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(81) **Designated States** *(unless otherwise indicated, for every kind of national protection available):* AE, AG, AL, AM, AT, AU, AZ, BA, **BB,** BG, **BH, BR,** BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, **DK,** DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, **HR,** HU, **ID, IL,** IN, **IS, JP,** KE, KG, KM, KN, **KP, KR,** KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PG, **PH,** PL, PT, **RO,** RS, RU, SC, SD, SE, SG, SK, SL, SM, SV, SY, **TJ,** TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW

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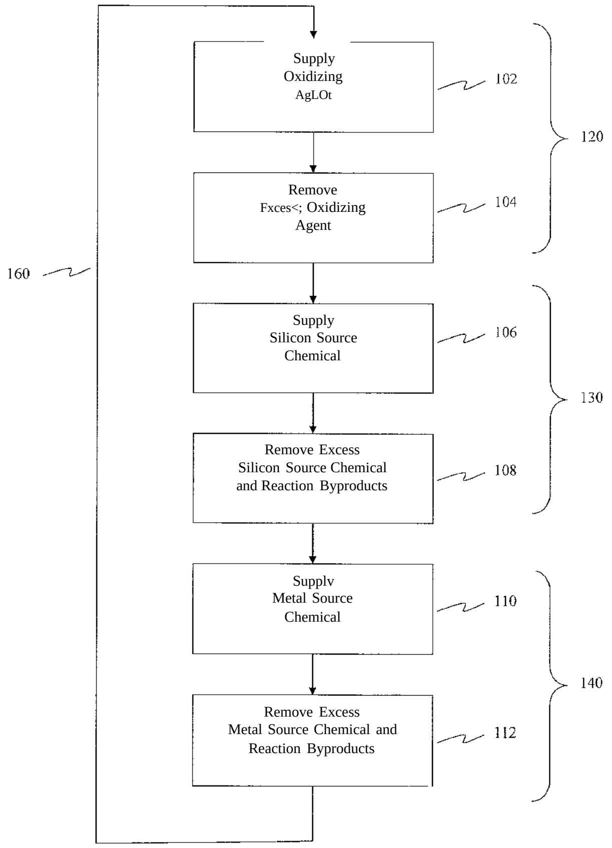
[Continued on next page]

(54) Title: ALD OF METAL SILICATE FILMS



(57) Abstract: Methods for forming metal silicate films are provided. The methods comprise contacting a substrate with alternating and sequential vapor phase pulses of a silicon source chemical, metal source chemical, and an oxidizing agent, wherein the metal source chemical is the next reactant provided after the silicon source chemical. Methods according to some embodiments can be used to form silicon-rich hafnium silicate and zirconium silicate films with substantially uniform film coverages on substrate surface.

PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:
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— *before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments*

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18 September 2008

INTERNATIONAL SEARCH REPORT

International application No
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A. CLASSIFICATION OF SUBJECT MATTER
INV. C23C16/40 C23C16/455

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)
C23C

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)

EPO-Internal , INSPEC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2005/271812 A1 (MYO NYI O [US] ET AL) 8 December 2005 (2005-12-08) page 3, paragraphs 32,33	1,3-15, 23
Y	page 7, paragraph 60 page 8, paragraph 62-69 page 9, paragraphs 74,76,77 page 10, paragraph 84 figures 4,5E -----	16,17
X	US 2005/271813 A1 (KHER SHREYAS [US] ET AL) 8 December 2005 (2005-12-08)	1-15,23
Y	page 3, paragraph 31-33 page 7, paragraphs 59,60 page 8, paragraph 64-68 page 9, paragraphs 75,76 page 10, paragraph 83 figures 4,5E -----	16,17
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☒ Further documents are listed in the continuation of Box C.

☒ See patent family 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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"T" later document published after the International filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"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.

"&" document member of the same patent family

Date of the actual completion of the international search 14 March 2008	Date of mailing of the International search report 04/07/2008
Name and mailing address of the ISA/ 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	Authorized officer Joffreau, P

Form PCT/ISA/210 (second sheet) (April 2005)

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2007/080342

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2005/113852 A (APPLIED MATERIALS INC [US] ; MYO NYI 00 [US] ; CHOI KENRIC [US] ; KHER SH) 1 December 2005 (2005-12-01) page 9, paragraph 28 - page 10, paragraph 30	1,3-15, 23
Y	page 21, paragraph 56 page 22, paragraph 58. page 23, paragraph 61 - page 25, paragraph 65 page 27, paragraph 70 page 28, paragraphs 72,73 page 31, paragraph 80 figures 4,5E -----	16,17
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Y	page 21, paragraph 56 page 22, paragraph 57 - page 25, paragraph 64 page 27, paragraph 71 - page 28, paragraph 19 page 31, paragraph 79 . figures 4,5E -----	16,17
X	KUKLI K ET AL: "Properties of Oxide Film Atomic Layer Deposited from -Tetraethoxy Silane, Hafnium Halides, and Water" JOURNAL .OF THE ELECTROCHEMICAL SOCIETY, vol . 151, **no. 5,** 17 March 2004 (2004-03-17) , pages F98-F104, XP002472772	1,2, 5-10,12
Y	page F99, left-hand column, paragraphs 3,4; tables I, II page FlOO, left-hand column , paragraphs 3,4; table III page FlOl ; table IV -----	13-15
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X	page 20, paragraph 54 page 22, paragraph 60 -----	23

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INTERNATIONAL SEARCH REPORT

International application No
PCT/US2007/080342

C(Continuation). **DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	KUKLI K ET AL: "Hafnium silicon oxide films prepared by atomic layer deposition" MATERIALS SCIENCE AND ENGINEERING B, ELSEVIER SEQUOIA, LAUSANNE, CH, vol. 109, no. 1-3, 15 June 2004 (2004-06-15), pages 2-5, XP004507161 ISSN: 0921-5107	1,2, 5-10,12
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X,P	DUEÑAS S ET AL: "Experimental investigation of the electrical properties of atomic layer deposited hafnium-rich silicate films on n-type silicon" JOURNAL OF AP-PLIED PHYSICS, AMERICAN INSTITUTE OF PHYSICS. NEW YORK, US, vol. 100, no. 9, 10 November 2006 (2006-11-10), pages 94107-1-94107-9, XP012090464 ISSN: 0021-8979 pages 94107-2, right-hand column, paragraph 3	1,2,5,6, 8-10
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INTERNATIONAL SEARCH REPORT

International application No
PCT/US2007/080342

C(Continuation). **DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X,P	DELABIE A ET AL: "Atomic layer deposition of hafnium silicate gate dielectric layers" JOURNAL OF VACUUM SCIENCE & TECHNOLOGY A (VACUUM, SURFACES, AND FILMS) AIP FOR AMERICAN VACUUM SOC. USA, vol. 25, no. 4, July 2007 (2007-07), pages 1302-1308, XP002472774 ISSN: 0734-2101 page 1302, right-hand column, last paragraph - page 1303, left-hand column, paragraph 1 page 1304, left-hand column, last paragraph - right-hand column, paragraph 1	1-10
X,P	WO 2007/043709 A (NIPPON ELECTRIC CO [JP]; NAKAGAWA TAKASHI [JP]) 19 April 2007 (2007-04-19) figures 18,19	1
E	WO 2008/011235 A (ASM INC [US]; WANG CHANG-GONG [US]; SHERO ERIC J [US]; WILK GLEN [US];) 24 January 2008 (2008-01-24) page 3, paragraphs 10,11 page 11, lines 20-25 page 13, paragraph 61	1,2,8-12

# INTERNATIONAL SEARCH REPORT	International application No. PCT/US2007/080342

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search 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.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

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

see additional sheet

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 fees, this Authority did not invite payment of additional fees.

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.:

see annex

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.

☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.

☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1-17,23

Atomic layer deposition method for forming a metal silicate film (e.g. hafnium silicate) characterized in that the metal source chemical is the next reactant provided after the silicon source chemical

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2. claims: 18-22

A silicon-rich metal silicate film having a silicon content greater than 65%

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/US2007/080342

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