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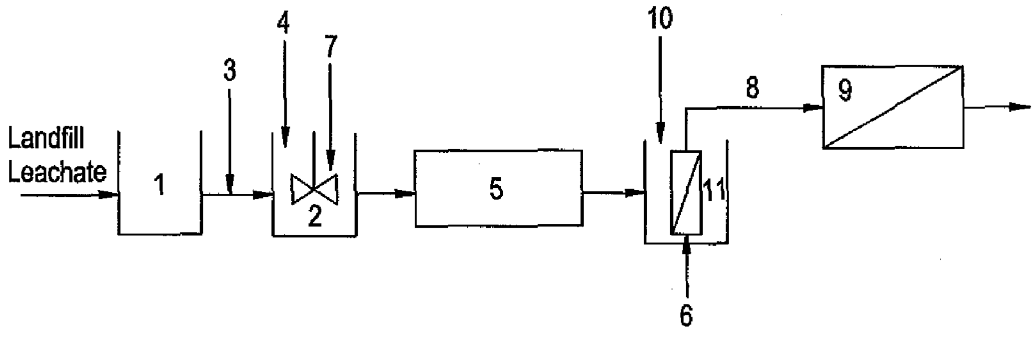
(54) Title: METHOD OF IMPROVING PERFORMANCE OF ULTRAFILTRATION OR MICROFILTRATION MEMBRANE PROCESS IN LANDFILL LEACHATE TREATMENT



(57) Abstract: A method of processing landfill leachate by use of a membrane separation process is disclosed. Specifically, the following steps are taken to process landfill leachate: collecting landfill leachate in a receptacle suitable to hold said landfill leachate; treating said landfill leachate with one or more water soluble polymers, wherein said water soluble polymers are selected from the group consisting of: amphoteric polymers; cationic polymers; zwitterionic polymers; and a combination thereof; optionally mixing said water soluble polymers with said landfill leachate; passing said treated landfill leachate through a membrane, wherein said membrane is an ultrafiltration membrane or a microfiltration membrane; and optionally back-flushing said membrane to remove solids from the membrane surface.

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METHOD OF IMPROVING PERFORMANCE OF ULTRAFILTRATION OR MICROFILTRATION MEMBRANE PROCESS IN LANDFILL LEACHATE TREATMENT

FIELD OF THE INVENTION

This invention pertains to a method of processing landfill leachate via the use of a membrane system including a microfiltration membrane or an ultrafiltration membrane.

BACKGROUND

Landfill leachate is a wastewater stream generated after rainwater percolates through the landfill of industrial or municipal solid waste. This stream contains high levels of contaminants such as suspended solids, colloidal material, bacteria, heavy metals and other soluble organics. Therefore, due to environmental concerns and regulations, the landfill leachate cannot be discharged in a river or other water bodies without treatment. Various membrane separation processes are utilized to combat this problem. Ultrafiltration (UF) and Microfiltration (MF), optionally followed by Nanofiltration (NF) or Reverse Osmosis (RO) membrane processes, are increasingly being used for removing the above-mentioned contaminants. The water treated by this method is very pure and can be discharged into bodies of water, or be reused, for example, as boiler feed water or for irrigation.

Although water quality obtained with these membrane processes is very high compared to the use of clarifiers or media filters, fouling of UF or MF membranes by landfill leachate contaminants results in reduced flux in membranes and an increase in frequency of membrane cleaning. As a result of the reduced flux rate, the number and / or size of UF/MF membranes needs to be increased, especially in the case where large amounts of wastewater are being processed. The requirement that membranes need to be larger and / or in greater number is problematic in that it increases capital costs. The requirement of frequent membrane cleaning is also problematic in that it increases operating cost. All of these problems make the process of using UF or MF membranes uneconomical to operate.

Therefore, it is of interest to minimize membrane fouling so that membranes: operate for a longer period between cleanings; operate at a rate of flux in accord with the chosen membrane; operate at higher than currently achievable fluxes; or a combination thereof. In addition, it of interest to lower the number and /or size of the membranes so that capital costs of new systems containing UF/MF membranes are lowered, especially when processing a large amount of wastewater.

SUMMARY OF THE INVENTION

The present invention provides a method of processing landfill leachate by use of one or more membrane separation processes comprising the following steps: collecting landfill leachate in a receptacle suitable to hold said landfill leachate; treating said landfill leachate with one or more water soluble polymers; optionally mixing said water soluble polymers with said landfill leachate; passing said treated landfill leachate through a membrane, wherein said membrane is an ultrafiltration membrane or a microfiltration membrane; and optionally back-flushing said membrane to remove solids from the membrane surface.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 illustrates a general process scheme for processing landfill leachate, which includes a microfiltration membrane/ultrafiltration membrane, wherein the membrane is submerged in a tank, as well as an additional membrane for further processing of the permeate from said microfiltration membrane/ultrafiltration membrane.

Figure 2 illustrates a general process scheme for processing landfill leachate, which includes a mixing tank, clarifier/filter and a microfiltration membrane/ultrafiltration membrane, wherein the membrane is submerged in a tank, as well as an additional membrane for further processing of the permeate from said microfiltration membrane/ultrafiltration membrane.

Figure 3 illustrates a general process scheme for processing landfill leachate, which includes a mixing tank, clarifier/filter and a microfiltration membrane/ultrafiltration membrane, wherein the membrane is external to a feed tank that contains the landfill leachate, as well as an additional membrane for further processing of the permeate from said microfiltration membrane/ultrafiltration membrane.

DETAILED DESCRIPTION OF THE INVENTION

Definitions of Terms:

"UF" means ultrafiltration.

"MF" means microfiltration.

"Amphoteric polymer" means a polymer derived from both cationic monomers and anionic monomers, and, possibly, other non-ionic monomer(s). Amphoteric polymers can have a net positive or negative charge. The amphoteric polymer may also be derived from zwitterionic monomers and cationic or anionic monomers and possibly nonionic monomers. The amphoteric polymer is water soluble.

"Cationic polymer" means a polymer having an overall positive charge. The cationic polymers of this invention are prepared by polymerizing one or more cationic monomers, by copolymerizing one or more nonionic monomers and one or more cationic monomers, by condensing epichlorohydrin and a diamine or polyamine or condensing ethylenedichloride and ammonia or formaldehyde and an amine salt. The cationic polymer is water soluble.

"Zwitterionic polymer" means a polymer composed from zwitterionic monomers and, possibly, other non-ionic monomer(s). In zwitterionic polymers, all the polymer chains and segments within those chains are rigorously electrically neutral. Therefore, zwitterionic polymers represent a subset of amphoteric polymers, necessarily maintaining charge neutrality across all polymer chains and segments because both anionic charge and cationic charge are introduced within the same zwitterionic monomer. The zwitterionic polymer is water soluble.

Preferred Embodiments:

As stated above, the invention provides for a method of processing landfill leachate by use of a microfiltration membrane or an ultrafiltration membrane.

After the landfill leachate is collected and treated with one or more water-soluble polymers, the landfill leachate is passed through a membrane. In one embodiment, the membrane may be submerged in a tank. In another embodiment, the membrane is external to a feed tank that contains said landfill leachate.

In another embodiment, the landfill leachate that passes through the microfiltration membrane or ultrafiltration membrane may be further processed through one or more membranes. In yet a further embodiment, the additional membrane is either a reverse osmosis membrane or a nanofiltration membrane.

Various landfill leachate processing schemes would be apparent to one of ordinary skill in the art. In one embodiment, the collected landfill leachate may be passed through one or more filters or clarifiers prior to its passage through an ultrafiltration membrane or a microfiltration membrane. In a further embodiment, the filter is selected from the group consisting of: a sand filter; a multimedia filter; a cloth filter; a cartridge filter; and a bag filter.

The membranes utilized to process landfill leachate may have various types of physical and chemical parameters.

With respect to physical parameters, in one embodiment, the ultrafiltration membrane has a pore size in the range of 0.003 to 0.1 μm. In another embodiment, the microfiltration membrane has a pore size in the range of 0.1 to 0.4 μm. In another embodiment, the membrane has a hollow fiber configuration with outside-in or inside-out filtration mode. In another embodiment, the membrane has a flat sheet configuration. In another embodiment, the membrane has a tubular configuration. In another embodiment, the membrane has a multi-bore structure.

With respect to chemical parameters, in one embodiment, the membrane is polymeric. In another embodiment, the membrane is inorganic. In yet another embodiment, the membrane is stainless steel.

There are other physical and chemical membrane parameters that may be implemented for the claimed invention.

Various types and amounts of chemistries maybe utilized to treat the landfill leachate. In one embodiment, the landfill leachate collected from a landfill site is treated with one or more water-soluble polymers. Optionally, mixing of the landfill leachate with the added polymer is assisted by a mixing apparatus. There are many different types of mixing apparatuses that are known to those of ordinary skill in the art.

In another embodiment, these water-soluble polymers typically have a molecular weight of about 2,000 to about 10,000,000 daltons.

In another embodiment, the water-soluble polymers are selected from the group consisting of: amphoteric polymers; cationic polymers; and zwitterionic polymers.

In another embodiment, the amphoteric polymers are selected from the group consisting of: dimethylaminoethyl acrylate methyl chloride quaternary salt (DMAEA.MCQ) /acrylic acid copolymer, diallyldimethylammonium chloride/acrylic acid copolymer, dimethylaminoethyl acrylate methyl chloride salt/N,N-dimethyl-N-methacrylamidopropyl-N-(3-sulfopropyl)-ammonium betaine copolymer, acrylic acid/N,N-dimethyl-N-methacrylamidopropyl-N-(3-sulfopropyl)-ammonium betaine copolymer and DMAEA.MCQ/Acrylic acid/N,N-dimethyl-N-methacrylamidopropyl-N-(3-sulfopropyl)-ammonium betaine terpolymer.

In another embodiment the water soluble polymers have a molecular weight of about 2,000 to about 10,000,000 daltons. In yet a further embodiment, the water soluble polymers have a molecular weight from about 100,000 to about 2,000,000 daltons.

In another embodiment, the dosage of the amphoteric polymers is from about 1ppm to about 500 ppm of active solids

In another embodiment, the amphoteric polymers have a molecular weight of about 5,000 to about 2,000,000 daltons.

In another embodiment, the amphoteric polymers have a cationic charge equivalent to anionic charge equivalent ratio of about 4.0:6.0 to about 9.8:0.2.

In another embodiment, the cationic polymers are selected from the group consisting of: polydiallyldimethylammonium chloride ("poly DADMAC"); polyethyleneimine; polyepiamine; polyepiamine crosslinked with ammonia or ethylenediamine; condensation polymer of ethylenedichloride and ammonia; condensation polymer of triethanolamine and tall oil fatty acid; poly(dimethylaminoethylmethacrylate sulfuric acid salt); and poly(dimethylaminoethylacrylate methyl chloride quaternary salt).

In another embodiment, the cationic polymers are copolymers of acrylamide ("AcAm") and one or more cationic monomers selected from the group consisting of: diallyldimethylammonium chloride; dimethylaminoethylacrylate methyl chloride quaternary salt; dimethylaminoethylmethacrylate methyl chloride quaternary salt; and dimethylaminoethylacrylate benzyl chloride ("DMAEA.BCQ") quaternary salt.

In another embodiment, the dosage of cationic polymers is from about 0.05 ppm to about 400 ppm active solids.

In another embodiment, the cationic polymers have a cationic charge of at least about 5 mole percent.

In another embodiment, the cationic polymers have a cationic charge of 100 mole percent.

In another embodiment, the cationic polymers have a molecular weight of about 100,000 to about 10,000,000 daltons.

In another embodiment, the zwitterionic polymers are composed of about 1 to about 99 mole percent of N,N-dimethyl-N-methacrylamidopropyl-N-(3-sulfopropyl)-ammonium betaine and about 99 to about 1 mole percent of one or more nonionic monomers.

Three potential landfill leachate processing schemes are shown in Figure 1 through Figure 3.

Referring to Figure 1, landfill leachate is collected in a landfill leachate receptacle (1). The landfill leacheate then flows through a conduit, wherein said in-line addition (3) of one or more polymers occurs. The treated landfill leachate then flows into a membrane unit (6) that is submerged in a tank (11). Also, polymer (10) may be added to the tank (11) containing the submerged membrane. The submerged membrane may be an ultrafiltration membrane or a microfiltration membrane. Optionally, the subsequent permeate (8) then flows through an additional membrane (9) that may be either a reverse osmosis membrane or a nanofiltration membrane. Referring to Figure 2, landfill leachate is collected in a landfill leachate receptacle (1). The landfill leacheate then flows through a conduit, wherein said in-line addition (3) of one or more polymers occurs. The treated landfill leachate subsequently flows into a mixing tank (2), wherein it is mixed with a mixing apparatus (7), optionally additional polymer (4) is added to the mixing tank (2). The treated landfill leachate then travels through a pre-filter (5) or clarifier (5). The treated landfill leachate then flows through a conduit into a membrane unit (6) that is submerged in a tank (11). Optionally polymer (10) may be added to the tank (11) containing the submerged membrane. The submerged membrane may be an ultrafiltration membrane or a microfiltration membrane. Optionally, the subsequent permeate (8) then flows through an additional membrane (9) that maybe either a reverse osmosis membrane or a nanofiltration membrane.

Referring to Figure 3, landfill leachate is collected in a landfill leachate receptacle (1). The landfill leacheate then flows through a conduit, wherein said in-line addition (3) of one or more polymers occurs. The treated landfill leachate subsequently flows into a mixing tank (2), wherein it is mixed with a mixing apparatus (7), optionally additional polymer (4) is added to the mixing tank (2). The treated landfill leachate travels through a pre-filter (5) or clarifier (5). The treated landfill leachate then flows through a conduit into a membrane unit (6), either containing a microfiltration membrane or an ultrafiltration membrane. Optionally the subsequent permeate (8) then flows through an additional membrane (9) that may be either a reverse osmosis membrane or a nanofiltration membrane. The resulting permeate is collected for various purposes known to those of ordinary skill in the art.

In another embodiment, the membrane separation process is selected from the group consisting of: a cross-flow membrane separation process; a semi-dead-end flow membrane separation process; and a dead-end flow membrane separation process.

The following examples are not intended to limit the scope of the claimed invention.

EXAMPLES

Membrane flux was studied by performing water turbidity studies. Based on the well-established literature on water treatment using membranes, the significant decrease in water turbidity is expected to minimize the membrane fouling and allow the UF/MF operation at the same flux but for much longer run times between cleanings or even at higher flux. Turbidity was measured by a Hach Turbidimeter (Hach, Ames, IA), that is sensitive to 0.06 NTU (Nephelometric Turbidimetric Unit).

Example 1

Landfill leachate obtained from the eastern United States and contained in a 500 ml jar was treated by mixing with various dosages of Product A (Core shell copolymer of DMAEA.MCQ, and AcAm, cationic with 50% mole charge) and Product B (Copolymer of DMAEA.MCQ and AcAm, cationic with 50% mole charge), for about 2 minutes. The treated leachate was then settled for 10 minutes, and the turbidity of supernatant was measured. The colloidal material, which is the main cause of higher turbidity in leachate, was coagulated and flocculated by this method.

Table 1: Turbidity (NTU) of treated and settled landfill leachate from eastern US

Polymer Dosage (ppm) based upon actives	Product A	Product B
0 (Untreated)	359	
50	256	390
100	176	296
150	99	207
200	68	159
250	61	118
500		26.4

As shown in Table 1, there is over 83% and 90% reduction in turbidity after treatment with 250 ppm Product A and 500 ppm Product B, respectively. Therefore, if leachate is treated by, e.g. 200 ppm Product-B (concentration lower than that required for maximum turbidity removal, so that free polymer does not exist to come in contact with membrane surface), a dramatic improvement in membrane performance is expected.

Example 2

A leachate sample obtained from south-eastern US was studied in the same manner as in Example 1. The leachate sample was treated with Polymer A (same as in Example 1), Polymer C (Copolymer of DMAEA.MCQ, DMAEA.BCQ and AcAm, cationic with 35% mole charge), and Polymer D (Polydiallyldimethylammoniumchloride, cationic with 100% mole charge). Turbidity removal results are shown in Table 2.

Table 2: Turbidity (NTU) of treated and settled landfill leachate from south-eastern US

Polymer Dosage (ppm)	Product-A	Product–C	Product-D
0 (Untreated)	21		
10	2.58		
20		3.5	
100			3.3

As shown in Table 2, over 83% of turbidity was removed by treatment with Product A, C or D, although at different dosages.

CLAIMS

In the claims:

1. A method of processing landfill leachate by use of a membrane separation process comprising the following steps:
    a. collecting landfill leachate in a receptacle suitable to hold said landfill leachate;
    b. treating said landfill leachate with one or more water soluble polymers, wherein said water soluble polymers are selected from the group consisting of: amphoteric polymers; cationic polymers; zwitterionic polymers; and a combination thereof;
    c. optionally mixing said water soluble polymers with said landfill leachate;
    d. passing said treated landfill leachate through a membrane, wherein said membrane is an ultrafiltration membrane or a microfiltration membrane; and
    e. optionally back-flushing said membrane to remove solids from the membrane surface.
2. The method of claim 1, wherein a driving force for passage of said landfill leachate through said membrane is positive or negative pressure.
3. The method of claim 1, wherein said ultrafiltration membrane has a pore size in the range of 0.003 to 0.1 μm.
4. The method of claim 1, wherein said microfiltration membrane has a pore size in the range of 0.1 to 0.4 μm.
5. The method of claim 1, wherein said membrane is submerged in a tank.
6. The method of claim 1, wherein said membrane is external to a feed tank that contains said landfill leachate.
7. The method of claim 1, wherein the water soluble polymers have a molecular weight of about 2,000 to about 10,000,000 daltons.
8. The method of claim 1, wherein the amphoteric polymers are selected from the group consisting of: dimethylaminoethyl acrylate methyl chloride quaternary salt/acrylic acid copolymer, diallyldimethylammonium chloride/acrylic acid copolymer, dimethylaminoethyl acrylate methyl chloride salt/N,N-dimethyl-N-methacrylamidopropyl-N-(3-sulfopropyl)-ammonium betaine copolymer, acrylic acid/N,N-dimethyl-N-methacrylamidopropyl-N-(3-sulfopropyl)-ammonium betaine copolymer and DMAEA.MCQ/Acrylic acid/N,N-dimethyl-N-methacrylamidopropyl-N-(3-sulfopropyl)-ammonium betaine terpolymer.
9. The method of claim 1, wherein the dosage of the amphoteric polymers are from about 1ppm to about 500 ppm of active solids

10. The method of claim 1, wherein the amphoteric polymers have a molecular weight of about 5,000 to about 2,000,000 dalton.
11. The method of claim 1, wherein the amphoteric polymers have a cationic charge equivalent to an anionic charge equivalent ratio of about 4.0:6.0 to about 9.8:0.2.
12. The method of claim 1, wherein the cationic polymers are selected from the group consisting of: polydiallyldimethylammonium chloride; polyethyleneimine; polyepiamine; polyepiamine crosslinked with ammonia or ethylenediamine; condensation polymer of ethylenedichloride and ammonia; condensation polymer of triethanolamine an tall oil fatty acid; poly(dimethylaminoethylmethacrylate sulfuric acid salt); and poly(dimethylaminoethylacrylate methyl chloride quaternary salt).
13. The method of claim 1, wherein the cationic polymers are copolymers of acrylamide and one or more cationic monomers selected from the group consisting of: diallyldimethylammonium chloride, dimethylaminoethylacrylate methyl chloride quaternary salt, dimethylaminoethylmethacrylate methyl chloride quaternary salt and dimethylaminoethylacrylate benzyl chloride quaternary salt.
14. The method of claim 1, wherein the dosage of cationic polymers are from about 0.05 ppm to about 400 ppm active solids.
15. The method of claim 1, wherein the cationic polymers have a cationic charge of at least about 5 mole percent.
16. The method of claim 1, wherein the cationic polymers have a cationic charge of 100 mole percent.
17. The method of claim 1, wherein the cationic polymers have a molecular weight of about 500,000 to about 10,000,000 daltons.
18. The method of claim 1, wherein the zwitterionic polymers are composed of about 1 to about 99 mole percent of N,N-dimethyl-N-methacrylamidopropyl-N-(3-sulfopropyl)-ammonium betaine and about 99 to about 1 mole percent of one or more nonionic monomers.
19. The method of claim 1 further comprising passing said landfill leachate through a filter or a clarifier prior to said landfill leachate's passage through said membrane.
20. The method of claim 1 further comprising: passing a filtrate from said membrane through an additional membrane.

FIG. 1

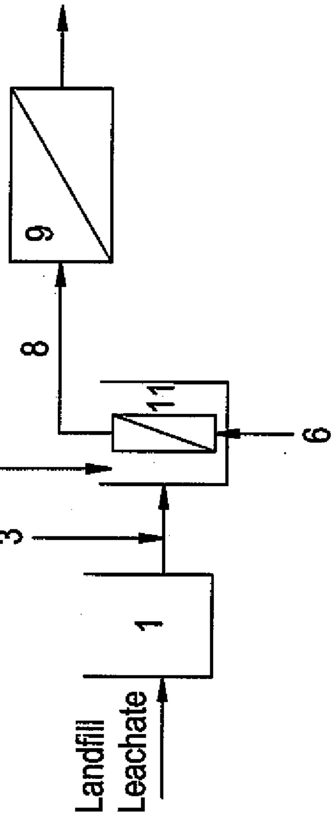


FIG. 2

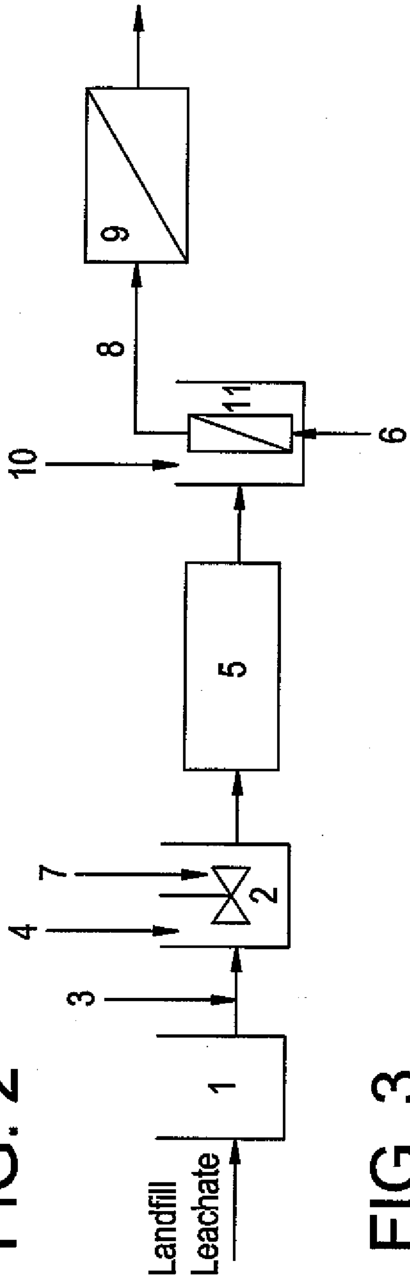
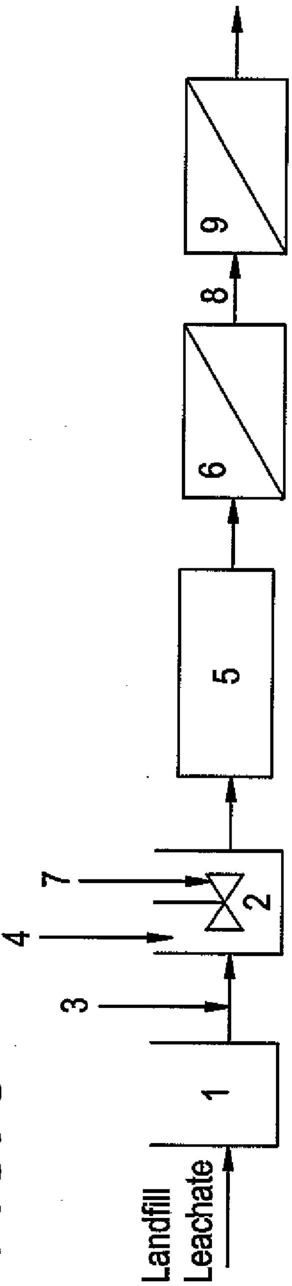


FIG. 3



INTERNATIONAL SEARCH REPORT

International application No.
PCT/US 07/69947

A. CLASSIFICATION OF SUBJECT MATTER
IPC(8) - B01D 61/00, C02F 1/44 (2007.01)
USPC - 210/650
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)
USPC - 210/650

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
USPC - 210/$

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
WEST (PGPB,USPT,USOC,EPAB,JPAB); Google
Search Terms: : membrane, separation, process, water, soluble, polymers, ultrafiltration, , microfiltration, zwitterionic, amphoteric, cationic

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	US 5,766,478 A (Smith et al.) 16 January 1998 (10.01.1998) fig. 1; col 2, 3, 10, 11, 12, 15, 33	1-2, 6 and 19-20 3-5 and 7-18
Y	US 2004/0168980 A1 (Musale et al.) 2 September 2004 (02.09.2004); para [0009]; [0079]; [0109]; [0114]-[0126]	3-5 and 7-18

☐ Further documents are listed in the continuation of Box C. ☐

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