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Has received an application for a patent for a new and useful invention. The title and description of the invention are enclosed. The requirements of law have been complied with, and it has been determined that a patent on the invention shall be granted under the law.

Therefore, this

United States Patent

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Michelle K. Lee

Director of the United States Patent and Trademark Office

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If the application for this patent was filed on or after June 8, 1995, the term of this patent begins on the date on which this patent issues and ends twenty years from the filing date of the application or, if the application contains a specific reference to an earlier filed application or applications under 35 U.S.C. 120, 121, 365(c), or 386(c), twenty years from the filing date of the earliest such application ("the twenty-year term"), subject to the payment of maintenance fees as provided by 35 U.S.C. 41(b), and any extension as provided by 35 U.S.C. 154(b) or 156 or any disclaimer under 35 U.S.C. 253.

If this application was filed prior to June 8, 1995, the term of this patent begins on the date on which this patent issues and ends on the later of seventeen years from the date of the grant of this patent or the twenty-year term set forth above for patents resulting from applications filed on or after June 8, 1995, subject to the payment of maintenance fees as provided by 35 U.S.C. 41(b) and any extension as provided by 35 U.S.C. 156 or any disclaimer under 35 U.S.C. 253.



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(12) **United States Patent**
Matsuda et al.

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(54) **ELECTROLYTE MEMBRANE, FUEL CELL,
AND ELECTROLYTE MEMBRANE
MANUFACTURING METHOD**

(58) **Field of Classification Search**
None
See application file for complete search history.

(75) Inventors: **Atsunori Matsuda**, Toyohashi (JP);
Song-Yul Oh, Toyohashi (JP); **Toshihiro
Yoshida**, Toyohashi (JP); **Go
Kawamura**, Toyohashi (JP); **Junichi
Hamagami**, Hachioji (JP); **Mototsugu
Sakai**, Kosai (JP)

(56) **References Cited**

U.S. PATENT DOCUMENTS

2003/0099874 A1 5/2003 Kim et al.
2005/0113547 A1* 5/2005 Li et al. 528/30

(Continued)

FOREIGN PATENT DOCUMENTS

EP 1624519 5/2003
EP 1309025 2/2006

(Continued)

OTHER PUBLICATIONS

Oh et al., Inorganic-organic composite electrolytes consisting of polybenzimidazole and Cs-substituted heteropoly acids and their application for medium temperature fuel cells, *J. Mater. Chem.* 2010 (20), pp. 6359-6366, Jun. 2010.*

(Continued)

Primary Examiner — Jonathan Crepeau
(74) *Attorney, Agent, or Firm* — Day Pitney LLP

(73) Assignee: **NATIONAL UNIVERSITY
CORPORATION TOYOHASHI
UNIVERSITY OF TECHNOLOGY**,
Toyohashi-Shi, Aichi (JP)

(*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 427 days.

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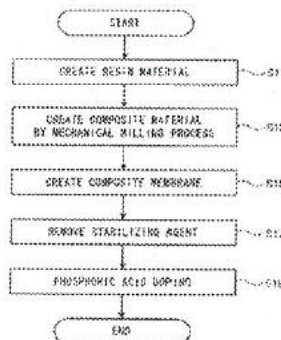
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CPC . **C08G 73/18** (2013.01); **C08J 5/22** (2013.01);
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(57) **ABSTRACT**

A composite membrane that can be used as an electrolyte membrane of a fuel cell is produced from a resin material that contains at least polybenzimidazole and from a composite material that is produced from of hydrogen sulfate and heteropoly acid. The composite membrane has a basic structure of an aromatic hydrocarbon that does not contain fluorine and has good efficiency, even at a low doping level, so it can be used for the electrolyte membrane of a medium temperature dry fuel cell. Phosphoric acid is doped into the composite membrane. Characteristics of the relationship between output current density and output voltage for an electrolyte membrane that is made from the created composite membrane (a specimen 4) are better than for a specimen 8, in which the amount of phosphoric acid doping is equivalent.

15 Claims, 20 Drawing Sheets



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(56)

References Cited

U.S. PATENT DOCUMENTS

2006/0027789 A1 2/2006 Ivanov et al.
 2006/0141316 A1 6/2006 Kang
 2007/0184323 A1* 8/2007 Lee et al. 429/33

FOREIGN PATENT DOCUMENTS

JP	2003-178777	6/2003
JP	2005-25943	1/2005
JP	2006-54179	2/2006
JP	2006-185919	7/2006
JP	2007-165006	6/2007
JP	2008-218299	9/2008

OTHER PUBLICATIONS

International Preliminary Report on Patentability and Written Opinion of the International Searching Authority (in the English and Japanese languages), for related international application No. PCT/JP2010/071131.

* cited by examiner