactions that don't involve materials or solids that would clog the microchannels and that don't produce material or solids that would clog the microchannels (FF8). The Examiner concludes that it would have been obvious to carry out a DME synthesis reaction in a microchannel reactor, not that it would have been obvious to carry out a slurry phase process in a microchannel reactor. One of ordinary skill in the art would understand that Shikada's solvent suspended catalyst particles would not be used in a microchannel reactor like that disclosed in the Tonkovich patent. The catalysts in a microchannel reactor are coated onto porous supports that are inserted in the reactor in the path of the reactants - carbon monoxide and hydrogen, in the case of DME synthesis from syngas (FFs 1, 4, 5). Appellants have not established that materials or solids that could clog the microchannel reactor are formed during DME synthesis from syngas.

Finally, Appellants argue that the use of the microchannel reactor provides a surprising and unexpected degree of selectivity as compared to a conventional slurry process (App. Br. 5; Decl. ¶ 5), and moreover, this improved selectivity is also unexpected in view of the Tonkovich patent because "a slurry process is well known as a means for limiting reaction exotherms and should be quite isothermal" (Decl. ¶ 5; FF6).

This argument is not persuasive. Contact time is also a factor in selectivity, and the Tonkovich patent explains that the shorter reaction time

made possible by the microchannel reactor suppresses undesirable products of secondary reactions or slow parallel reactions (FF4). Therefore, we agree with the Examiner that the results shown in the Examples and Table 2 of the Specification, and discussed in Dr. Tonkovich's declaration would not have been unexpected, given the teachings of the Tonkovich patent.

CONCLUSION

The evidence of record supports the Examiner's conclusion that it would have been obvious for one of ordinary skill in the art to produce dimethyl ether in a microchannel reactor, given the teachings of the prior art relied on. Appellants have not provided evidence of unexpected results sufficient to outweigh the evidence supporting the prima facie case of obviousness.

Accordingly, we affirm the rejection of claims 28 and 30 as unpatentable over Shikada, Tonkovich, Grady, and Wegeng. Claims 29, 31-43, 46-49, 51, and 52 have not been argued separately and therefore fall with claim 28. 37 C.F.R. § 41.37(c)(1)(vii).

SUMMARY

The rejection of claims 28-43, 46-49, 51, and 52 under 35 U.S.C. § 103(a) is affirmed.

TIME PERIOD FOR RESPONSE

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

AFFIRMED

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