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UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE PATENT TRIAL AND APPEAL BOARD

Ex parte ERICH DILK, DETLEF GEISEL, and STEFAN LAMBRECHT

Appeal 2019-006459
Application¹ 15/575,076
Technology Center 1600

Before RICHARD M. LEBOVITZ, ULRIKE W. JENKS, and
RACHEL H. TOWNSEND, *Administrative Patent Judges*.

TOWNSEND, *Administrative Patent Judge*.

DECISION ON APPEAL

This is an appeal under 35 U.S.C. § 134 involving claims to a process for the oxidation of limonene, which have been rejected as obvious. We have jurisdiction under 35 U.S.C. § 6(b).

We affirm.

STATEMENT OF THE CASE

“The aromatic main constituents of citrus essential oils are monoterpenes, sesquiterpenes and the oxygen-containing derivatives thereof.” (Spec. ¶ 2.) “The sensory properties of citrus aromas mainly

¹ We use the word “Appellant” to refer to “applicant” as defined in 37 C.F.R. § 1.42. Appellant identifies the real party in interest as Symrise AG. (Appeal Br. 2.)

depend on the content of oxygen-containing terpene derivatives, alcohols, aldehydes, esters and ketones.” (*Id.*) Limonene is a monoterpene that is in high concentration in citrus oil and “negatively influence[s] the sensory properties of citrus aromas.” (*Id.*) “[I]t is generally known . . . [to be] used as an educt for the production of particular perfumes, aromas or flavours, and also of intermediate compounds thereof.” (*Id.* ¶ 6.) Appellant’s invention “relates to a process for the production of oxidized derivatives of limonene.” (*Id.* ¶ 1.)

Claims 1–21 are on appeal. Claim 1 is representative and reads as follows:

1. A process for the oxidation of limonene, comprising the reaction of limonene with hydrogen peroxide in the presence of a catalyst containing atoms and/or ions of at least one metal selected from the group consisting of molybdenum, tungsten, scandium, titanium, lanthanum, zirconium, praseodymium, neodymium, samarium, europium, terbium, dysprosium, erbium or ytterbium and mixtures thereof, wherein the molecular weight of the catalyst is less than 2,000 g/mol and the reaction is performed at a pH value of more than 8.0.

(Appeal Br. 27.)

The prior art relied upon by the Examiner is:

Name	Reference	Date
Emsenhuber et al.	US2004/0220416 A1	Nov. 4, 2004
Bain et al.	US 3,014,047	Dec. 19, 1961
V. Nardello et al., <i>Olefin oxidation by the system H_2O_2/MoO_4^{2-}: competition between epoxidation and peroxidation</i> , 117 J. Molec. Catalysis A: Chemical, 439–47 (1997) (Nardello 1)		
V. Nardello, <i>Singlet oxygen generation from H_2O_2/MoO_4^{2-}: peroxidation of hydrophobic substrates in pure organic solvents</i> , 43 Tetrahedron Letters, 8731–34, (2002) (Nardello 2)		

The following grounds of rejection by the Examiner are before us on review:

Claims 1–11 and 19–21 under 35 U.S.C. § 103 as unpatentable over Emsenhuber, Nardello 1, and Nardello 2.

Claims 12–18 under 35 U.S.C. § 103 as unpatentable over Emsenhuber and Bain.

DISCUSSION

Obviousness: Claims 1–11 and 19–21

The Examiner finds that Emsenhuber teaches a process for oxidation of organic substrates, including olefins such as tiglic acid and limonene, with 1O_2 via catalytic decomposition of H_2O_2 to give water and 1O_2 . (Final Action 4; Ans. 9.) The Examiner notes that Emsenhuber teaches a number of catalysts including those based on all of the metals identified in claim 1 and teaches the use of a catalyst having a molecular weight of less than 2,000 g/mol as claimed. (*Id.*) The Examiner further finds that Emsenhuber teaches the pH of the reaction mixture can be between 0 and 14, and preferably between 4 and 14, and the particular pH depends on the chosen

substrate and catalyst. (*Id.*) The Examiner notes that Emsenhuber provides a specific example of using Na_2MoO_4 catalyst in a process in which the pH is about 10 which meets the claim limitation of a pH value of more than 8.0. (*Id.* at 5.)

The Examiner recognizes that Emsenhuber does not specifically identify carrying out the reaction of limonene at a pH of more than 8, or more than 9. (*Id.*) However, the Examiner concludes that doing so would have been obvious to one of ordinary skill in the art in light of Emsenhuber and Nardello 1. (*Id.* at 5–6; Ans. 8–12.)

The Examiner finds that Nardello 1 teaches quantitatively creating singlet oxygen, $^1\text{O}_2$, by catalysis of hydrogen peroxide in alkaline media, pH of 9–12, in the presence of molybdate ions. (Final Action 5–6.) The Examiner further finds that Nardello 1 teaches “the pH of the reaction effects selectivity to the epoxidized or peroxidized product.” (*Id.* at 5; Ans. 8.) Thus, explains the Examiner, Nardello 1 “demonstrate[s] that pH is a result effective variable.” (Ans. 8.)

The Examiner finds further that the reaction disclosed in Nardello 1 “is analogous to the reaction disclosed by Emsenhuber et al. [in] that it involves oxidation of an organic substrate, in particular an olefin, by means of $^1\text{O}_2$ that has been generated by H_2O_2 .” (Ans. 9.) Consequently, the Examiner finds that it would have been obvious to one of ordinary skill in the art to conduct the Emsenhuber process with limonene at pH of 9 or above where “one desires to obtain the peroxide compounds” rather than epoxide compounds of limonene. (*Id.*)

The Examiner relies on the teachings of Nardello 2 to address the limitation of claims 19 and 20 that the oxidation reaction be complete in less

than 24 hours. (Final Action 6.) In particular, the Examiner finds that Nardello 2 teaches the rate of molybdate catalyzed H_2O_2 disproportionation to generate $^1\text{O}_2$ “is strongly influenced by temperature.” (*Id.*) The Examiner finds, for example, Nardello 2 teaches that decomposition of 0.5 M H_2O_2 by 0.1 M Li_2MoO_4 at 25 °C takes more than 12 hours but that the reaction time is decreased to only 28 min when carried out at 40 °C. (*Id.*) The Examiner further finds that “although limonene is not the substrate discussed or employed in Nardello et al. 2, the skilled artisan would reasonably expect the teachings of Nardello et al. 2 to be pertinent to limonene, since Nardello et al. 2 uses similar substrates as Emsenhuber et al.” (Ans. 21.)

The Examiner notes further that “temperature is not the only result-effective variable in the reaction of Nardello et al. 2. However, Nardello et al. 2 show that temperature is one of the variables that [a]ffects the reaction time and in particular the temperature is inversely proportional to the reaction time.” (Ans. 15–16.) Consequently, the Examiner concludes that one of ordinary skill in the art would have reasonably expected “that when using a catalyst having the molybdate ions in the process of Emsenhuber et al. that the reaction time can be completed in less than 24 hours by modifying the temperature in the manner taught by Nardello et al. 2.” (Final Action 6.)

We agree with the Examiner’s factual findings and conclusion of obviousness regarding claim 1. As the Examiner explained, Emsenhuber teaches limonene is one of the preferred olefinic organic substrates that can be oxidized with $^1\text{O}_2$, obtained from the chemical catalysis of H_2O_2 in the presence of a molybdenum based catalyst at a pH between 4 and 14. (Emsenhuber ¶¶ 14, 17, 20–29, 72–75.) And, Emsenhuber teaches a specific

example of oxidizing a “test solution” of sodium tiglate, (E)-2-methylcrotonic acid, at pH of about 10, with H₂O₂ in the presence of Na₂MoO₄. (*Id.* ¶¶73–74 (Example 4).) Furthermore, as the Examiner points out (Ans. 11–12), Emsenhuber itself discloses that “it may be advantageous, for better activation of the catalyst, if customary basic and acidic additives are added to the reaction mixture,” (Emsenhuber ¶ 25), and that these additives would necessarily adjust the pH of the reaction mixture. In other words, Emsenhuber teaches that pH is a result effective variable. “[T]he discovery of an optimum value of a variable in a known process is normally obvious.” *In re Antonie*, 559 F.2d 618, 620 (CCPA 1977).

However, even if it were not deemed obvious to carry out the oxidation of limonene as described in Emsenhuber at an alkaline pH of 9 or higher based on Emsenhuber alone, as the Examiner further explained, Nardello 1 demonstrates that the pH of the catalytic system employing H₂O₂ and MoO₄⁻² is result effective where alkaline pH in the range of 9–12 promotes peroxidation over epoxidation of olefins. (*See, e.g.*, Nardello 1 439, 444–46.) Tiglic acid was chosen as a model olefin in the analysis because it is water soluble and known to react both by epoxidation and by peroxidation with ¹O₂. (*Id.* at 443–44.)

In light of the foregoing, we agree with the Examiner that Nardello 1 demonstrates pH is a result effective variable known in the art in the reaction process taught by Emsenhuber for olefinic substrates (Ans. 8), and that one of ordinary skill in the art would have found it obvious to carry out the Emsenhuber reaction at alkaline pH of 9 or more in order to preferentially achieve peroxidation rather than epoxidation (Ans. 9).

We do not find persuasive Appellant’s argument that because Emsenhuber provides an “expansive description of the ‘organic substrate’” that there is “*no guidance* to one skilled in the art as to *which* pH would function for the specifically chosen substrate of limonene” (Appeal Br. 12–13). Nor do we find persuasive Appellant’s argument that because Nardello 1 only discloses oxidation of tiglic acid, one would not have been able to determine that limonene would benefit by the same pH range (Appeal Br. 15–16). One of ordinary skill in the art would have had a reasonable expectation of being able to oxidize each one of the specifically identified olefins (which includes tiglic acid, as well as limonene, and salts thereof (Emsenhuber ¶¶ 17–18)) of Emsenhuber with singlet oxygen made through the catalytic decomposition of H₂O₂ at a pH of between 4 and 14 because Emsenhuber describes each of them as “[e]xamples of particularly suitable [olefin] substrates” for use in the described oxidation reaction. And, Nardello 1 provides a specific reason to use an alkaline pH of greater than 9, i.e., to achieve peroxidation of an olefin rather than epoxidation. *Sinclair & Carroll Co. v. Interchemical Corp.*, 325 U.S. 327, 335 (1945) (“[r]eading a list and selecting a known compound to meet known requirements is no more ingenious than selecting the last piece to put into the last opening in a jig-saw puzzle. It is not invention.”); *see also Merck & Co. Inc. v. Biocraft Laboratories, Inc.*, 874 F.2d 804, 807 (Fed. Cir. 1989). “Only a reasonable expectation of success, not absolute predictability, is necessary for a conclusion of obviousness.” *In re Longi*, 759 F.2d 887, 897 (Fed. Cir. 1985) (“The fact that some titanium compounds function more effectively, and that the exact magnitude of the increased catalytic activity might not be predictable, does not preclude a conclusion of obviousness.”).

Although Nardello 1 chose tiglic acid as the olefin to study, Nardello 1 concludes more generally that in substrates that are subject to epoxidation and peroxidation in the “double reactivity of the system hydrogen peroxide/molybdate ions,” alkaline medium the formation of the intermediate $\text{MoO}(\text{O}_2)_3^{2-}$ allows one to obtain the hydroperoxide via the generation of singlet oxygen, whereas in an acidic medium the formation of $\text{Mo}_2\text{O}_3(\text{O}_2)_4^{2-}$ leads to the epoxide. (Nardello 1 446.) Limonene was an olefin well known to be a substrate that undergoes both epoxidation and peroxidation. (See, e.g., Bain 3:30–36.) For the foregoing reason, we also do not find persuasive of non-obviousness, the fact that Example 4 of Emsenhuber discloses carrying out the reaction at pH 10 with sodium tiglate and not limonene (Appeal Br. 13).²

We also agree with the Examiner that Emsenhuber does not teach away from Nardello 1, as argued by Appellant (Appeal Br. 14–15). One of

² We also do not find persuasive, Appellant’s argument that one of ordinary skill in the art would not combine the teachings of Nardello 1 with Emsenhuber because Nardello 1 used water as the solvent which is in “stark contrast to Emsenhuber.” (Appeal Br. 15.) As the Examiner finds Emsenhuber teaches “[t]he reaction is conducted in a water-miscible organic solvent or in water or in a mixture of water and water-miscible solvent, which includes C1-C8 alcohols and amides.” (Final Action at 4–5; Emsenhuber ¶ 20 (“The oxidation according to the invention takes place in a water-miscible organic solvent **or in water** or in a mixture of water and a water-miscible organic solvent.” (Emphasis added).) Moreover, as the Examiner explains, which Appellant does not respond to,

The presence of water is not taught to be avoided by Emsenhuber et al., instead Emsenhuber et al. provides a means to overcome the presence of water by removing the water with a membrane.

(Ans. 17.)

ordinary skill in the art would not have been discouraged from carrying out the $^1\text{O}_2$ oxidation at an alkaline pH as described in Nardello 1 by the fact that Emsenhuber teaches that single O_2 generated is lost as a result of quenching by water molecules. As the Examiner, explains “Emsenhuber et al. disclose an improved way to carry out the $^1\text{O}_2$ oxidation of Nardello et al. 1 by using membranes to remove the water contained in the reaction mixture (see paragraphs 0009-0012 of Emsenhuber et al.)” (Ans. 12.) Furthermore, as the Examiner explained, Emsenhuber teaches that water is a suitable reaction solvent for the described oxidation process with singlet oxygen catalytically generated from H_2O_2 . (*Id.*)

We also do not find persuasive Appellant’s argument that Nardello 1 describes that choosing the appropriate pH is complex and difficult to forecast (Appeal Br. 15). As the Examiner explains (Ans. 13–14), Nardello 1 states that “the kinetics of the reactions under study are very complex and it is difficult to forecast exactly th[e] pH value [for which both the tetraperoxodimolybdate and triperoxomolybdate oxidizing intermediates are in equal amounts] only by considering the equilibrium constants.” (Nardello 1 446.) It does not say that identifying a pH by routine experimentation to achieve peroxidation of an olefin is difficult to forecast or complex.

Appellant does not separately argue claims 5, 7, 8, or 21 and therefore these claims fall with claim 1. 37 C.F.R. § 41.37(c)(1)(iv).

Regarding the requirement in claims 19 and 20 that the reaction is complete in less than 24 hours, we agree with the Examiner that such would have been obvious in light of the combination of Emsenhuber, Nardello 1, and Nardello 2. Appellant’s argument against this part of the rejection is that there is no illustration in Nardello 2 with data that “with the use of

ethanol solvent an increase in the temperature would influence the reaction rate” and that “[o]ther factors also can play a dramatic role” in reaction time. (Appeal Br. 16.) We do not find these arguments persuasive. First, the Examiner provides where in Nardello 2 data is provided that exemplifies the inverse relationship of temperature to reaction time (Ans. 15 (quoting from Nardello 2 8733)), which Appellant does not respond to. Furthermore, we agree with the Examiner (Ans. 15–16), that while Nardello 2 may not describe temperature as being the only result-effective variable, Nardello 2 demonstrates that it is a variable that is known to be alterable (increasing it) to significantly reduce the reaction time.

Appellant argues dependent claims 3, 4, 6, 9, and 11 separately. (Appeal Br. 17–19.) The Examiner explains why each argument fails to overcome the obviousness argument (Ans. 16–20), and Appellant does not respond. We agree with the Examiner that Appellant has not established non-obviousness of these claims for the reasons expressed by the Examiner.

For the reasons discussed, we affirm the Examiner’s rejection of claims 1–11 and 19–21.

Obviousness: Claims 12–18

The Examiner notes that although Emsenhuber discloses “that hydroperoxides or peroxides can further react to give alcohols (see paragraph 0019),” Emsenhuber does not disclose using “a reduction step of the oxidized limonene to obtain the alcohols or . . . separat[ing] the reduced reaction product by distillation.” (Final Action 7.)

The Examiner finds that Bain discloses a method for the decomposition of peroxides that develop during an oxidation process by

carrying out a reduction under alkaline conditions and then separating the oxidation products via a physical process such as distillation. (*Id.*) The Examiner further finds that Bain teaches that it was “known that terpenic compounds are valuable ingredients of essential oils and are prized individually and/or in combination as perfume ingredients (see column 1, lines 29-34).” (*Id.* at 8.) The Examiner concludes that it would have been obvious to produce an aroma composition for the oxidized limonene compounds of Emesnhuber in light of Bain’s teaching (*id.*) and to use the reduction process of Bain with Emsenhuber to obtain carveol, which is an alcohol product of limonene (*id.* at 7).

We agree with the Examiner’s findings and conclusion of obviousness. Bain teaches that it was known to oxidize limonene with oxygen “and to work up such oxidation products to obtain carvone and carveol” which are prized terpenic compounds. (Bain 1:29–44.) Furthermore, Bain explains that it is “desirable to reduce . . . peroxides” in a process of oxidizing limonene

Prior to attempts to separate the various oxidation products from each other, since the peroxides are unstable and tend to decompose in undesirable directions when heated as in distilling them.

(*Id.* at 4:1–6.) Bain goes on to state:

Through reduction of the peroxides under neutral, preferably alkaline conditions, we thus produce a limonene oxidation mixture which is readily separable into epoxides, p-menthane derivatives oxygenated at the 2-position and p-menthane derivatives oxygenated at the 1-position. We avoid the various decomposition products of the peroxides, oxides and other sensitive compounds and thereby avoid formation of a difficultly separable mixture containing large quantities of high boiling, tar-like products and unsuited to clean separations.

(*Id.* at 4:42–51.)

In light of Emsenhuber teaching oxidation of limonene in an oxidized reaction mixture, we agree with the Examiner that it would have been obvious to include a reduction step “to ensure that peroxides that would hinder the separation of the desired oxidation products would be removed prior to distillation.” (Ans. 21.)

Regarding Appellant’s arguments concerning Bain’s teaching of neutralizing (Appeal Br. 22–23), we agree with and adopt the Examiner’s response (Ans. 22) to which Appellant does not respond.

Regarding Appellant’s argument that Emsenhuber teaches away from claims 15 and 18 (Appeal Br. 24–25), we agree with and adopt the Examiner’s response (Ans. 22–23) to which Appellant does not respond.

Appellant does not separately argue claim 14.

In light of the foregoing, we affirm the Examiner’s rejection of claims 12–18 as being obvious from Emsenhuber and Bain.

DECISION SUMMARY

In summary:

Claims Rejected	35 U.S.C. §	Reference(s)/Basis	Affirmed	Reversed
1–11, 19–21	103	Emsenhuber, Nardello 1, Nardello 2	1–11, 19–21	
12–18	103	Emsenhuber, Bain	12–18	
Overall Outcome			1–21	

Appeal 2019-006459
Application 15/575,076

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). *See* 37 C.F.R. § 1.136(a)(1)(iv).

AFFIRMED