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APPLICATION NO.	FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
12/577,785	10/13/2009	Yasuhiko MATSUOKA	348777US0CONT	5012

22850 7590 09/23/2016  
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ALEXANDRIA, VA 22314

EXAMINER
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BUIE-HATCHER, NICOLE M

ART UNIT	PAPER NUMBER
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1767

NOTIFICATION DATE	DELIVERY MODE
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09/23/2016

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UNITED STATES PATENT AND TRADEMARK OFFICE

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BEFORE  
THE PATENT TRIAL AND APPEAL BOARD

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*Ex parte* YASUHIKO MATSUOKA,<sup>1</sup>  
Kunio Watanabe, and Hiroki Kamiya

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Appeal 2014-008335<sup>2</sup>  
Application 12/577,785  
Technology Center 1700

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Before CATHERINE Q. TIMM, MARK NAGUMO, and  
JEFFREY R. SNAY, *Administrative Patent Judges*.

NAGUMO, *Administrative Patent Judge*.

DECISION ON APPEAL

Yasuhiko Matsuoka, Kunio Watanabe, and Hiroki Kamiya  
("Matsuoka") timely appeal under 35 U.S.C. § 134(a) from the Final  
Rejection<sup>3</sup> of claims 1, 3, 5, 7, 9–11, and 13–17, which are all of the pending  
claims. We have jurisdiction. 35 U.S.C. § 6. We affirm.

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<sup>1</sup> The real party in interest is identified as Asahi Glass Company, Ltd.  
(Appeal Brief, filed 24 April 2014 ("Br."), 1.)

<sup>2</sup> Heard 15 September 2016. Counsel's clear and vigorous advocacy at Oral  
Argument is noted with appreciation.

<sup>3</sup> Office action mailed 13 November 2013 ("Final Rejection").

## OPINION

### A. Introduction<sup>4</sup>

The subject matter on appeal relates to a method of making fluoropolymers by emulsion polymerization in the presence of a fluorocarboxylic acid as the emulsifier, at a pH of 1 to 3.0. (Spec. 1, ll. 7–9.) The Specification reveals that the present reagent of choice for emulsifiers for such polymerization reactions is ammonium perfluoro-octanoate, which is said not to hinder polymerization to high molecular weights by chain transfer reactions. (*Id.* at ll. 12–16.) The ’785 Specification discloses, however, that there are concerns about the accumulation of this emulsifier in the environment as well as adverse health effects. (*Id.* at ll. 20–22.) As a consequence, alternatives are being sought, but, according to the Specification, the yield of polymer per unit time is not improved. (*Id.* at ll. 22–23, *citing* prior art patent documents at ll. 26–27.)

The claimed process is said to provide improved yields of polymer per unit time, as well as high molecular weight. (*Id.* at 3, ll. 10–12.) The etheric oxygen content, fluorine content, and chain length of the fluorocarboxylic acid may be adjusted to modify the surface activity and emulsion stabilization properties. (*Id.* at 4, ll. 14–26.) Moreover, the Specification teaches that using the fluorocarboxylic acid, rather than its salt, means it is not necessary to add a mineral acid or strong organic acid to the salt to adjust

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<sup>4</sup> Application 12/577,785, *Method for producing fluoropolymer using fluorocarboxylic acid compound*, filed 13 October 2009 as a continuation under 35 U.S.C. § 371 of PCT/JP08/56699, filed 3 April 2008, claiming the benefit of an application filed in Japan on 13 April 2007. We refer to the “’785 Specification,” which we cite as “Spec.”

the pH. (*Id.* at 5, ll. 3–6.) “Accordingly,” in the words of the Specification, “the fluorocarboxylic acid is superior to the alkali metal or the ammonium salt of the same fluorocarboxylic acid, since some steps can be omitted in the production and the production is easy.” (*Id.* at 5, ll. 8–10.)

Fluorinated polymer products are said to include high molecular weight modified polytetrafluoroethylene (“PTFE”) that is not melt-processable, melt-processable fluoropolymers, and fluoroelastomers. (*Id.* at 1, ll. 12–14) The emulsion polymerization provides fine powders (*id.* at 9, ll. 14–16) with very low amounts of residual fluorinated emulsifier (*id.* at ll. 1–3), which can be recovered and re-used (*id.* at ll. 6–9). The Specification teaches that the product powder may be used, e.g., as a raw material for molded products, dispersed as an emulsion useful as a coating material, or as a binder or additive. (*Id.* at 13, ll. 4–10.)

Sole independent claim 1 is representative and reads:

A method for producing a fluoropolymer, which comprises  
mixing an aqueous medium with  
a fluoromonomer and  
a polymerization catalyst,  
polymerizing the fluoromonomer by emulsion polymerization  
in the presence of the polymerization catalyst in the  
*aqueous medium containing a fluorocarboxylic acid  
compound* having at most 6 carbon atoms and  
at most 3 etheric oxygen atoms per molecule,  
*wherein the aqueous medium has a pH of 1 to 3.0*  
during a period *beginning before* the polymerization  
catalyst and the fluoromonomer are mixed with the  
aqueous medium *through the end* of the polymerizing, and  
*wherein the fluorocarboxylic acid compound is in the acid form.*

(Claims App., Br. 36; some indentation, paragraphing, and emphasis added.)

The Specification states expressly that “the time period of ‘during the polymerization’ is a period of from the beginning to the end of the polymerization,” and “[t]he pH at the [beginning] of the polymerization means a pH of an aqueous medium before the polymerization catalyst and the fluoromonomer are introduced thereto.” (Spec. 4, ll. 7–10.)

The Examiner maintains the following grounds of rejection:<sup>5</sup>

- A. Claims 1, 3, 7, 9–11, and 13–17 stand rejected under 35 U.S.C. § 103(a) in view of the combined teachings of Nomura<sup>6</sup> and Hintzer.<sup>7</sup>
- A1. Claims 1, 3, 5, 11, 13, and 14 stand rejected under 35 U.S.C. § 103(a) in view of the combined teachings of Nomura and Funaki.<sup>8</sup>
- B. Claims 1, 3, 7, 9–11, and 13–17 stand rejected under 35 U.S.C. § 103(a) in view of the combined teachings of Tanaka<sup>9</sup> and Hintzer.

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<sup>5</sup> Examiner’s Answer mailed 23 May 2014 (“Ans.”).

<sup>6</sup> Junpei Nomura et al., *Process for producing elastic fluorocopolymer and crosslinked fluororubber*, U.S. Patent Application Publication 2007/0167581 A1 (19 July 2007), based on an application filed 3 January 2007; assigned to Asahi Glass Co., Ltd.; H. Kamiya is a common inventor.

<sup>7</sup> Klaus Hintzer et al., *Method of making fluoropolymer dispersion*, U.S. Patent Application Publication 2007/0015864 A1 (18 January 2007), based on an application filed 25 May 2006.

<sup>8</sup> Hiroshi Funaki et al., *Process for the recovery of fluorine-containing emulsifiers*, EP 1 514 848 A1 (2005); assigned to Asahi Glass Co., Ltd.; H. Kamiya is a common inventor.

<sup>9</sup> Hiroyuki Tanaka et al., *Fluorine-containing elastomer composition excellent in plasma-aging prevention effect and shaped article made thereof*, U.S. Patent Application Publication 2006/0235140 A1 (19 October 2006), based on a PCT application filed 21 April 2004.

- B1. Claims 1, 3, 5, 9–11, 13, and 14 stand rejected under 35 U.S.C. § 103(a) in view of the combined teachings of Tanaka and Funaki.

B. Discussion

Findings of fact throughout this Opinion are supported by a preponderance of the evidence of record.

Briefly, the Examiner finds that Nomura and Tanaka disclose emulsion polymerization of fluorinated monomers at acidic pH ranges that overlap (Ans. 2, *citing* Nomura 3 [0052]) or that meet (Ans. 6, *citing* Tanaka 6 [0097]) the range of pH 1–3.0 required by the claims before and during polymerization. The Examiner finds that neither Nomura nor Tanaka discloses a fluorocarboxylic acid as required by claim 1, but finds that both Hintzer (Ans. 2, *citing* Hintzer [0114], [0117] and [0085]) and Funaki (Ans. 4, *citing* Funaki [0049]–[0052]) disclose using such fluorinated carboxylic acids as emulsifiers in the emulsion polymerization of fluorinated monomers. The Examiner reasons that it would have been obvious to use disclosed materials for their designated purposes.

*Rejections A (Nomura)*

Matsuoka urges that although Nomura teaches that the pH for emulsion polymerization is adjusted within a broad range (pH 1 to 9) (Br. 6, ll. 4–7), those broad teachings do not include a teaching or suggestion that the pH be maintained in the range of pH 1 to 3.0 throughout the entire polymerization (*id.* at ll. 16–17). In particular, Matsuoka points out, the pH described in the Examples varies greatly during polymerization. (*Id.* at ll. 17–18, and at para. bridging 15–16, *citing* Nomura 6 [0089]–

[0090], Example 1, “Acidic Polymerization,” starting at pH 7.0 and ending at pH 2.0.) Moreover, as Matsuoka argues, although Nomura does disclose fluorocarboxylic acids as emulsifiers, Nomura “clearly prefers ammonium salts, listing no acids but several salts . . . .” (Br. 18, 1st para., citing Nomura [0038] and [0039].)

Matsuoka urges that Hintzer treats carboxylic acids and their salts as interchangeable. (Br. 16, last para, citing Hintzer [0017].) Moreover, Matsuoka points out, Hintzer exemplifies only the use of ammonium salts. (Br. 16, last para., *citing* Hintzer 10–11 [0170] (Table 1).) Accordingly, in Matsuoka’s view, the Examiner erred in holding that Hintzer would have suggested using fluorocarboxylic acids within the scope of the appealed claims in the processes taught by Nomura.

These arguments are not persuasive of harmful error. As the Examiner points out repeatedly (Ans. 10–12<sup>10</sup>), all of the teachings of a reference, including non-preferred embodiments, must be considered. As for the silence of Nomura on maintaining the pH below 3.0 throughout the polymerization, Nomura’s teachings that the pH at the time of the emulsion polymerization is adjusted from 1 to 7, preferably from 1 to 5 (Nomura 3 [0052]) would have suggested starting the polymerization in the range of pH 1 to 3. Because the evidence of record indicates that the change in hydrogen ion concentration is not large compared to the hydrogen ion concentration in the pH range of 1 to 3,<sup>11</sup> we conclude that the pH would,

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<sup>10</sup> We caution that constant repetition of a principal may diminish the impact of the underlying fact-finding, analysis and reasoning.

<sup>11</sup> In the examples in Nomura as well as in the comparative examples of the ’785 Specification, the pH tends to fall by several pH units, e.g.,

more likely than not, remain within that range during polymerization, and the Examiner's holding of prima facie obviousness therefore is supported by a preponderance of the evidence of record.

Matsuoka urges further that the artisan would not have expected any difference between using an ammonium salt emulsifier and an acid emulsifier in a fluoropolymer production process. (Br. 18, ll. 10–13.) Indeed, Hintzer states, “[f]or the sake of convenience, the term ‘fluorinated carboxylic acid’ is hereinafter used to indicate the free acid as well as salts thereof.” (Hintzer 2 [0017].) In contrast, according to Matsuoka, the acid emulsifiers of Examples 1 and 2 show superior results compared to the corresponding ammonium salt emulsifiers of Comparative Examples 1 and 2 reported in the Specification at pages 11–13 and in Table 1. (*Id.* at 18, ll. 13–18; Table 1 is reproduced on the following page.) In particular, Matsuoka urges that the polymerization rate is substantially higher (about 25% higher for Example 1, about 22% higher for Example 2) than the polymerization rates of the comparative examples. (Br. 19, l. 8.) Similarly, Matsuoka urges that the standard specific gravities (“SSG”) (2.163 compared to 2.173, and 2.167 compared to 2.171) are substantially lower, and indicate high molecular weight and excellence in physical properties. (*Id.* at 20, ll. 20–24.)

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from 6.69 to 2.94 (corresponding to an increase in  $H^+$  concentration from about  $2 \times 10^{-7}$  ( $\log^{-1}(-6.69)$ ) to about  $4 \times 10^{-3}$  ( $\log^{-1}(-2.94)$ ) mol/l. In Example 2 in the Specification, the pH drops from about 2.41 ( $H^+$  concentration of  $3.9 \times 10^{-3}$ ) to about 2.08 ( $H^+$  concentration of  $8.3 \times 10^{-3}$ ). (See Table 1, reproduced *infra*.)



{Table 1 is shown below}

TABLE 1

	Unit	Ex. 1	Comp. Ex. 1	Ex. 2	Comp. Ex. 2
Fluorocarboxylic acid compound		PFDOA	APFDO	PFHxA	APFHx
pH of aqueous medium at the beginning of polymerization	-	2.69	6.69	2.41	6.31
pH of aqueous medium at the end of polymerization	-	2.36	2.94	2.08	2.82
Polymerization time	minute	156	198	212	259
Polymerization rate	g/L·Hr	127	100	94	77
Average particle size of primary particles	μm	0.21	0.24	0.23	0.25
SSG	-	2.163	2.173	2.167	2.171

{Table 1 shows emulsion polymerization conditions and products}

The difficulty with these arguments is that, based on only two Examples and two Comparative Examples, we have little evidence to support the conclusion that the differences are significant (in particular, we have no way to assess the scatter in the data), let alone to determine if the results are unexpected. Weighed against the suggestions by Nomura that acidic emulsion polymerizations in the range of pH 1 to 3 would be useful, discussed *supra*, we find the weight of the evidence insufficient to carry Matsuoka's arguments for unexpected results.

Matsuoka's further arguments for the patentability of dependent claims 5, 7, 10, and 13 rely on the arguments for unexpected results, which, in Matsuoka's view, "unquestionably show unexpected results." (Br. 29, ll. 11–12; 30, l. 12; 31, ll. 12–13; 32, l. 17.) As indicated *supra*, however,

the quantity of the data are not such that the significance of the better results can be assessed as unquestionably showing unexpected results.<sup>12</sup>

Matsuoka urges that the Examiner errs in finding that the fluorine-containing acids listed in Funaki paragraph [0051] are emulsifiers. Rather, according to Matsuoka, “it is the salts of those listed acids that are the emulsifiers for polymerization.” (Br. 22, last two lines.) On this point, the weight of the evidence supports Matsuoka. The grammatical subject of the first sentence of Funaki [0050] is “the fluorine-containing emulsifier,” which is described as “preferably a salt of a perfluoroalkanoic acid, a  $\omega$ -hydroperfluoroalkanoic acid, . . . .” The subject of each of the remaining sentences of Funaki [0050] is the salt. Thus, the acids named in paragraphs [0050], [0051] and [0052] are most naturally read as the acids used to form the salts named in paragraphs [0053]–[0061].

It has not escaped our notice that Funaki is primarily concerned with recovering fluorine-containing emulsifiers (*see* the title), after coagulation of an emulsion-polymerization product. Thus, the teachings of Funaki are not directed towards the conditions of the emulsion polymerization. We have also noted that Funaki teaches that, “[i]n the present invention, the fluorine-

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<sup>12</sup> In the event of further examination, we remind the Examiner of a caution issued long ago by our reviewing court: “Prima facie obviousness is a legal conclusion, not a fact. Facts established by rebuttal evidence must be evaluated along with the facts on which the earlier conclusion was reached, not against the conclusion itself. . . . [A] final finding of obviousness may of course be reached, but such finding will rest upon evaluation of all facts in evidence, uninfluenced by any earlier conclusion reached by an earlier board upon a different record.” *In re Rinehart*, 531 F.2d 1048, 1052 (CCPA 1976), quoted in *In re Piasecki*, 745 F.2d 1468, 1472–73 (Fed. Cir. 1984).

containing emulsifier highly concentrated in the aqueous solution (B) *can be precipitated in the form of a free acid by adjusting the pH of the aqueous solution (B) to an acidity of at most pH 4.*” (Funaki col. 8 [0045].)

On the present record, the weight of the evidence is that the Examiner erred in finding that Funaki teaches free fluorinated acids as emulsifiers, or that Funaki would have suggested that free fluorinated acids be used as emulsifiers. However, the evidence of record indicates that at some pH below pH 4 and above pH 1, the salts would convert to free acids. Thus, the “condition” recited in claim 1, that “the fluorocarboxylic acid compound is in the acid form” is not an independent condition, but merely a statement of fact for that pH range. In particular, claim 1 does not require that the pH be obtained by any particular route. The pH may be obtained by adding an appropriate amount of strong mineral acid to ammonium salts of fluorocarboxylic acids, as well as by adding only fluorinated carboxylic acids. As long as either addition is done before the introduction of the initiator and the fluoromonomer, the conditions defining the period recited in claim 1 would be met.

*Rejections B (Tanaka)*

Matsuoka urges that Tanaka discloses salts of fluorocarboxylic acids as emulsifiers, not the acids themselves. (Br. 8, 1st full para., and 23, last para., both citing Tanaka 6 [0095].) Matsuoka quotes (Br. 25) the most relevant part of Tanaka [0097] cited by the Examiner, which reads,

[o]ther polymerization conditions are not specifically limited, but it is preferable *that the pH of polymerization solution is set as strongly acidic of less than 3* in order to obtain the

polymer product having a carboxyl group at the end and/or a branched chain *without acid treatment described later.*”

(Tanaka 6 [0097].) However, Matsuoka urges that the following two paragraphs, [0098] and [0099], show that “the pH of the polymerization reaction mixture is not maintained at not more than 3 during the entire polymerization process. Rather, the pH must be adjusted to at most 3 by acid treatment in order for coagulation, after the polymerization.” (Br. at para. bridging 25–26.)

Read in context, we find it more plausible to read Tanaka [0098] and [0099] as describing the alternative acid treatment method “described later” in Tanaka [0097]. Thus, Tanaka [0097] describes a process in which a carboxylic acid group is added to the end of the emulsion-polymerized elastomeric fluoropolymer during polymerization, rather than a terminal ammonium or metallic salt is converted to a carboxyl group by an acid treatment following the polymerization.

Matsuoka’s arguments regarding the combinations of the teachings of Hintzer and the teachings of Funaki with the teachings of Tanaka parallel the arguments regarding Nomura, and we find them unpersuasive for the same reasons.

#### C. Order

It is ORDERED that the rejection of claims 1, 3, 7, 9–11, and 13–17 under 35 U.S.C. § 103(a) in view of the combined teachings of Nomura and Hintzer is affirmed.

It is ORDERED that the rejection of claims 1, 3, 5, 11, 13, and 14 under 35 U.S.C. § 103(a) in view of the combined teachings of Nomura and Funaki is affirmed.

It is ORDERED that the rejection of claims 1, 3, 7, 9–11, and 13–17 under 35 U.S.C. § 103(a) in view of the combined teachings of Tanaka and Hintzer is affirmed.

It is ORDERED that the rejection of claims 1, 3, 5, 9–11, 13, and 14 under 35 U.S.C. § 103(a) in view of the combined teachings of Tanaka and Funaki is affirmed.

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