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**NOTICE OF ENTITLEMENT**

We, FOSECO INTERNATIONAL LIMITED

of 285 Long Acre, Nechells, Birmingham, B7 5JR, GREAT BRITAIN being the applicant in respect of Application No.....42703/93..... state the following:

DAVID LLEWELLYN JONES  
MARTIN SEAN SWIFT and  
REINHARD STOETZEL

are the actual inventors.

The person nominated for the grant of the patent has entitlement from the actual inventors by virtue of their Contract Of Employment with David Llewellyn JONES and Martin Sean SWIFT and by virtue of an Assignment from Reinhard STOETZEL.

The person nominated for the grant of the patent is the applicant of the application listed in the declaration under Article 8 of the PCT.

The basic application listed in the declaration made under Article 8 of the PCT is the first application made in a Convention country in respect of the invention.

*W. J. Eyles*  
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W. J. EYLES - EXECUTIVE OFFICER



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**FILTER FOR LIGHT METALS**
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- (56) Prior Art Documents  
**EP 507463**  
**GB 2207913**  
**EP 388010**
- (57) Claim

1. A filter for the filtration of molten light metals comprising a reticular foam formed from a composition comprising graphite, wollastonite, silica and a glass characterised in that the filter consists of a crystalline phase comprising graphite and wollastonite dispersed in a substantially amorphous matrix of borosilicate glass.

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<b>(51) International Patent Classification<sup>5</sup> :</b> <b>B01D 39/20, C03C 14/00</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 93/25296</b>
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<b>(21) International Application Number:</b> PCT/GB93/00956 <b>(22) International Filing Date:</b> 10 May 1993 (10.05.93) <b>(30) Priority data:</b> 9211947.8 5 June 1992 (05.06.92) GB <b>(71) Applicant (for all designated States except US):</b> FOSECO INTERNATIONAL LIMITED [GB/GB]; 285 Long Acre, Nechells, Birmingham B7 5JR (GB). <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> JONES, David, Llevellyn [GB/GB]; 97 Bittell Road, Barnt Green, Birmingham B45 (GB). SWIFT, Martin, Sean [GB/GB]; 15 Thornley Close, Moseley, Birmingham B13 9UQ (GB). STOETZEL, Reinhard [DE/DE]; Nonnenettweide 20, D-4280 Borken (DE).	<b>(74) Agents:</b> MOORE, John, Hamilton et al.; Foseco International Limited, Group Patents Department, 285 Long Acre, Nechells, Birmingham B7 5JR (GB). <b>(81) Designated States:</b> AU, BR, CA, JP, KR, US, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). <b>Published</b> <i>With international search report.</i> <b>658760</b>	

**(54) Title:** FILTERS FOR LIGHT METALS

**(57) Abstract**

Filters for filtering molten light metals comprise a reticular foam formed from a composition comprising graphite, wollastonite, silica and a borosilicate glass and consist of a crystalline phase comprising graphite and wollastonite dispersed in a substantially amorphous matrix of borosilicate glass. The filters can readily be machined accurately from large pieces of the filter material and they do not pick up moisture from the atmosphere when stored. The borosilicate glass used to produce the filters preferably has a softening temperature ( $T_1$ ) in the range of 600 °C, more preferably 650 °C to 900 °C and a melting temperature ( $T_2$ ) in the range of 700 °C to 1100 °C, as measured according to the tests in German Standard DIN 51730.

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FILTERS FOR LIGHT METALS

This invention relates to filters for filtering light metals, by which term is to be understood aluminium, magnesium, alloys of these two metals, and alloys of either or both of these metals with other metals in which alloys, aluminium and/or magnesium predominate. In particular the invention relates to filters of relatively small size for use in dies for the gravity diecasting of aluminium or aluminium alloys.

It is well known to use open-cell ceramic foams to filter molten light metals. Such ceramic foam filters are generally made by impregnating an organic foam, usually polyurethane foam, with an aqueous slurry containing the material which is to form the filter and a binder, removing excess slurry, drying to remove water, and firing the dried impregnated foam to burn off the organic foam to form a ceramic foam.

Using the above method of manufacture individual filters are produced of the required shape and size, or alternatively a large piece of ceramic foam is produced and cut or machined to form individual filters. Both methods of manufacture suffer from disadvantages.

When individual filters of a given nominal size are produced directly from organic foam of a similar size to the filters their dimensional variation is such that problems arise in use, particularly in the gravity die casting of aluminium and its alloys, in which the filter is inserted in a recess in the die. As oversize filters cannot be used because they cannot

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be inserted in the recess it is common practice to compensate for the dimensional variations by deliberately making the filter slightly too small for the size of the recess. As a result small gaps remain between the edge of the filter and the wall of the recess, and because metal can penetrate through the gaps and bypass the filter, filtration efficiency is reduced. If the filter is loose, crushing of the filter can occur when the die is closed and this can lead to particles of ceramic material from the filter passing through the filter, and into the casting cavity of the die, thus resulting in inclusions in the casting.

When individual filters are produced from large pieces of ceramic foam by cutting or machining, manufacturing costs are high and productivity rates low, due to the nature of the materials from which the filters have hitherto been made. As the materials are brittle fracture of the foam tends to occur when the foam is cut or machined.

US Patent 3524548 describes a rigid, porous, filter medium for molten aluminium comprising particles of refractory material and a vitreous bonding agent of the type described in US Patent 2997402. The vitreous or glassy bonding agent consists essentially of boron oxide, calcium oxide and aluminium oxide and contains not more than 10% by weight of silica.

British Patent 1428437 describes a filter medium for molten aluminium made by shaping, drying and calcining at a temperature not lower than 1000°C a mixture comprising aggregate particles of fused alumina, sintered alumina, silicon carbide and/or

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silicon nitride, a combustible material, a caking material, water and an inorganic bonding material having a chemical composition of 10 to 50% by weight of silica and 5 to 20% by weight of boric oxide, at least 70% by weight of the inorganic bonding material being a non-vitreous substance. The inorganic bonding agent is prepared by mixing and pulverising at least 70% by weight of a natural or synthetic non-vitreous substance and not more than 30% by weight of a vitreous substance such as a commercially available glaze for porcelain.

EP 0507463A describes reticular foam filters which can be machined or cut to size from large pieces of filter material and which have a composition comprising graphite, wollastonite, silica, an alkali phosphate glass and monoaluminium orthophosphate.

It has now been found that the filters described in EP 0507463A can be improved if a borosilicate glass is used instead of the alkali phosphate glass and the monoaluminium orthophosphate.

According to the invention there is provided a filter for the filtration of molten light metals comprising a reticular foam formed from a composition comprising graphite, wollastonite, silica and a glass characterised in that the filter consists of a crystalline phase comprising graphite and wollastonite dispersed in a substantially amorphous matrix of borosilicate glass.

The filters of the invention may be made by impregnating a large piece of reticulated organic foam, such as polyurethane foam, with an aqueous slurry containing the graphite, wollastonite, silica,

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borosilicate glass, one or more binders which function during manufacture, and optionally other components, removing excess slurry (for example by squeezing the impregnated organic foam), drying and firing the impregnated organic foam, and then cutting or machining the resulting piece of inorganic foam to smaller pieces of the required shape and size. When the impregnated organic foam is fired the maximum firing temperature will usually be of the order of 800°C.

The aqueous slurry used for producing the filters of the invention will usually contain 5 - 25% by weight graphite, 1 - 10% by weight of wollastonite, 5 - 20% by weight silica and 35 - 50% by weight of borosilicate glass.

The wollastonite may be a powder or in the form of small fibres (for example of length 150 microns).

The silica used to produce the filters should be a very fine particle size silica such as a fumed silica, a colloidal silica, or the fine silica which is produced as by product in the production of silicon metal.

The borosilicate glass used to produce the filters preferably has a softening temperature ( $T_1$ ) in the range of 600°C, more preferably 650°C to 900°C and a melting temperature ( $T_2$ ) in the range of 700°C to 1100°C, as measured according to the tests in German Standard DIN 51730. Suitable borosilicate glasses include those sold by Bayer AG as ES71630, X0800215,

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X0800216 and X0800286, the chemical composition (% by weight) and T<sub>1</sub> and T<sub>2</sub> values for which are tabulated below:-

	ES71630	X0800215	X088216	X0800286
SiO <sub>2</sub>	39.08	37.7-42.3	32.0-34.2	53.66
B <sub>2</sub> O <sub>3</sub>	14.50	31.0-34.6	29.7-31.3	20.30
Na <sub>2</sub> O	9.52	12.8- 6.5	15.1-17.5	5.63
CaO	13.12	8.7- 9.8	-	5.72
MgO	9.63	2.0- 3.1	-	0.42
Al <sub>2</sub> O <sub>3</sub>	10.06	1.5- 2.1	14.0-14.9	9.85
K <sub>2</sub> O	0.37		4.4- 5.9	1.35
ZrO <sub>2</sub>	0.61			-
TiO <sub>2</sub>	0.20			-
Fe <sub>2</sub> O <sub>3</sub>				0.27
Other oxides	rest			rest
T <sub>1</sub>	712°C	641°C	621°C	790°C
T <sub>2</sub>	950°C	750°C	780°C	970°C

In the table the figures quoted for the composition of ES71630 and X0800286 are values determined by chemical analysis of a particular sample, and the figures for the other three borosilicate glasses are ranges quoted in the supplier's literature.

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The preferred borosilicate glasses are ES71630 and X0800286.

Alternatively, if desired the borosilicate may be formed in situ during the production of the filter from sodium silicate or calcium-sodium silicate and boric oxide added to the starting composition.

After firing the filters of the invention consist of a crystalline phase comprising principally graphite, wollastonite, and, if the borosilicate glass contains significant quantities of calcium oxide or magnesium oxide, diopside (a calcium-magnesium-silicate having the formula  $\text{Ca Mg} (\text{SiO}_3)_2$ ), and possibly cristobalite, dispersed in a substantially amorphous matrix of borosilicate glass.

In addition to the graphite the composition which is used to produce the filter of the invention may also contain amorphous carbon, such as carbon black, usually in an amount of up to about 5% by weight. The carbon black has the effect of improving the rheology of the slurry and loading of the strand of the reticular organic foam during impregnation of the foam with the slurry. In order to improve the appearance of the filter and to overcome the tendency of the graphite to produce a mottled appearance when the reticular foam filter material is fired, the composition also preferably contains a minor proportion, usually up to about 5% by weight, of silicon metal. It is believed that the silicon also reduces the tendency for some of the graphite to be lost due to oxidation during firing.

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The composition used to produce the filters may also contain a proportion, usually in an amount of up to about 10% by weight, of mica which because of its lamellar structure improves the strand loading of the organic reticular foam when the foam is impregnated with the slurry and also gives improved strength of the fired filter material.

Binders which can be used to produce green strength in impregnated large pieces of foam and a bond during drying and the initial stages of firing include a carbohydrate material such as dextrin, a polyacrylate, a polyvinylmethylether, a copolymer of an acrylic ester and styrol and a lignosulphonate such as an ammonium lignosulphonate. The amount of binder used will usually be up to 5% by weight based on the weight of the slurry. Monoaluminium orthophosphate may be used as a binder provided it has been neutralised to prevent reaction with the borosilicate glass, and provided it is only used in small quantities in order to avoid problems due to the finished filters being hygroscopic.

A material, such as glycerol, may be added to the slurry, usually in an amount of less than 1% by weight based on the weight of the slurry to prevent premature drying out of the slurry when it has been applied to the organic foam.

The slurry used to produce the filters may also contain additives to improve the applicational properties of the slurry during impregnation of the organic foam. Examples of such additives are

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rheological agents, for example organic thickening agents or titania, wetting agents and antifoaming agents.

When used the amount of rheological agent, wetting agent or antifoaming agent will usually be of the order of 0.1 - 2.0% by weight based on the weight of the slurry.

When the impregnated organic foam is fired it is desirable that the firing temperature does not exceed 850°C, unless special precautions are taken, otherwise excessive oxidation of the graphite, and amorphous carbon if present, will occur. The firing temperature will usually be in the range of 600°C - 800°C.

In order to alleviate any tendency for the edges to be friable and to improve strength to facilitate handling, in a preferred embodiment of the invention the exterior surfaces of large filters are coated with a composition comprising graphite and a binder and optionally silicon metal. If desired the composition used to coat the exterior surfaces of large bodies of filter material may be the same as or a more dilute version of the slurry which is used to produce the large bodies. After the large filters have been dried they are coated with the composition, for example by spraying the composition on to the dried filter, the filter is then heated to dry the coating, and finally fired. The fired large filter is then machined to produce small filters of a desired size.

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If desired dust produced when large bodies of the filter material are cut or machined to produce individual filters may be recycled by adding it to the slurry which is used subsequently to produce other bodies of filter material, or to coat the external surfaces of other large bodies.

The filters of the invention have all the desirable characteristics and properties of the filters described in EP 0507463A and they also have additional advantages compared to those filters.

When the filters described in EP 0507463A are stored in contact with the atmosphere they have a tendency to pick up moisture. The filters of the present invention do not suffer from this disadvantage.

The filters of the present invention also have superior strength, both at ambient and elevated temperatures compared to the filters described in EP 0507463A.

The following Examples will serve to illustrate the invention:-

EXAMPLE 1

Reticular foam filters were produced using an aqueous slurry of the following composition by weight:-

graphite	12.40%
fine silica (byproduct from silicon production)	10.94%
wollastonite fibres (length 150 microns)	7.29%
wollastonite powder	3.36%

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borosilicate glass (ES71630)	32.82%
amorphous carbon	1.82%
titania	0.73%
mica	3.36%
silicon	3.65%
dispersant	0.44%
thickening agent	0.15%
glycerol	0.44%
neutralised monoaluminium orthophosphate (60% w/w solution)	0.88%
polyvinylmethylether (50% W/W solution)	0.88%
water	20.84%

Pieces of reticulated polyurethane foam measuring 485 mm x 640 mm x 22 mm and having pores per cm were impregnated with the slurry, and excess slurry was removed by squeezing the foam.

After impregnation the pieces were coated by spinwheel spraying with a diluted version of the same slurry and the impregnated and coated foam was dried at 80 °C for 1 hour and at 120 °C for 30 minutes.

The coating procedure was repeated with the pieces inverted so as to coat what had been the underside in the first coating operation. After drying at 120 °C for 10 minutes the pieces were fired at 800 °C for 30 minutes, and finally cut to produce individual filters of nominal size 50 mm x 50 mm x 22 mm.

The suitability of the individual filters for filtering molten aluminium was then assessed by an impingement test after the filters had been weighed and measured using test apparatus of the type described and

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shown in Canadian Patent Application No. 2007450. The filters were inserted in a frame and approximately 25 kg of molten aluminium at a temperature of approximately 800°C were allowed to fall on to the filters from a ladle through a down pipe 350 cm high. The filters passed the test if they did not disintegrate under the action of the molten aluminium stream.

The results obtained are tabulated below:-

<u>Filter No.</u>	<u>Weight (g)</u>	<u>Dimensions (mm)</u>	<u>Pour Time</u>	<u>Pass/ (s)Fail</u>
1	19.6	50.3x49.4x21.8	22.7	Pass
2	18.2	49.8x49.8x21.8	17.6	Pass
3	13.4	49.5x49.5x21.6	18.9	Pass
4	18.3	50.4x49.5x21.8	20.1	Pass
5	13.4	49.3x49.6x21.9	16.0	Pass
6	18.0	49.3x49.6x22.0	19.4	Pass

#### EXAMPLE 2

Reticular foam filters were produced using an aqueous slurry of the following composition by weight:-

graphite	18.18%
fine silica (by product from silicon production)	9.73%
wollastonite fibres	12.45%
borosilicate glass (ES71630)	29.36%
amorphous carbon	1.64%
silicon	1.64%

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ammonium lignosulphonate (50% w/w solution)	0.95%
thickening agent (RHODOPOL)	0.14%
rheological agent (NORDFLOC AP 30E 1% w/w solution)	9.45%
acrylic ester/styrol copolymer (ACRONAL 290D)	2.82%
neutralised monoaluminium orthophosphate (60% w/w solution)	0.73%
water	12.91%

Individual filters of nominal size 50 mm x 50 mm x 22 mm were produced and tested using the procedures described in Example 1 except that in the impingement tests the temperature of the molten aluminium was approximately 850°C.

The results obtained are tabulated below:-

<u>Filter</u> <u>No.</u>	<u>Weight</u> (g)	<u>Dimensions</u> (mm)	<u>Pour Time</u> (s)	<u>Pass/</u> <u>Fail</u>
7	19.3	49.6x49.7x20.4	21.8	P
8	19.7	49.6x48.8x20.6	21.6	P
9	19.0	49.7x49.2x20.4	20.0	P

Chemical analysis of the filters by X-Ray Fluorescence and X-Ray Diffraction showed that the filters were composed of principally graphite, wollastonite and diopside dispersed in a borosilicate glass matrix.

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EXAMPLE 3

Reticular foam filters were produced using an aqueous slurry of the following composition by weight:-

graphite	17.63%
fine silica (by product from silicon production)	9.43%
wollastonite fibres	12.07%
bottle glass	26.67%
boric acid	1.45%
amorphous carbon	1.59%
silicon	1.59%
ammonium lignosulphonate (50% w/w solution)	0.92%
thickening agent (RHODOPOL)	0.14%
rheological agent (NORDFLOC NF 10E)	0.05%
acrylic ester/styrol copolymer (ACRONAL 290D)	2.73%
dextrin	1.75%
water	23.98%

The bottle glass contained, by weight, 70.31% silica, 11.84% sodium oxide, 11.73% calcium oxide, 3.45% alumina and 1.52% magnesium oxide.

Individual filters of nominal size 50 mm x 50 mm x 22 mm were produced and tested using the procedures described in Example 1 except that in the impingement tests the temperature of the molten aluminium was 850°C.

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The results obtained are tabulated below:-

<u>Filter</u> <u>No.</u>	<u>Weight</u> (g)	<u>Dimensions</u> (mm)	<u>Pour Time</u> (s)	<u>Pass/</u> <u>Fail</u>
10	15.2	50.2x50.2x21.9	13.2	P
11	15.5	50.1x50.5x21.9	14.2	P
12	16.3	50.4x50.2x22.0	11.8	P
13	15.6	50.5x50.3x22.4	12.8	P

Chemical analysis of the filters by X-Ray Fluorescence and X-Ray Diffraction showed the filters were composed of principally graphite, wollastonite, diopside and cristobalite dispersed in a borosilicate glass matrix.

A borosilicate glass made from the bottle glass and boric acid in the proportions used in the above composition had a softening temperature ( $T_1$ ) of 790°C and a melting temperature ( $T_2$ ) of 1050°C as measured according to the tests in German Standard DIN51730.

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CLAIMS

1. A filter for the filtration of molten light metals comprising a reticular foam formed from a composition comprising graphite, wollastonite, silica and a glass characterised in that the filter consists of a crystalline phase comprising graphite and wollastonite dispersed in a substantially amorphous matrix of borosilicate glass.

2. A filter according to Claim 1 characterised in that the borosilicate glass used to produce the filter has a softening temperature ( $T_1$ ) in the range 600°C to 900°C and a melting temperature ( $T_2$ ) in the range of 700°C to 1100°C as measured according to the tests in German Standard DIN51730.

3. A filter according to Claim 2 characterised in that the borosilicate glass has a softening temperature ( $T_1$ ) in the range 650°C to 900°C.

4. A filter according to any one of Claims 1 to 3 characterised in that the borosilicate glass is formed in situ during production of the filter from sodium silicate or calcium-sodium silicate and boron oxide.

5. A filter according to any one of Claims 1 to 4 characterised in that the crystalline phase contains diopside and/or cristobalite.

6. A filter according to any one of Claims 1 to 5 characterised in that the composition from which the filter is formed also contains amorphous carbon.

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7. A filter according to any one of Claims 1 to 6 characterised in that the composition from which the filter is formed also contains silicon.

8. A filter according to any one of Claims 1 to 7 characterised in that the composition from which the filter is formed also contains mica.

9. A filter according to any one of Claims 1 to 8 characterised in that the filter is formed from an aqueous slurry containing 5 - 25% by weight graphite, 1 - 10% by weight wollastonite, 5 - 20% by weight silica and 35 - 50% by weight of borosilicate glass.

## INTERNATIONAL SEARCH REPORT

PCT/GB 93/00956

International Application No

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (if several classification symbols apply, indicate all) <sup>6</sup>		
According to International Patent Classification (IPC) or to both National Classification and IPC		
Int.Cl. 5 B01D39/20; C03C14/00		
<b>II. FIELDS SEARCHED</b>		
Minimum Documentation Searched <sup>7</sup>		
Classification System	Classification Symbols	
Int.Cl. 5	B01D ; C03C ; C04B ; C22B	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched <sup>8</sup>		
<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT<sup>9</sup></b>		
Category <sup>10</sup>	Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>
P,A	EP,A,0 507 463 (FOSECO INTERNATIONAL LIMITED) 7 October 1992 cited in the application see the whole document	1,4,9
A	--- CHEMICAL ABSTRACTS, vol. 109, no. 10, 1988, Columbus, Ohio, US; abstract no. 78487c, 'Porous molded body from glass' see abstract & PATENT ABSTRACTS OF JAPAN vol. 12, no. 333 (C-526)8 September 1988 & JP,A,63 095 141 ( KUBOTA LTD. ) 26 April 1988 see abstract --- -/--	1-3,9
<p><sup>10</sup> Special categories of cited documents:</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"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</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"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.</p> <p>"A" document member of the same patent family</p>		
<b>IV. CERTIFICATION</b>		
Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report	
03 AUGUST 1993	11. 08. 93	
International Searching Authority	Signature of Authorized Officer	
EUROPEAN PATENT OFFICE	STEVNSBORG N.	

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category °	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No.
A	GB,A,2 207 913 (CENTRAL GLASS COMPANY LIMITED) 15 February 1989 see abstract see page 6, line 3 - page 7, line 15 see page 14, line 12 - page 15, line 1 see page 28, line 12 - line 22 see tables 4-1,4-2 ---	1-3,8
A	EP,A,0 388 010 (FOSECO INTERNATIONAL LIMITED) 19 September 1990 see the whole document -----	1-4,6,9

**ANNEX TO THE INTERNATIONAL SEARCH REPORT  
ON INTERNATIONAL PATENT APPLICATION NO.**

GB 9300956  
SA 73689

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The members are as contained in the European Patent Office EDP file on The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information. 03/08/93

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP-A-0507463	07-10-92	US-A- 5169542	08-12-92
GB-A-2207913	15-02-89	JP-B- 4049499	11-08-92
		JP-A- 63319232	27-12-88
		JP-A- 1087532	31-03-89
		US-A- 4992321	12-02-91
EP-A-0388010	19-09-90	AU-A- 5003790	30-08-90
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