



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<p>(51) International Patent Classification ⁵ : C08K 5/45, 3/16, C01B 7/19, C07C 2/58</p>	<p>A1</p>	<p>(11) International Publication Number: WO 95/16740 (43) International Publication Date: 22 June 1995 (22.06.95)</p>
<p>(21) International Application Number: PCT/US94/08157 (22) International Filing Date: 20 July 1994 (20.07.94) (30) Priority Data: 08/167,010 16 December 1993 (16.12.93) US (71) Applicant: MOBIL OIL CORPORATION [US/US]; 3225 Gallows Road, Fairfax, VA 22037 (US). (72) Inventors: DEL ROSSI, Kenneth, J.; 8 LaTour Court, Wood- bury, NJ 08096-5741 (US). HUSS, Albin, Jr.; 51 Stir- ling Way, Chaddsford, PA 19317-9412 (US). LANDIS, Michael, E.; 26 N. Horace Street, Woodbury, NJ 08096 (US). SAEGER, Roland, Bernard; 207 W. Evesham Road, Runnemedede, NJ 08078-1932 (US). TREWELLA, Jeffrey, C.; 283 Hickory Drive, Kennett Square, PA 19348 (US). (74) Agents: ROBERTS, Peter, W. et al.; Mobil Oil Corporation, 3225 Gallows Road, Fairfax, VA 22037 (US).</p>	<p>(81) Designated States: AU, CA, JP, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Published <i>With international search report.</i></p>	
<p>(54) Title: HYDROGEN FLUORIDE COMPOSITION</p>		
<p>(57) Abstract</p>		
<p>A composition comprising HF and a viscoelastic polymer having an average molecular weight of at least about 100,000 is useful in isoparaffin/olefin alkylation as a substitute for concentrated HF catalyst.</p>		

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HYDROGEN FLUORIDE COMPOSITION

The present invention relates to a composition which contains hydrogen fluoride and which avoids many of the safety and environmental concerns associated with concentrated hydrofluoric acid but which is substantially interchangable with concentrated hydrofluoric acid in many industrial applications.

Hydrogen fluoride, or hydrofluoric acid (HF) is highly toxic and corrosive, but is useful as a catalyst in a variety of isomerization, condensation, polymerization and hydrolysis reactions. As used herein, the term "concentrated hydrofluoric acid" refers to an essentially anhydrous liquid containing at least about 85 weight percent HF. The petroleum industry uses anhydrous hydrogen fluoride primarily as a liquid catalyst for alkylation of olefinic hydrocarbons to produce alkylate for increasing the octane number of gasoline. Years of experience in its manufacture and use have shown that HF can be handled safely, provided the hazards are recognized and precautions taken.

Alkylation is a reaction which adds an alkyl group to an organic molecule. Thus an isoparaffin can be reacted with an olefin to provide an isoparaffin of higher molecular weight. The principal alkylation reaction of interest in petroleum refining reacts a C₂ to C₃ olefin with isobutane in the presence of an acidic catalyst producing a so-called alkylate. This alkylate is a valuable blending component in the manufacture of gasolines due not only to its high octane rating but also to its compatibility with octane-enhancing additives such as ethers.

Given the many and varied uses for HF, it would be desirable to provide a safer HF composition which may readily be substituted for concentrated HF in commercial applications such as isoparaffin/olefin alkylation.

Accordingly, this invention provides a composition comprising HF and a viscoelastic polymer having a molecular weight of at least about 100,000.

The concentration of the viscoelastic polymer additive typically ranges from 1 to 5000 ppm, preferably from 5 to 2000 ppm, more preferably from 10 to 1500 ppm. The composition of the invention is typically characterized by extensional viscosities within the range of from 10 to 10,000 cP, preferably from 100 to 10,000 cP, and more preferably from 1000 to 10,000 cP. The composition preferably contains no added metal halide. The composition may optionally contain other components, for example, water or other solvents which are miscible with HF.

The composition of this invention is useful as an isoparaffin/olefin alkylation catalyst in accordance with the disclosures of U.S. Patents 5,196,628 to Del Rossi and Huss, and 5,202,518 to Del Rossi.

The viscoelastic polymer employed in the composition of the invention is not a surfactant. As used herein, the term surfactant is intended to indicate a compound which satisfies many of the six fundamental characteristics which are generally understood to be associated with surfactants: solubility in at least one phase of a liquid system, an amphiphatic structure, the tendency to form oriented monolayers at phase interfaces, preferential equilibrium concentration at a phase interface as compared to the bulk of a solution, micelle formation, and the possession of some combination of the functional properties of surfactants, such as detergency, foaming, wetting, emulsifying, solubilizing, and dispersing. For a general discussion of surfactant properties, see 22 Kirk-Othmer Encyclopedia of Chemical Technology 332 (2nd ed., 1969).

The viscoelastic polymer of this invention may comprise more than one monomer and may comprise a random copolymer. Block copolymers which function as surfactants are not preferred for use in the present invention, and the composition of the invention preferably excludes block copolymers.

The invention further comprises a method for rendering

a liquid hydrofluoric acid composition less susceptible to forming a tenaceous vapor cloud, said method comprising adding a viscoelastic polymer to said composition in an amount sufficient to increase the extensional viscosity of
5 said composition.

For a discussion of other additives useful for mitigating the tenacity of HF vapor clouds, see U.S. Patent 4,938,935 to Audeh et al.

The invention still further comprises a process for
10 alkylating an isoparaffin with an olefin comprising contacting said isoparaffin and said olefin in the presence of the composition of the invention to produce alkylate.

Solvents

The composition of this invention comprises HF and a
15 viscoelastic polymer having a molecular weight of at least about 100,000. The composition may optionally comprise a solvent which is not deleterious to the viscoelastic behavior of the mixture. Solvents which may be added to the mixture of HF and viscoelastic polymer in accordance
20 with the invention include, but are not limited to, those solvents characterized by a Donor Number of less than about 40.

The term "donicity" describes the propensity of a solvent to donate electron pairs to acceptor solutes. The
25 term "Donor Number" (DN) as used herein is a measure of donicity, and is defined as the negative of the enthalpy change, measured in $\text{Kcal}\cdot\text{mol}^{-1}$, for the reaction of the solvent with SbCl_5 to form a 1:1 adduct, where both reactants are in dilute solution in 1,2-dichloroethane
30 (DCE). For a discussion of donicity and Donor Numbers, see Y. Marcus, "The Effectivity of Solvents as Electron Pair Donors", 13 Journal of Solution Chemistry 599 (1984). Table 1 below reports donor numbers listed in the Marcus article for various solvents.

35 Solvents useful in the present invention include nitroalkanes, carbonates, perhalogenated alkanes,

halogenated alcohols, sulfonic acids, sulfones, acetyl halides, benzoyl halides, phosphorous oxychloride, alkyl sulfites, anhydrides, esters, and sulfuryl halides. Non-limiting examples of these additives include nitromethane, 5 1-nitropropane, propylene carbonate, perfluorodecalin, 2,2,2-trifluoroethanol, methanesulfonic acid, a low donicity solvent, acetyl chloride, benzoyl fluoride, methyl propionate, sulfuryl chloride, and sulfuryl chloride fluoride.

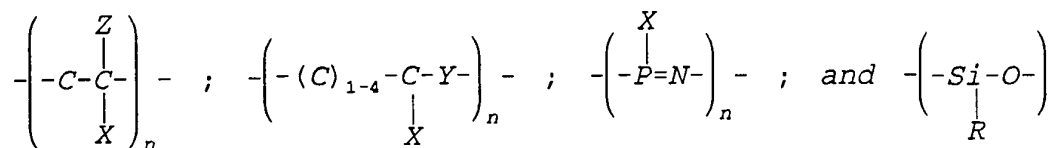
Table 1

	<u>Solvent</u>	<u>DN</u>	<u>Solvent</u>	<u>DN</u>
	1,2-dichloroethane	(0)	Methyl-t-butylketone	17.0
	Acetyl Chloride	0.7	Diethyl Ether	19.2
	Benzoyl Chloride	2.3	Tetrahydrofuran	20.0
	Sulfuryl Chloride	0.1	Triethylamine	30.5
5	Thionyl Chloride	0.4	Pyridine	33.1
	Selenoyl Chloride	12.2	Acetonitrile	14.1
	Phosphoryl Chloride	11.7	Propanonitrile	16.1
	Tetrachloroethylene		Butanonitrile	16.6
	Carbonate	0.8	Isobutanonitrile	15.4
10	Dichloroethylene		Benzyl Cyanide	15.1
	Carbonate	2.7	Benzonitrile	11.9
	Nitromethane	2.7	N,N-Dimethylformamide	26.6
	Nitrobenzene	4.4	N,N-Diethylformamide	30.9
	Acetic Anhydride	10.5	N,N-Dimethylacetamide	27.8
15	Methyl Acetate	16.4	N,N-Diethylacetamide	32.2
	Ethyl Acetate	17.1	Tetramethyl Urea	29.6
	2-Propyl Acetate	17.5	Hexamethyl Phosphoric	
	Ethyl Propanoate	17.1	Triamide	38.8
	Ethyl Butanoate	16.8	Ethylene Sulfite	15.3
20	Ethyl Isobutanoate	16.4	Dimethylsulfoxide	29.8
	Ethyl t-Pentanoate	12.9	Tetramethylene Sulfone	14.8
	Diethylcarbonate	16.0	Phenyldifluorophosphine	
	Ethylene Carbonate	16.4	Oxide	16.4
	1,2-Propylene		Phenyldichlorophosphine	
25	Carbonate	15.1	Oxide	18.5
	Acetone	17.0	Diphenylchlorophosphine	
			Oxide	22.4
	2-Butanone	17.4	Methylisopropyl-ketone	17.1
	Trimethyl Phosphate	23.0	Tri-n-butyl Phosphate	23.7

Viscoelastic Polymer

The viscoelastic polymers useful in the invention generally range in molecular weight from 10^5 to 3×10^7 , preferably from 10^6 to 3×10^7 , and more preferably from 10^7 to 3×10^7 . The viscoelastic polymers may comprise one or more monomers. Random copolymers are useful in the invention, and while some block copolymers are useful, block copolymers which function as surfactants are not preferred for the present invention. Thus the viscoelastic polymers of the invention preferably exclude block copolymers.

Viscoelastic polymers useful in this invention include polymers having the following structures:



wherein X and Z are selected from H, alkyl groups, alkenyl groups, halogens, substituted and unsubstituted phenyl groups, CONH_2 , CONHR , $\text{CONR}'\text{R}$, COOR , and $\text{C}\equiv\text{N}$; wherein Y is selected from O, NR, and NX; wherein R and R' are selected from hydrogen and alkyl groups.

Examples of useful viscoelastic polymer additives are disclosed in the following U.S. Patents.

U.S. Patent 4,089,804 to Falk discloses alkeneoxyalkenes and alkyleneiminoalkenes of 2 to 12 carbon atoms at column 1, line 64, through column 2 at line 10.

U.S. Patent 4,257,903 to Kucera et al. discloses useful viscoelastic polymers at column 1, line 61, through column 2, line 24.

U.S. Patent 4,363,886 to Lipowski et al. discloses monomers which can be polymerized by convention means to

produce polyacrylamides useful in the present invention.
See column 3, lines 4-64.

U.S. Patent 4,505,828 and 4,552,670 to Lipowski et al.
similarly disclose monomers which can be polymerized by
5 known methods to produce polyacrylamides useful in the
present invention.

U.S. Patent 4,599,372 to Bardoliwalla et al. discloses
useful viscoelastic polymers at column 5, line 33 through
column 6 at line 29.

10 U.S. Patent 4,646,834 to Bannister discloses useful
viscoelastic polymers at column 2, lines 3-38.

U.S. Patent 4,796,703 to Gabel et al. discloses
polymeric flocculating agents (which include viscoelastic
polymers useful in the present invention) at column 2,
15 lines 15-63.

U.S. Patent 4,847,342 to Peiffer discloses
polyacrylamide polymers at column 3, lines 10-26, and at
column 4, lines 3-59.

U.S. Patent 4,913,585 to Thompson et al. discloses
20 useful polyacrylamide polymers at column 3, line 57,
through column 4 at line 34.

U.S. Patent 5,071,934 to Peiffer discloses useful
viscoelastic polymers at column 2, lines 52-69.

U.S. Patent 5,132,284 to Tsai discloses useful viscoelastic
25 polymers at column 1, line 66, through column 3, line 56.

U.S. Patent 5,132,285 to Tsai discloses useful
viscoelastic polymers at column 1, line 67, through column
3, line 56.

Feedstocks

30 Feedstocks useful in the alkylation process of the
invention include at least one isoparaffin and at least one
olefin. The isoparaffin reactant used in the present
alkylation process has from 4 to 8 carbon atoms.
Representative examples of such isoparaffins include
35 isobutane, isopentane, 3-methylhexane, 2-methylhexane, 2,3-
dimethylbutane and 2,4-dimethylhexane.

The olefin component of the feedstock includes at least one olefin having from 2 to 12 carbon atoms. Representative examples of such olefins include butene-2, isobutylene, butene-1, propylene, ethylene, hexene, octene, and heptene. The preferred olefins include the C₄ olefins, for example, butene-1, butene-2, isobutylene, or a mixture of one or more of these C₄ olefins, with butene-2 being the most preferred. Suitable feedstocks for the process of the present invention are described in U.S. Patent 3,862,258 to Huang et al. at column 3, lines 44-56.

The molar ratio of isoparaffin to olefin is generally from 1:1 to 100:1, preferably from 1:1 to 50:1, and more preferably from 5:1 to 20:1.

Process Conditions

The composition of the present invention may be readily substituted for the concentrated hydrofluoric acid catalyst in an existing hydrofluoric acid alkylation process unit, for example, a riser reactor alkylation process unit, without substantial equipment modifications. Accordingly, the conversion conditions for the process of the present invention resemble those of typical commercial hydrofluoric acid alkylation processes.

The present alkylation process is suitably conducted at temperatures of from -18 to 66°C (0 to 150°F), preferably from 10 to 66°C (50 to 150°F), and more preferably from 21 to 43°C (70 to 110°F). Pressure is maintained to ensure a liquid phase in the alkylation reaction zone and typically ranges from 240 to 8375 kPa (20 to 1200 psig), preferably from 445 to 3550 (50 to 500 psig). The reaction zone is preferably free from added hydrogen. Olefin feed rates generally range from 0.01 to 50 WHSV and more preferably from 0.5 to 20 WHSV. The mixed isoparaffin-olefin reactants may be contacted with the catalyst composition of the invention in any suitable reaction vessel, examples of which include stirred-tank reactors as well as riser-type reactors. Contact time for

the mixed isoparaffin-olefin feed and the catalyst composition of the invention typically are within the range of from 0.1 second to 50 seconds, and are preferably from 8 seconds to 25 seconds.

5 The relative amounts of catalyst and reactants are defined herein by the acid-to-oil ratio. The volumetric acid-to-oil ratio (as used herein) is the ratio of the sum of the volumes of ASO (acid soluble oil), acid, a suitable solvent (if present), and the viscoelastic polymer to the
10 total isoparaffin and olefin reactor feed. The volumetric acid-to-oil ratio typically falls within the range of 0.1:1 to 10:1, preferably from 0.1:1 to 5:1.

 The viscoelastic polymer may be added by injection directly into the alkylation process unit, or may be mixed
15 with the hydrocarbon charge, or may be mixed with the fresh and/or the circulating acid catalyst component, or with a stream of mixed acid/additive catalyst. Downstream from the alkylation reaction zone, the viscoelastic polymer may be separated from the alkylate product stream, mixed with
20 fresh and/or circulating acid and/or circulating acid/additive catalyst mixture, and recycled to the alkylation reaction zone. Alternatively, the viscoelastic polymer may be used in a concentration which is sufficiently low (i.e. ~50 ppm) to eliminate the economic
25 incentive for recovering and recycling the viscoelastic polymer.

 The invention will now be more particularly described with reference to accompanying drawings, in which:

 Figure 1 is a simplified schematic diagram showing a
30 ductless siphon apparatus useful for measuring the viscoelasticity (expressed in terms of extensional viscosity, μ_e) for catalysts of the present invention.

 Figure 2 is a plot of ductless siphon height (the y-axis, mm) as a function of ionic strength (the x-axis, mol/l) for the solutions of Examples 1-7.
35

 Figure 3 is a plot of ductless siphon height (the

y-axis, mm) as a function of polymer concentration (the x-axis, ppm) for the solutions of Examples 8-11.

Figure 4 is a plot of ductless siphon height (the y-axis, mm) as a function of polyacrylamide additive
5 molecular weight for the solutions of Example 12.

Examples 1-7

Referring now to Figures 1 and 2, the ductless siphon as schematically illustrated provides a direct measurement of polymer/solvent system's viscoelasticity. See, e.g.,
10 G. Astarita, et al., "Extensional Flow Behaviour of Polymer Solutions" 1 Chem. Eng. J. 57 (1970). In this apparatus, an applied pressure to the free surface of the polymer/solvent in a reservoir 11 generates a free-standing column of fluid 12 when a nozzle 13 is withdrawn from the
15 polymer/solvent. The height, h , of the fluid column (or siphon) at snap-off, when column self-weight balances the elastic forces generated within the fluid, measures the polymer/solvent system's anti-misting tendencies. See, e.g., K.K. Chao, et al. "Antimisting Action of Polymeric
20 Additives in Jet Fuels", 30 AIChE J. 111 (1984). Thus the siphon height, h , measured when polymer is added to the HF/sulfolane or HF/solvent mixture indicates the degree of potential rainout-improvement. For a given flowrate, Q , and a given nozzle diameter, (and thus a given upward fluid
25 velocity v_x) the siphon breakage height, h , is proportional to $(\mu_e/\mu_s)^{1/2}$, where μ_e is extensional viscosity and μ_s is the shear viscosity.

Examples 1 and 2 examined the behavior of polyethylene oxide (5,000,000 mw) in aqueous HCl at polymer
30 concentrations of 1000 ppm (Example 1) and 100 ppm (Example 2) as a function of ionic strength in solution. The effectiveness of the polyethylene oxide (PEO) additive decreased with increasing ionic strength.

Example 3 examined the behavior of polyethylene oxide
35 (5,000,000 mw) in NaCl at polymer concentration of 100 ppm as a function of ionic strength in solution. The

effectiveness of the polyethylene oxide (PEO) additive decreased with increasing ionic strength.

Examples 4 and 5 examined the behavior of polyacrylamide (5,000,000 mw) in aqueous HCl at polymer concentrations of 1000 ppm (Example 4) and 100 ppm (Example 5) as a function of ionic strength in solution. The effectiveness of the polyacrylamide (PAM) additive surprisingly increased with increasing ionic strength in solution.

Example 6 examined the behavior of polyacrylamide (PAM) (5,000,000 mw) in NaCl at polymer concentration of 100 ppm as a function of ionic strength in solution. The effectiveness of the polyethylene oxide (PEO) additive surprisingly increased with increasing ionic strength.

Example 7 examined the behavior of polyethylene oxide (5,000,000 mw) in a solution of HF and sulfolane (49/49/2 wt/wt/wt HF/sulfolane/PEO). The effectiveness of the polyethylene oxide (PEO) additive decreased with increasing ionic strength.

20 Examples 8-11

Referring now to Figure 3, Examples 8-11 demonstrate the effect of polymer concentration (x-axis, ppm) on ductless siphon height (y-axis, mm). Increasing polymer concentration improved ductless siphon height for solutions of PEO or PAM (5,000,000 m.w.) with water or aqueous (9.2 M) HCl.

Example 12

Figure 4 (Example 12) shows the effect of polyacrylamide (PAM) polymer molecular weight (x-axis) on ductless siphon height (y-axis, mm). Higher molecular weight viscoelastic additives are preferred for use in accordance with the present invention.

Examples 13-25

Examples 13-25 show the ductless siphon heights for HF/sulfolane/water mixtures. Examples 13-16 show the ductless siphon heights for HF/sulfolane/water mixtures

with the addition of PEO, while Examples 17-25 show the ductless siphon heights for HF/sulfolane/water mixtures with the addition of PAM.

Table 2

Ductless Siphon Heights

Example No.	Solvent	Polymer	Polymer conc., ppm	siphon height, mm
13	water	PEO	100	12
14	water	PEO	500	17
15	water	PEO	1000	25
16	HF/sulfolane/ water 49/49/2 (weight)	PEO	1000	1-2
17	HF/sulfolane/ water 49/49/2 (weight)	Nalco 9905	100	5
18	HF/sulfolane/ water 69/29/2 (weight)	Nalco 9905	100	3
19	HF/sulfolane/ water 69/29/2 (weight)	Nalco 9905	1000	15
20	HF/sulfolane/ water 84/14/2 (weight)	Nalco 9905	1000	23

Example No.	Solvent	Polymer	Polymer conc., ppm	siphon height, mm
21	HF/sulfolane/ water 69/29/2 (weight)	Magnafloc 905N	100	7
22	HF/sulfolane/ water 69/29/2 (weight)	Magnafloc 905N	1000	15
23	HF/sulfolane/ water 69/29/2 (weight)	Magnafloc 496C	100	2
24	HF/sulfolane/ water 69/29/2 (weight)	Magnafloc 496C	1000	16
25	HF/sulfolane/ water 69/29/2 (weight)	Magnafloc 866A	100	27

Table 3
Polymer Properties

Polymer	Description	Supplier	Molecular Weight, millions
PEO	polyethylene oxide	Aldrich	3-5
Nalco 9905	cationic polyacrylamide	Nalco	>1
Magnafloc 866A	anionic polyacrylamide	American Cyanimid	16-19
Magnafloc 905N	nonionic polyacrylamide	American Cyanimid	14-17
Magnafloc 496C	cationic polyacrylamide	American Cyanimid	3-5

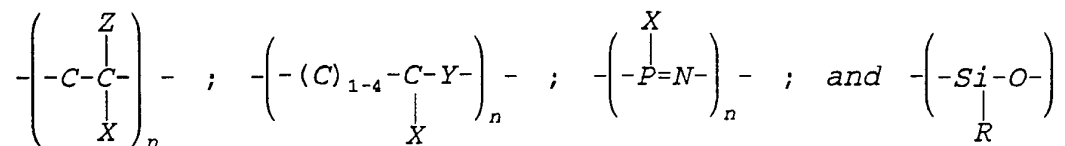
Claims:

1. A composition comprising HF and a viscoelastic polymer having an average molecular weight of at least 100,000.

2. A composition as claimed in claim 1 including 1 to 5000 ppm of said viscoelastic polymer.

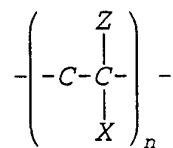
3. A composition as claimed in claim 1 including 10 to 1500 ppm of said viscoelastic polymer.

4. A composition as claimed in any preceding claim wherein said viscoelastic polymer has a structure selected from:



wherein X and Z are selected from the group consisting of H, alkyl groups, alkenyl groups, Br, Cl, I, F, substituted and unsubstituted phenyl groups, CONH₂, CONHR, CONR'R, COOR, and C≡N; wherein Y is selected from the group consisting of O, NR, and NX; wherein R and R' are the same or different, and are selected from the group consisting of hydrogen and alkyl groups.

5. A composition as claimed in claim 4 wherein said viscoelastic polymer has the structure



6. A composition as claimed in claim 4 or claim 5 wherein X is CONH_2 .

7. A composition as claimed in any one of claims 4 to 6 wherein n is at least about 10,000.

8. A composition as claimed in any preceding claim wherein said viscoelastic polymer has a molecular weight of at least about 10^6 .

9. A composition as claimed in claim 8 wherein said viscoelastic polymer has a molecular weight of from 10^7 to 3×10^7 .

10. A composition as claimed in any preceding claim further comprising a solvent having a Donor Number of less than about 40.

11. A composition as claimed in any preceding claim further comprising sulfolane.

12. A method for rendering a liquid hydrofluoric acid composition less susceptible to forming a tenaceous vapor cloud, said method comprising adding to said composition a viscoelastic polymer having an average molecular weight of at least 100,000 in an amount sufficient to increase the extensional viscosity of said composition.

13. A process for alkylating an isoparaffin with an olefin comprising reacting said isoparaffin with said olefin in the presence of an alkylation catalyst comprising HF and a viscoelastic polymer having an average molecular weight of at least about 100,000 wherein said viscoelastic polymer is present in an amount sufficient to enhance the extensional viscosity of the composition.

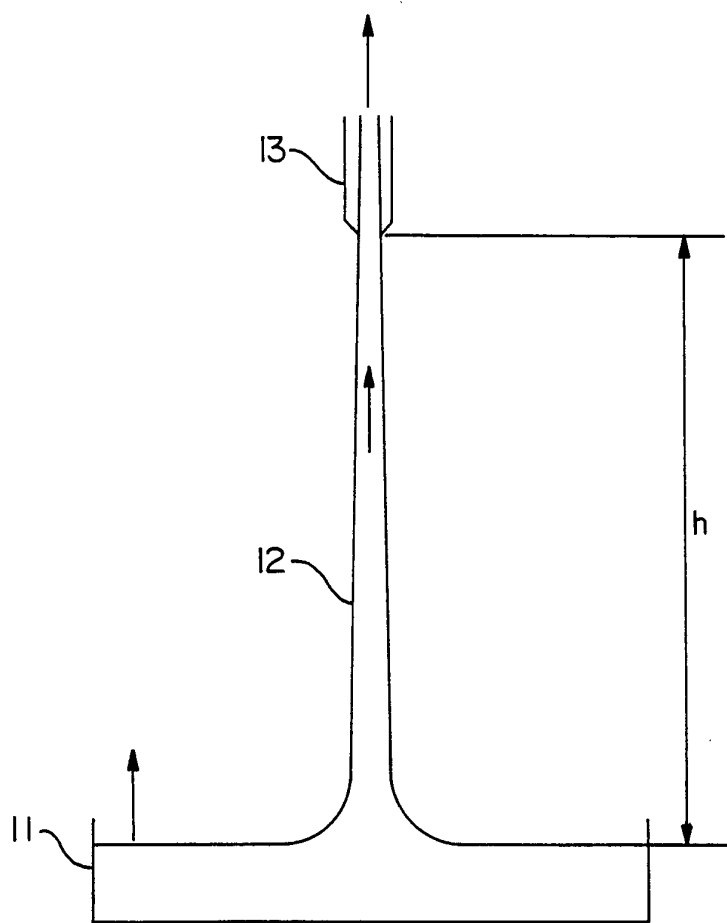


FIG. 1

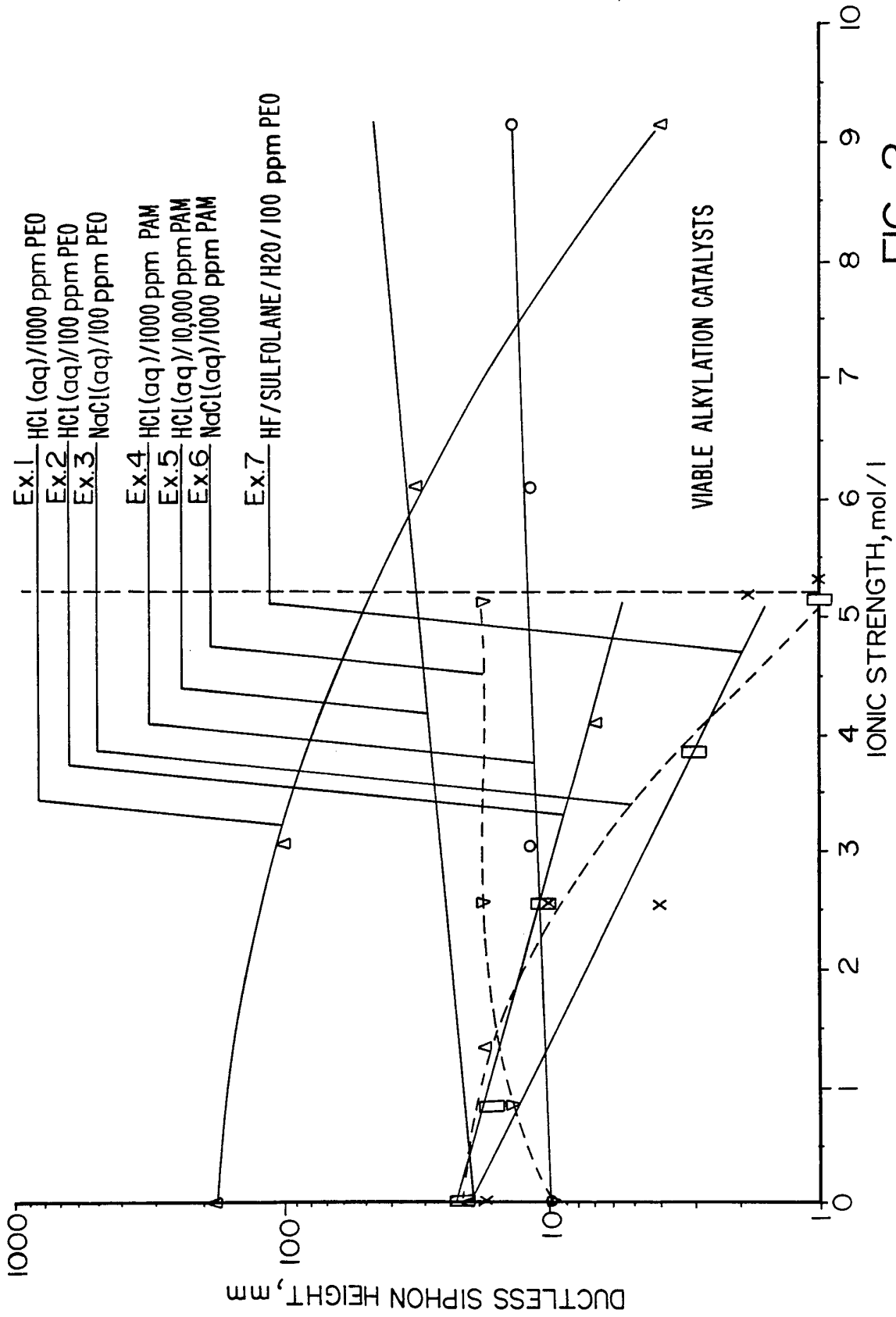


FIG. 2

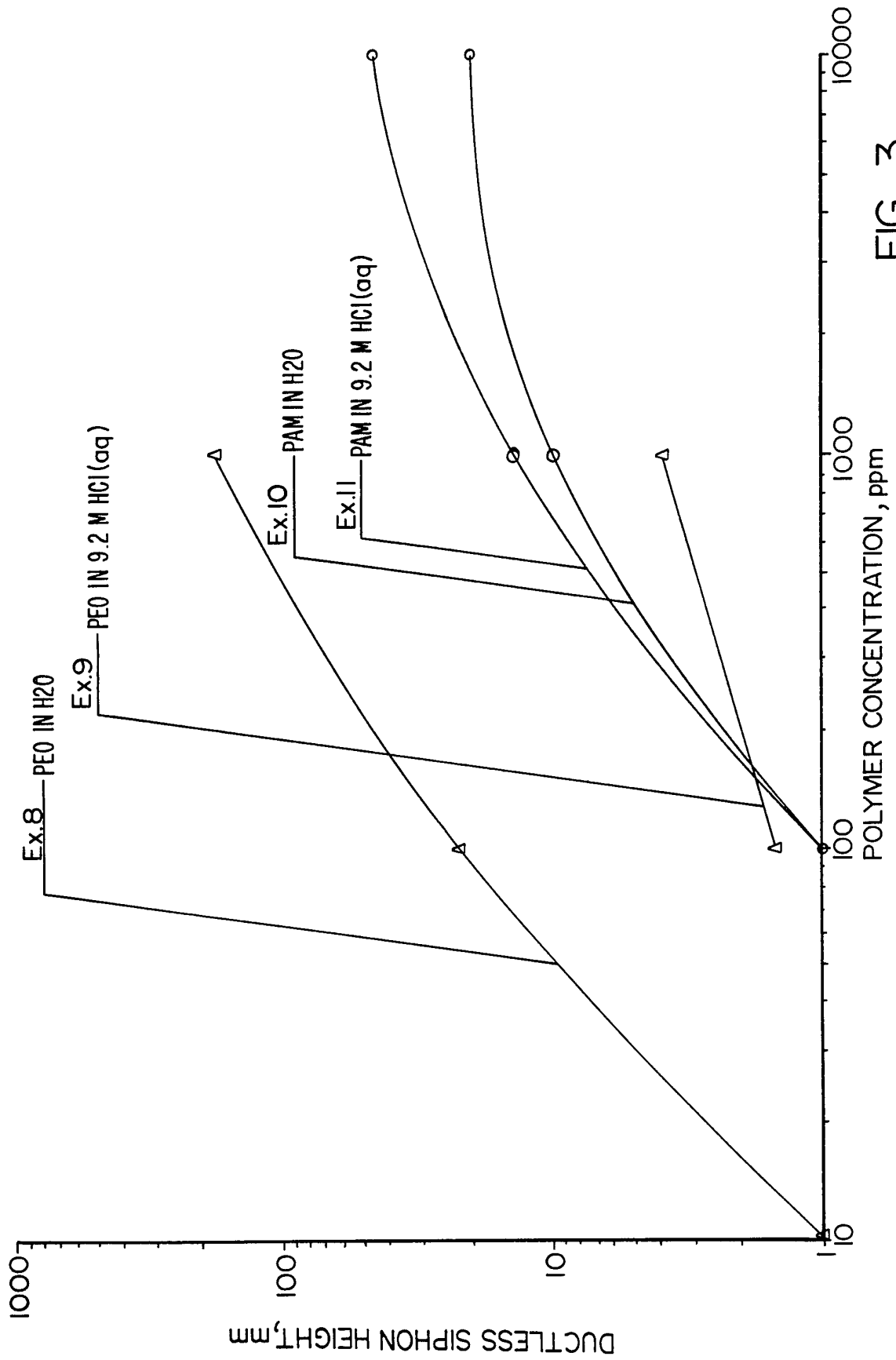


FIG. 3

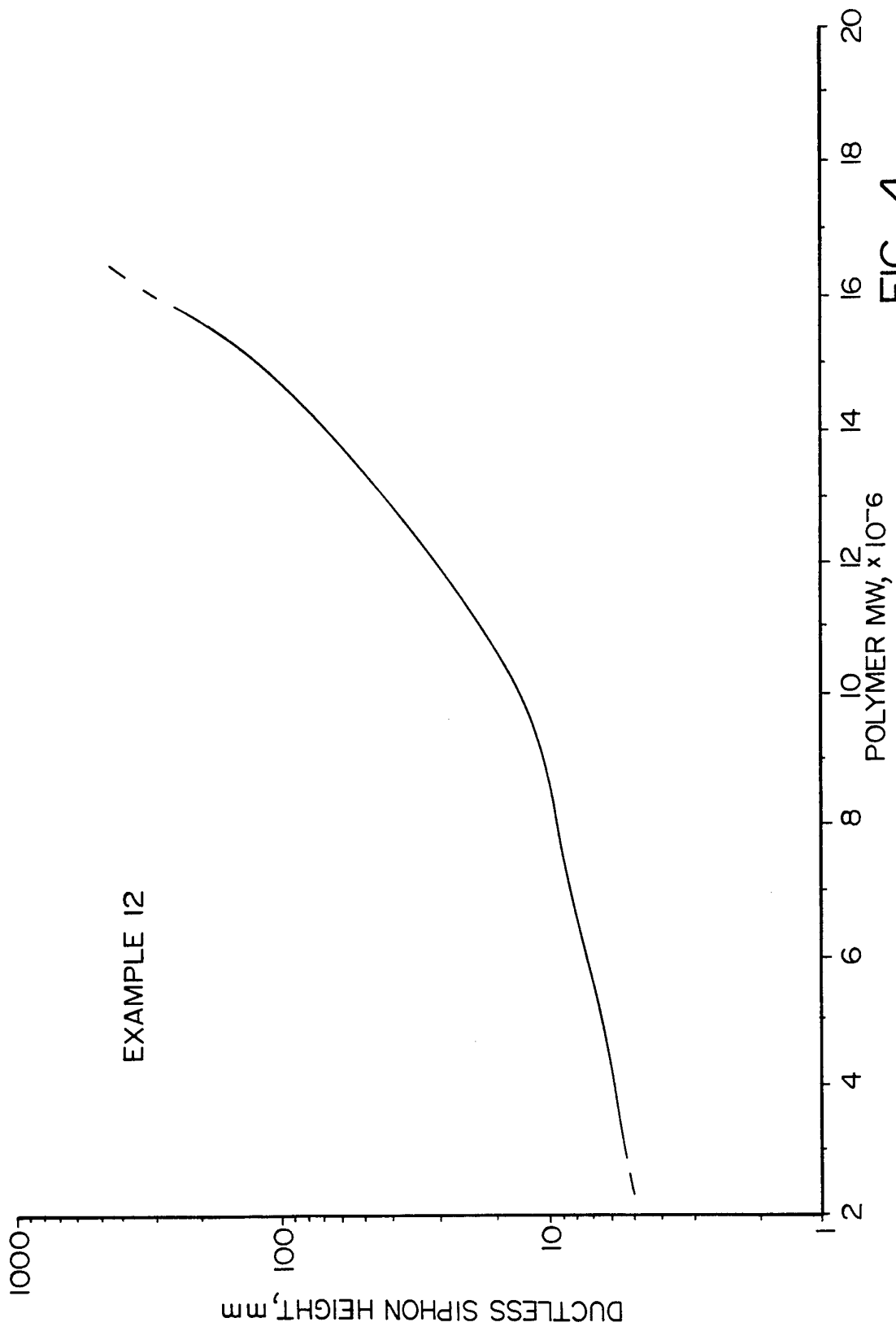


FIG. 4

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US94/08157

A. CLASSIFICATION OF SUBJECT MATTER
 IPC(5) : C08K 5/45, 3/16; C01B 7/19; C07C 2/58
 US CL : 524/84, 401; 423/483; 585/721, 723
 According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED
 Minimum documentation searched (classification system followed by classification symbols)
 U.S. : 524/84, 401; 423/483; 585/721, 723

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X, P	US, A, 5,286,456 (SCHATZ) 15 February 1994. See entire document.	1-5, 12, 13
X	US, A, 5,191,150 (CHILD et al.) 02 March 1993. See Abstract and Examples.	1-5, 12, 13
A	US, A, 4,795,728 (KOCAL) 03 January 1989. See entire document.	1-5, 12, 13

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/US94/08157**Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)**

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. Claims Nos.: 6-11
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- The additional search fees were accompanied by the applicant's protest.
 No protest accompanied the payment of additional search fees.