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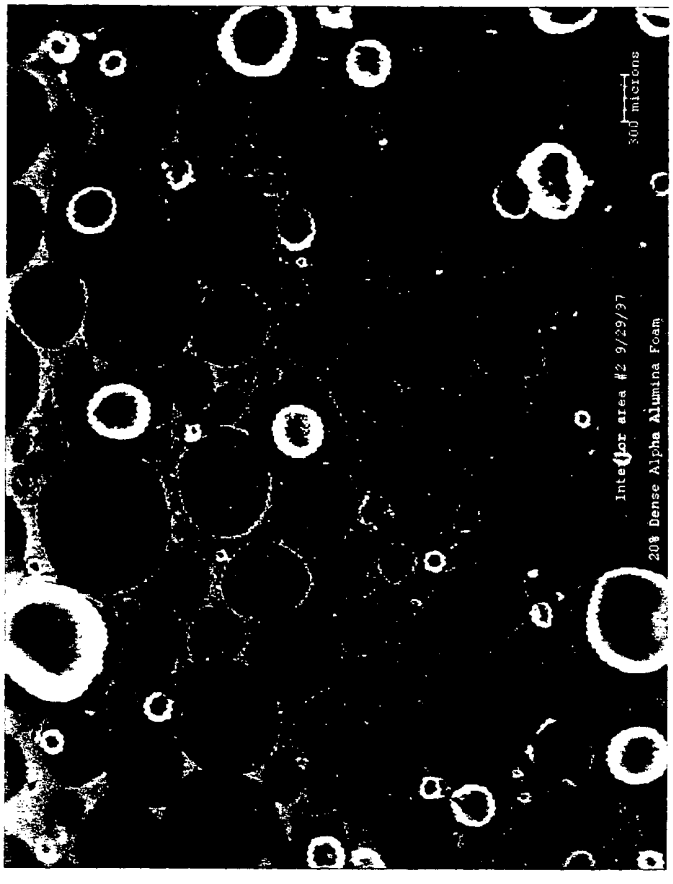
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(54) Title: CATALYTIC OXIDATION PROCESS



(57) Abstract: A process for the catalytic oxidation of a feed comprising contacting said zone, wherein: (a) said catalyst comprises at least one catalytically active metal selected from the group consisting of silver and the elements of Group VIII of the Periodic Table of the Elements supported on a porous ceramic carrier, (b) said catalyst is retained within said reaction zone in a fixed arrangement; and (c) said carrier has a distribution of total pores wherein: (1) about 70 percent of said total pores have a volume to surface area ratio (V/S) that is within about 20 percent of the mean V/S value for said total pores and no pores have a volume to surface area ratio that is greater than twice the mean V/S value for said total pores; and (2) for about 70 percent of said total pores, the pore-to-pore distance between neighboring pores is within about 25 percent of the mean pore-to-pore distance between neighboring pores; and (3) about 50 percent of said total pores have a coordination number that is within about 50 percent of the mean coordination number between neighboring pores and wherein less than about 15 percent of said total pores have a coordination number that is less than 3.

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ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZA, ZW.

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INTERNATIONAL SEARCH REPORT

Inte onal Application No
PCT/US 01/23610

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C01B3/38 C01B3/40 B01J35/04 B01J35/10

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)
IPC 7 C01B B01J

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 practical, search terms used)
API Data, EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	M. A. PENA ET AL.: "Partial oxidation of methane to sythesis gas over Ni-loaded ultrastable HY zeolite catalysts" STUDIES IN SURFACE SCIENCE AND CATALYSIS, vol. 107, 1997, pages 441-446, XP001058996 the whole document ---	1-20, 27-30
X	NAKAGAWA K ET AL: "Partial oxidation of methane to synthesis gas over supported iridium catalysts" APPLIED CATALYSIS A: GENERAL, ELSEVIER SCIENCE, AMSTERDAM, NL, vol. 169, no. 2, 11 May 1998 (1998-05-11), pages 281-290, XP004271437 ISSN: 0926-860X table 1 ---	1-19, 27-30

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

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"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

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Date of the actual completion of the international search	Date of mailing of the international search report
5 February 2002	21/02/2002

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Van der Poel, W

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Form PCT/ISA/210 (second sheet) (July 1992)

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
X	MATSUMOTO H ET AL: "REACTION MECHANISM OF METHANE OXIDATION TO SYNTHESIS GAS OVER AN ACTIVATED PDY ZEOLITE" JOURNAL OF THE CHEMICAL SOCIETY. FARADAY TRANSACTIONS, ROYAL SOCIETY OF CHEMISTRY, CAMBRIDGE, GB, vol. 90, no. 19, 7 October 1994 (1994-10-07), pages 3001-3006, XP000467711 ISSN: 0956-5000 page 3001 ---	1-19, 27-30
X	HICKMAN D A ET AL: "SYNTHESIS GAS FORMATION BY DIRECT OXIDATION OF METHANE OVER RH MONOLITHS" CATALYSIS LETTERS, BALTZER, SCIENTIFIC PUBL, BASEL, CH, vol. 17, no. 3/4, 1 February 1993 (1993-02-01), pages 223-237, XP000471634 ISSN: 1011-372X the whole document ---	1-20, 27-29
X	WO 99 37580 A (PIETERSE COEN WILLEM JOHANNES ;SCHADDENHORST DAVID (NL); HUISMAN H) 29 July 1999 (1999-07-29) examples ---	1-20, 27-29
X	US 5 741 440 A (INGRAM EUGENE MICHAEL ET AL) 21 April 1998 (1998-04-21) examples 1-3 ---	1
A	SU 1 780 831 A (GNI PI AZOTNOJ PROMY PRODUKTOV) 15 December 1992 (1992-12-15) abstract ---	1
A	US 5 705 448 A (SAMBROOK RODNEY MARTIN ET AL) 6 January 1998 (1998-01-06) cited in the application the whole document -----	1

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

Continuation of Box I.2

Claims Nos.: 1-30 (all partially)

Present claims 1-30 relate to a method defined by reference to a desirable characteristic or property, namely, the catalyst carrier has a distribution of pores wherein (1) about 70 percent of said total pores have a volume to surface area ratio (V/S) that is within about 20 percent of the mean V/S value for said total pores and no pores have volume to surface area ratio that is greater than twice the mean V/S value for said total pores; and (2) for about 70 percent of said total pores, the pore-to-pore distance netween neighboring pores is within about 25 percent of the mean pore-to-pore distance between neighboring pores; and (3) about 50 percent of said total pores have a coordination number that is within about 50 percent of the mean coordination number between neighboring pores and wherein less than about 15 percent of said total pores have coordination number that is less than 3.

The claims cover all methods using a carrier having this characteristic or property, whereas the application provides support within the meaning of Article 6 PCT and/or disclosure within the meaning of Article 5 PCT for only a very limited number of such methods. In the present case, the claims so lack support, and the application so lacks disclosure, that a meaningful search over the whole of the claimed scope is impossible. Independent of the above reasoning, the claims also lack clarity (Article 6 PCT). An attempt is made to define the method by reference to a result to be achieved. Again, this lack of clarity in the present case is such as to render a meaningful search over the whole of the claimed scope impossible.

Furthermore, present claims 1-30 relate to a method defined (inter alia) by reference to the following parameters:

P1: volume to surface ratio of the pores
P2: pore-to-pore distance
P3: coordination number of the pores

All three parameters furthermore have to fulfil complicated relationships, in which percentages of the total number of pores have to fulfil at least certain values of the parameters.

The use of these parameters in the present context is considered to lead to a lack of clarity within the meaning of Article 6 PCT. It is impossible to compare the parameters the applicant has chosen to employ with what is set out in the prior art. The lack of clarity is such as to render a meaningful complete search impossible.

Consequently, the search has been restricted to catalytic oxidation processes using catalysts carried on an inorganic oxide foam. The search also revealed catalytic oxidation processes using catalyst support materials that seem to fulfil the parametric requirements of claim 1. This does not mean that the search in this respect has been complete.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

The applicant's attention is drawn to the fact that claims, or parts of claims, relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure.

INTERNATIONAL SEARCH REPORT

Information on patent family members

Inte onal Application No
PCT/US 01/23610

Patent document cited in search report		Publication date	Patent family member(s)		Publication date
WO 9937580	A	29-07-1999	AU	734820 B2	21-06-2001
			AU	3250099 A	09-08-1999
			BR	9907110 A	24-10-2000
			CN	1288442 T	21-03-2001
			WO	9937580 A1	29-07-1999
			EP	1049645 A1	08-11-2000
			NO	20003700 A	19-07-2000
			PL	341869 A1	07-05-2001
			ZA	9900313 A	19-07-1999
US 5741440	A	21-04-1998	AU	7244196 A	17-04-1997
			WO	9711904 A1	03-04-1997
SU 1780831	A	15-12-1992	SU	1780831 A1	15-12-1992
US 5705448	A	06-01-1998	GB	2289466 A	22-11-1995
			AU	2415195 A	29-11-1995
			BR	9507674 A	19-08-1997
			CN	1148375 A ,B	23-04-1997
			DE	69508074 D1	08-04-1999
			DE	69508074 T2	15-07-1999
			EP	0759020 A1	26-02-1997
			WO	9530633 A1	16-11-1995
			JP	10500387 T	13-01-1998