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D E C I S I O N
of 21 September 1999

Case Number: T 0835/95 - 3.3.5

Application Number: 88300228.9

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Title of invention:
Glass microbubbles

Patentee:
Minnesota Mining and Manufacturing Company

Opponent:
Asahi Glass Company, Ltd.

Headword:
Microbubbles/3M

Relevant legal provisions:
EPC Art. 84, 54, 56

Keyword:
"Novelty (yes), no implicit disclosure"
"Inventive step (yes) warning in the prior art"

Decisions cited:
T 0990/96

Catchword:
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Boards of Appeal

Chambres de recours

Case Number: T 0835/95 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 21 September 1999

Appellant: Asahi Glass Company, Ltd.
(Opponent) 1-2 Marunouchi 2-chome
Chiyoda-ku, Tokyo 100
JAPON

Representative: Wächtershäuser, Günter, Prof. Dr.
Patentanwalt
Tal 29
80331 München
ALLEMAGNE

Respondent: Minnesota Mining and Manufacturing Company
(Proprietor of the patent) 3M Center
P.O. Box 33427
St. Paul
Minnesota 55133-3427
ETATS-UNIS D'AMERIQUE

Representative: Vossius & Partner
Postfach 86 07 67
81634 München
ALLEMAGNE

Decision under appeal: Interlocutory decision of the Opposition Division
of the European Patent Office posted 12 August
1995 concerning maintenance of European patent
No. 0 276 921 in amended form.

Composition of the Board:

Chairman: R. K. Spangenberg
Members: M. M. Eberhard

Summary of Facts and Submissions

I. European patent No. 0 276 921 based on application No. 88 300 228.9 was granted on the basis of seven claims. The appellant (opponent) filed a notice of opposition requesting revocation of the patent on the grounds of lack of novelty and lack of inventive step. During the opposition procedure, the parties relied *inter alia* on the following documents:

D3: US-A-4 391 646

D5: UCRL-51609, "Fabrication of the glass microballon laser target", P.C. Souers & al., 12 July 1974

D7: JP-A-49-37565, 1974, and English translation thereof

D12: Technical report from H. J. Marshall

D13: G. W. Morey, "The Properties of Glass", pages 42 to 54, 57 to 68, 75, 77, (1954)

D21: Affidavit of H. J. Marshall and Exhibits 1 to 13

D22: Handbook of Glass Properties, N. P. Bansal, H. R. Doremus, 1986, pages 31, 40 to 41, 49 to 51, 66 to 67, 126 to 128, 147

D23: "Glas", H. Thiene, 1939, pages 818, 823 to 824, 831, 850, 853 to 854, 856 to 858, 860, 862 to 863, 905, 938, 990.

II. The opposition division decided to maintain the patent

in an amended form. It considered that claims 1 to 6 according to the first of the five auxiliary requests filed on 12 May 1995 met the requirements of the EPC. Claim 1 of this request reads as follows:

"1. Microbubbles of glass having an alkaline earth metal oxide:alkali metal oxide weight ratio in the range of 1.2:1-3.0:1, at least 90% of the glass weight consisting of 70-80% SiO₂, 8-15% RO, 3-8% R₂O, 0.125-1.50% SO₃ and 2-6% B₂O₃, wherein R is at least one metal having the indicated valence, RO being an alkaline earth metal oxide and R₂O an alkali metal oxide, provided that the optional balancing components of the said glass weight shall not comprise the said oxides already specifically herein mentioned."

The opposition division held that the glass microbubbles B18A, B22A and B35D having the compositions reported in D5 were part of the state of the art. The claimed microbubbles were new over those of D5 in that they contained a specified amount of SO₃ in the glass. It further took the view that the skilled person had to overcome a prejudice when preparing glass microbubbles from glasses with a high RO/R₂O ratio. The respondent's experiments in the letter of 28 October 1994 underlined the difficulties the skilled person would have encountered in producing microbubbles of the composition reported in D5 in acceptable yields. Neither D5 nor any other document taught any particular benefits of using high RO/R₂O glass formulations so that the skilled person would not have been encouraged to pursue this approach.

III. The appellant lodged an appeal against this decision.

Oral proceedings were held on 21 September 1999. At the appeal stage the appellant referred to additional documents in order to show *inter alia* that the volatilisation of alkalis, boron oxide and sulfur from glass melts was a well-known phenomenon. The following documents were in particular relied on in connection with novelty and inventive step:

D24: US-A-3 365 315

D34: Kirk-Othmer Encyclopedia of chemical technology, vol. 4, 1964, pages 788 to 789

D35: Ullmanns Encyklopädie der technischen Chemie , volume 8, 1957, pages 135, 147, 163 to 165.

IV. The appellant's arguments concerning claim 1 of the first auxiliary request upheld by the opposition division can be summarised as follows:

It was ambiguous whether the SO₃ content indicated in claim 1 was that of the glass only or of the complete bubbles in view of some statements in the description which seemed to be inconsistent with claim 1.

D5 was relied on not as evidence to prove a prior use but as a regular written prior art document.

Microbubbles according to claim 1 lacked novelty over the disclosure of D5. The skilled person would have deduced directly from D5 and D24 that sulphur had been used as the blowing agent for preparing the microbubbles of D5 since SO₂ and O₂ were inside the bubbles. The SO₃ content stated in claim 1 was implicitly disclosed in D5 in that a SO₃ content falling

within the claimed range was inevitably obtained by following the instructions of D5 and D24, as shown by the appellant's test report dated 8 December 1995. Although the SO₃ content of the bubbles was not indicated in D5, it was in fact a different way of expressing or measuring the bubble density. The latter could be derived from the data given in D5 about glass density, diameter distribution and wall thickness. The density could also be obtained by asking the microbubble manufacturer. The test report, which was performed so as to achieve a density of 0.34 g/cm³, led to a SO₃ content falling within the claimed range. In a second line of argument regarding the lack of novelty, the appellant contended that a range of 0.05-1.2 g/cm³ for the bubble density was derivable from D5/D24 and that the selected density range of 0.08-0.8 g/cm³ represented only the range the skilled person would in practice have selected. Furthermore, the claimed glass bubbles were in fact bubbles of the same composition as those of D5 but containing SO₃ as impurities since the sulphur had no effect on the bubbles properties. According to decision T 990/96 impurities could not render a product novel. The disclosure of D5/D24 was an "enabling disclosure" since it was well-known at the date of publication of D5 that volatilisation of the volatile components occurred in particular with particles of high surfaces and that water quenching avoided devitrification.

D3 represented the closest prior art. The problem to be solved relative to D3 was to increase the water resistance of the bubbles. D3 taught that higher RO/R₂O ratios increased the chemical durability and were beneficial to the strength of the microbubbles.

Furthermore, it was well-known to increase the CaO content of a silicate glass in order to improve its water resistance, as shown by D35. Therefore, the claimed microbubbles lacked an inventive step in view of the teaching of D3 alone. Furthermore, in view of D5 which disclosed glass microbubbles made of compositions having a $R_0/R_2O > 1.2$ and also dealt with the problem of water resistance, it was obvious to the skilled person to arrive at the claimed subject-matter by combining the teaching of D3 and D5. As D5 disclosed three examples of glass microbubbles with a ratio $R_0/R_2O > 1.2$, this was a clear refutation of the alleged prejudice. Taking into account the composition shift known at the priority date, the R_0/R_2O ratios of the glass bubbles exemplified in D7 lay within the claimed range. The opinion expressed in D7 regarding the CaO and alkali oxide contents was far from being a prejudice. If D5 were regarded as the closest prior art, the problem to be solved would have been to modify the density of the microbubbles. This problem found a simple solution by combining the compositions of D5 with the process disclosed in D3.

- V. The respondent considered that the appellant's objection regarding the ambiguity of the SO_3 content stated in claim 1 was not relevant and did not want to take a position on this point. He argued *inter alia* that D5 did not destroy the novelty of the claimed microbubbles since the skilled person was unable to reproduce the glass microbubbles of D5, without undue burden, at the time of publication of D5, even in view of the teaching of D24. Furthermore, D5 did not implicitly disclose the use of SO_2 as the blowing agent, let alone the claimed amount of SO_3 . The appellant's

tests had been performed with the hindsight knowledge of a composition shift and a water quenching to form the frit and, thus, did not prove the alleged lack of novelty. The appellant's arguments concerning the relation between the density and the SO₃ content were not correct, since other parameters such as, for example, the operating conditions during the bubble formation, also had an influence on the bubble density.

The problem to be solved with respect to D3 as the closest prior art was to provide glass microbubbles which exhibited an improved water resistance while retaining a high compressive strength, and which could be produced over a broad density range in a convenient way and in high yields. Up to the date of the patent in suit, it was a wide spread belief that glass compositions with a high RO/R₂O ratio had a tendency to devitrify and thus should be avoided, as evidenced for example by D13. D5 did not indicate any beneficial properties of the bubbles. D7 warned about the risk of devitrification when using a high RO/R₂O ratio and taught that devitrification resulted in a decreased strength. If the skilled person had increased the RO content or the RO/R₂O ratio of the glass bubbles of D3 in order to improve the water insolubility, he would not have gone beyond the limit considered feasible. Furthermore, neither D3 nor the other prior art documents taught that increasing the RO/R₂O ratio would have allowed the incorporation of higher levels of SO₃ into the glass frit and, thus, the production of microbubbles with low densities in high yields and in an easy way. D5 could not be taken as the starting point for assessing inventive step since it was neither an "enabling disclosure" nor could it be

considered as the closest prior art.

- VI. The appellant requested that the decision under appeal be set aside and that the patent be revoked. The respondent requested that the appeal be dismissed. As an auxiliary request, the respondent requested that the case be remitted to the opposition division for further prosecution. As further auxiliary requests, the respondent requested that the patent be maintained on the basis of any of the auxiliary requests 2 to 5 filed on 12 May 1995, with further amendments to the fourth auxiliary request indicated in the respondent's letter dated 21 April 1999.

Reasons for the Decision

1. The appeal is admissible.
2. Claims 1 to 6 of the main request, which are amended claims 1 to 6 according to the first auxiliary request submitted on 12 May 1995 and upheld by the opposition division, meet the requirements of Article 123(2) and (3) EPC. In particular, the sulphur content of the glass (expressed as SO₃) incorporated into claim 1 is disclosed in the application as filed, on page 3, lines 15 to 20, and in original claim 7. By the introduction of this additional feature into granted claim 1, the scope of protection has been restricted.
3. At the oral proceedings the appellant pointed out for the first time that although the SO₃ content was, according to the wording of claim 1, that of the glass,

there were doubts as to whether this content related to the glass itself or to the glass microbubbles. In the appellant's view the statements on page 14, lines 2 to 4, and page 18, lines 3 to 6, of the original description regarding the method of measurement of the sulphur content as well as Tables II and IV on pages 8 and 12 would appear to be inconsistent with the SO₃ content of claim 1 being that of the glass. The said passages correspond respectively to page 9, lines 18 to 19; page 11, lines 17 to 18, Table II and Table IV of the patent in suit. Taking into account that the sulphur content of the glass microbubbles is higher than the sulphur content of the glass because of the additional sulphur present as gas (SO₂) inside the bubbles, this issue has to be examined first to determine whether or not amended claim 1 meets the requirements of Article 84 EPC and on which basis novelty and inventive step should be assessed. The respondent did not want to comment on the appellant's objection. In the board's view the wording of claim 1 itself is not ambiguous and it is clear that the SO₃ content is that of the glass. This was also not contested by the appellant. The SO₃ content in the glass as indicated in claim 1 is also in agreement with the statement on page 2, lines 51 to 54, of the patent in suit and claim 6 as granted so that claim 1 is supported by the description. The two passages referred to by the appellant disclose that the amount of sulphur (expressed as SO₃) was determined by iodine titration, measuring SO₂ evolved from a bubble **feed sample** when heated to 1500°C. Both passages, thus, relate to the determination of the amount of sulphur in the bubble **feed sample** and **not in the final bubbles**. As these statements concern the bubble feed, they are not

inconsistent with the fact that claim 1 gives the sulphur content of only the glass of the final microbubbles. The SO₃ contents reported in Table II are those of the glass bubble feeds for Examples 1, 3, 5, 7 and 8, and Table IV gives the analysis of the corresponding **glass bubbles**. However, as Table IV does not indicate the sulphur content of the glass itself but of the glass bubbles, the board cannot conclude that there is an inconsistency between the data of Table IV and the sulphur content of the glass stated in claim 1. The board observes in this respect that the patent in suit does not disclose which proportion of the total sulphur present in the final bubbles remains in the glass walls or as gas (SO₂) in the space inside the bubbles. No reliable assumptions can be made in this respect on the basis of the values given in Example 3 of D3 taking into account the differences in the glass compositions and in the operating conditions used during bubble formation. The appellant himself has provided no evidence that the SO₃ content of the glass in the said examples of Table IV lies outside the claimed range, nor has he raised objections against claim 6 as granted during the opposition procedure. Although the upper limit of 1.5 wt% stated in claim 1 might appear to be surprisingly high, there is no indication in the description from which it could be concluded that a mistake has occurred and that this value is not obtainable or relates to the whole microbubbles and not to the glass. For the preceding reasons the board considers that there is no inconsistency between the description and claim 1. Therefore, claim 1 meets the requirements of Article 84 EPC and it is considered that the sulphur content stated in claim 1 is actually that of the glass as

clearly indicated therein.

4. Novelty was disputed over the disclosure of D5. The board assumes to the appellant's benefit that D5 is an "enabling disclosure", ie that the skilled person would have been able to reproduce the microbubbles having the composition stated therein at the date of D5.

4.1 D5 discloses the chemical analysis of glass microballoons manufactured by the respondent. The three samples of microballoons designated B18A, B22A and B35D have SiO_2 , RO, R_2O and B_2O_3 contents as well as RO/ R_2O ratios lying within the claimed ranges (see D5, pages 7 and 8; D12, page 2, table 1). However, D5 is silent as to whether or not the glass of the microballoons contains sulphur. According to page 3, these microballoons are made by permeating gas into solid glass particles and reference is made to D24 for the manufacturing process. D5 further discloses that the finished microballoons contain 0.034 MPa SO_2 and O_2 (see page 3, right-hand column, third paragraph). The appellant's argument that the manufacture of the glass microbubbles of D5 by the process of D24 inevitably leads to microbubbles having a SO_3 content lying within the claimed range as shown by the test report dated 8 December 1995, is not convincing for the following reasons:

D24 discloses two methods for preparing the glass microbubbles, ie the first method or "permeation" method which is described in column 5, lines 21 to 32, and used in Examples 1 to 4 of D24, and the second method in which the compound which liberates gases during the reheating step is incorporated directly into

the glass of the particles (see column 5, lines 33 to 41). Although D5 indicates that the microbubbles were made by the "permeation" method of D24, the appellant did not use this method but the second method disclosed in D24. Therefore, he did not follow the instructions given in D5 for manufacturing the microbubbles. The appellant has provided no evidence that the second method would lead to similar results regarding the sulphur content of the glass of the resulting microbubbles. Furthermore, in the test report the molten glass was quenched into water to prepare the glass frit although this step, which is essential in the case of glasses with a high RO/R₂O ratio, is not disclosed in D24. It is observed that another method of preparing a glass frit, which avoids a drying step, was also well-known at the date of D5 (see D34, page 788). Neither the sulphur content of the microballoons nor that of the frit are given in D5; therefore, the amount of sulfate to be incorporated into the raw material batch could not be calculated back from the said content for reproducing the microbubbles. At the oral proceedings the appellant's representative explained that the SO₃ content of the microbubbles was in fact another way to express the bubble density, and that, accordingly, the appellant had determined by trial and error experiments the sulphate amount of the raw material which led to a density of 0.34 g/cm³. He had thus obtained the SO₃ content of 0.30 wt% stated in Tables 2 and 3 of the test report for the final bubbles. The board observes that although the density can be controlled by the content of blowing agent in the bubble feed (glass frit), this content is not the only parameter influencing the density. As pointed out by the respondent, the operating conditions used during

the reheating step to form the bubbles, in particular the temperature of the flame, the feed rate or the residence time in the flame also have an influence on the bubble density. The appellant's allegation that the influence of these parameters was negligible was not supported by evidence and was contested by the respondent. Furthermore, the density of 0.34 g/cm^3 selected by the appellant is not disclosed in D5. The appellant's arguments that the bubble density can be derived therefrom since D5 gives the glass density, the size distribution of the bubbles and the wall thickness were not accompanied by any concrete calculation showing that the data stated in D5 actually lead to the value of 0.34 g/cm^3 nor did the appellant refer to passages of D5 indicating the size distribution and wall thickness. D5 discloses a bubble diameter distribution of the as-received microballoons on page 16, Figure 5. Table 11 on page 19 discloses the mean wall thickness determined on sectioned microballoons of two sieve cuts (45-60 μm and 70-75 μm) of the B35D microballoons. The average wall thickness determined by helium permeation is also indicated on page 23 for B35D microballoons but it concerns only the narrow sieve cut 70-75 μm . As shown by Figure 5 the sample B35D contains substantial amounts of microballoons having diameters between 10-45 μm and 60-70 μm for which the wall thickness is not disclosed. Therefore, the data given in D5 are incomplete. In the absence of further information from the appellant as to which assumptions and calculations were made to arrive at the density value of 0.34 g/cm^3 on the basis of the data given in D5, the board cannot accept that this density is directly and unambiguously derivable from D5. The appellant further argued that the skilled

person would have obtained the density of the B35D sample by simply asking the manufacturer and would have been told the density of 0.30-0.34 indicated in Exhibit 13 of D21. These arguments are not convincing since the manufacturer of the glass microbubbles B35D analysed in D5 is the respondent, and the latter argued in the opposition and appeal proceedings that he had not manufactured B35D, B18A and B22A microballoons having the compositions stated in D5 before the date of D5 and that the analysis was not correct. The appellant also referred to the SO₃ value of 0.13 wt% indicated by the appellant in D12, page 3, as a further piece of evidence for the SO₃ content of the microbubbles. However, the estimated value of 0.13% does not concern the microbubbles B35D of D5 but the sample B18A which contains a surprisingly high amount of manganese oxide and about 5 wt% of unknown components. It is not clear to the board how this value might confirm the SO₃ content of 0.30% obtained for the B35D microbubbles in the appellant's test report. Regarding the said SO₃ content of 0.30%, the question also arises whether or not the sulphur content of the glass itself lies within the claimed range.

For the reasons given above the board considers that the appellant's test report does not prove that the skilled person would inevitably arrive at microbubbles having the sulphur content as defined in claim 1 by following the teaching of D5/D24.

- 4.2 The second line of argument presented by the appellant is based on the assumption that the sulphur content of the microbubbles is a measure of their density and that the claimed subject-matter consists in fact of a

selection of a density range of 0.08-0.8 g/cm³ within the broader range of density 0.05-0.5 g/cm³ disclosed in D5/D24. These arguments cannot be accepted for the following reasons:

The claimed subject-matter cannot be considered as a selection of a density range disclosed in D5 since D5 discloses the composition of **individual** samples of microbubbles and mentions neither their density nor their sulphur content. D5 refers to D24 only for illustrating the "permeation" method used to manufacture the bubbles and not for the SO₃ content or the average density of the microbubbles. Although D24 teaches a range of 0.05-1.2 g/cm³ for the average density of the microbubbles (see claim 3), this density range concerns glass microbubbles having the compositions indicated in claim 3 of D24. With respect to this teaching the skilled person would have had to select not only a narrower range of density (which by the way is already disclosed in D24, column 3, line 40) but also the claimed composition itself, ie the appropriate ranges for the different oxides of the glass composition. Furthermore, claim 1 does not define a density range for the microbubbles but the sulphur content of the glass and D24 discloses no range for this parameter.

- 4.3 The appellant further argued at the oral proceedings that the claimed microbubbles lacked novelty because they were in fact the same microbubbles as those of D5 but with impurities, and according to decision T 990/96 (OJ EPO 1998, 489) the degree of purity of a product could not bring novelty. In the appellant's view, as the sulphur contained in the glass of the claimed

microbubbles has no effect, it should be considered as an impurity. This argument cannot be accepted by the board since it is in contradiction with the whole teaching of the patent in suit. It is evident from the patent in suit that the sulphur contained in the glass of the microbubbles is not an impurity and that, on the contrary, sulphur is **deliberately** introduced into the raw material batch and serves as a **blowing agent** during the bubble forming step.

- 4.4 It follows from the above that the subject-matter of claim 1 is new over the disclosure of D5. It is also novel over the disclosure of the remaining documents. This was not disputed so that further considerations in this respect are not necessary.
5. Concerning the issue of inventive step, the appellant considered at the oral proceedings that D3 represents the closest prior art, in particular Example 3 thereof. The board can follow this approach. D3 discloses glass microbubbles of increased collapse strength having an average particle density of at least 0.4 g/cm^3 , which consist essentially of the following ingredients in wt%: SiO_2 60-90%, alkali metal oxide 2-20%, B_2O_3 1-30%, sulphur 0.005-0.5% (ie 0.0125-1.25% expressed as SO_3), R'O (R'O being CaO, MgO, BaO, SrO, ZnO and/or PbO) 0-25%, RO_2 (other than SiO_2) 0-10%, R_2O_3 (other than B_2O_3) 0-20%, R_2O_5 0-10%, F 0-5%, other ingredients 0-2%. Sulphur may be present either in the glass wall of the bubble or in the space enclosed within the bubble. The glass in the microbubbles of Example 3 contains 77.77% SiO_2 , 4.64% B_2O_3 , 0.88% P_2O_5 , 6.75% CaO, 0.17% MgO, 1.30% ZnO, 7.70% Na_2O , 0.23% K_2O , 0.006% Li_2O and 0.02% SO_3 . The sulphur content of the bubbles is 0.079%, ie 0.197%

SO₃. These glass bubbles have an average density of 0.499 g/cm³ and a relative compressive strength greater than 50.5% (see abstract; claim 1; column 1, line 48 to column 2, line 12; column 4, lines 25 to 30; Example 3).

The technical problem to be solved with regard to D3 can be seen in the provision of glass microbubbles having an improved water resistance while retaining a good relative strength, which can be produced over a broad density range in an easy way and with high yields.

It is proposed that this problem be solved by the glass microbubbles as defined in claim 1. The claimed microbubbles differ from those of Example 3 of D3 by a higher alkaline earth metal oxide/alkali metal oxide (RO/R₂O) ratio, a higher alkaline earth metal oxide content and a higher sulphur content of the glass. In view of (i) the examples of the patent in suit, (ii) the statement on page 2, lines 31 to 36 that the microbubbles are water resistant and can have the superior performance properties attributed to the microbubbles of D3 and (iii) the additional comparative examples submitted by the respondent with his letter of 21 April 1999, it is credible, in the absence of evidence to the contrary, that this problem has actually been solved by the claimed microbubbles. The appellant's argument that the comparative tests are not suitable to show an improvement of the water resistance because the microbubbles of Example 3 of D3 should have been compared with those of Example 1 of the patent in suit instead of Examples 3 or 5, cannot be accepted by the board. The microbubbles of Examples 1, 3 and 5 of

the patent in suit have a RO/R_2O ratio of 1.21, 1.86 and 2.24 respectively, whereas in Example 3 of D3 the said ratio is 0.87. As pointed out by the appellant the RO/R_2O ratio in Example 1 is closer to that of the prior art. However, the improvement of water resistance obtained with the microbubbles of Example 3 over those of D3 is so important that it is plausible that an improvement is still achieved with the lower value of 1.2 for RO/R_2O . In any case, in view of the respondent's comparative examples, the mere allegation that no improvement is achieved with a ratio of about 1.2, without evidence in support thereof, would not be sufficient to deny the said improvement.

- 5.1 D3 itself discloses that the glass bubbles can be given increased water insolubility by including R'O oxides in an amount of at least 1 wt%, preferably at least 3 wt%. CaO and/or MgO are preferred, but other R'O oxides can be used in addition or instead, including BaO, SrO, ZnO, and PbO (see column 4, lines 25 to 30). Regarding the alkali metal oxides, D3 discloses that they are included together with the silica to assist in obtaining a desired molten low-viscosity condition for the formation of the glass bubbles. At least 2 wt% are used, preferably at least 5 wt% of the final glass bubbles. The alkali metal oxide content generally does not exceed 20 wt%, preferably 15 wt%, to avoid making the melt too fluid and to improve the chemical durability of the finished bubbles (see column 4, lines 5 to 16). D3 further teaches that the change during the process of bubble formation in the composition of the glass particles, eg through loss of volatile ingredients, is believed to contribute to the strength and other desired properties of the glass

bubbles (see column 3, lines 34 to 39).

In the appellant's view, the skilled person confronted with the problem of improving the water resistance of the bubbles of D3 without impairing their strength would, in view of this teaching, have contemplated increasing the amount of alkaline earth metal oxide of the glass of Example 3 to such an extent that the RO/R_2O ratio would lie within the claimed range. This argument is not convincing for the following reasons. It was indeed well-known before the priority date of the patent in suit that the water resistance of silicate glasses can be improved by adding alkaline earth metal oxides, such as CaO , MgO and BaO , and that CaO was mainly used to achieve this goal (see D35, page 135). However, the skilled person was also aware of the fact that soda lime silicate glasses tend to become unstable and to devitrify when the RO/R_2O ratio is increased beyond a certain limit unless stabilizing agents such as alumina are included into the composition. D13, which illustrates the general knowledge before the priority date, discloses this tendency toward devitrification for commercial silicate glasses (see pages 75 and 77). Although D22 and D23 show that soda lime silicate glasses not containing alumina and having a RO/R_2O ratio of 1.3 or more exist, this does not mean that they can be produced in an easy way or on a commercial scale and have a low tendency toward devitrification. Further, D3 was not the sole prior art document available to the skilled person in the technical field concerned. D7, like D3, concerns the manufacture of glass microbubbles from soda lime silicate glasses optionally containing up to 10% boron oxide and deals with the problems of water resistance

and strength of the microbubbles together with the problem of devitrification. D7 discloses a process for the manufacture of glass microballoons in which a glass frit containing sulphur (SO_3) is reheated to convert the glass particles to the bubble state, the sulphur compound serving as a blowing agent during the bubble forming step. The glass frit has the following composition in wt%: SiO_2 60-75%, Na_2O 10-18%, K_2O 0-5%, $\text{Na}_2\text{O} + \text{K}_2\text{O}$ 13-19%, MgO 0-7%, CaO 5-15% BaO 0-7%, $\text{MgO} + \text{CaO} + \text{BaO}$ 5-15%, B_2O_3 0-10%, Al_2O_3 0-3%, SO_3 0.3-1.0% (see claim 1; page 1; page 2, lines 16 to 18, of the translation). D7 teaches that the glasses having a tendency toward devitrification have the drawbacks of being difficult to handle and lead to microbubbles having a decreased strength due to devitrification. According to D7, when Na_2O is less than 10% or when the total amount of $\text{Na}_2\text{O} + \text{K}_2\text{O}$ is less than 13%, the glass becomes hard or may tend to devitrify. With $\text{Na}_2\text{O} > 18\%$ or $\text{Na}_2\text{O} + \text{K}_2\text{O} > 19\%$ the water resistance of the microbubbles becomes defective. Workability and water resistance may be improved by the presence of up to 5% K_2O . When CaO or the total amount of $\text{CaO} + \text{MgO} + \text{BaO}$ exceeds 15% the glass tends to devitrify while it tends to be hard for amounts $< 5\%$. Although MgO improves the workability, the glass also tends to devitrify if MgO exceeds 7%. Boron oxide in amounts up to 10% is said to improve the water resistance of the glass and to decrease its viscosity. Al_2O_3 improves the water resistance of the glass; however the glass becomes too hard when the amount exceeds 3%. Of the six examples disclosed in D7, only the glass frit of Example 6 contains boron oxide and the $\text{RO}/\text{R}_2\text{O}$ ratio of the frit is 0.62. The glass frit of Example 1 which has a $\text{RO}/\text{R}_2\text{O}$ ratio of 1, leads to microbubbles whose strength is lower than that obtained

in Example 6 for an identical density of 0.5 g/cm³. In view of this document the skilled person is warned about the increased risk of devitrification when decreasing the amount of alkali metal oxides and increasing the amount of CaO or alkaline earth metal oxides. Furthermore, as set out above, D7 suggests other possible solutions for improving the water resistance which do not increase the risk of devitrification. In these circumstances, the skilled person confronted with the problem stated above might have considered increasing the content of alkaline earth metal oxides in the glass frit of Example 3 of D3 but not to such an extent that the RO/R₂O ratio comes close to the limit where devitrification might occur. The board considers that working close to the limit where the glass tends to devitrify does not represent an easy or convenient way of preparing the glass microbubbles. Particularly for production on a commercial scale, the skilled person usually tries to select compositions for which the tendency toward devitrification is greatly reduced, in particular because of the difficulty of keeping a uniform high temperature in all parts of the furnace on this scale. Therefore, if the skilled person had increased the RO/R₂O ratio, he would have done it only slightly and would have envisaged the other possibilities of improving the water resistance suggested in the prior art, for example in D7. Furthermore, D7 teaches that the maximum SO₃ amount remaining in the glass frit is 1 wt%. There is no suggestion in the cited prior art that an increase of the RO/R₂O content of the glass frit would have allowed the incorporation of higher amounts of SO₃ into the glass frit and thus the production of glass bubbles with low densities in an easy way and

high yields.

5.2 The Appellant further argued that D5 also dealt with the problem of water resistance of the microbubbles and thus would have given the skilled person an incentive to combine the teachings of D3 and D5. However, the board cannot agree that D5 deals with the problem of improving the water resistance. On page 15, which the appellant referred to at the oral proceedings, it is disclosed that the microballoons were washed in water and then in reagent grade ethanol, and dried. The mere mention of this washing step in water does not mean, however, that the document deals with the problem of water resistance of the microbubbles in the sense of D3 or D7. The fact that a washing step was performed in water does not give the skilled person information about this water resistance. The three compositions disclosed in D5 for the microbubbles B18A, B22A and B35D contain alkali metal oxides and alkaline earth metal oxides in such amounts that the RO/R₂O ratios are 1.99, 1.37 and 1.32 respectively. However, B18A and B22A comprise a relatively high amount of unknown components since the total amount of components is 94.6 wt% and 95.7 wt% (see page 8, Table 5). Furthermore, D5 is a scientific publication concerning the fabrication of glass microballoon laser targets for laser-induced thermonuclear fusion. The microbubble properties required in this special technical field are different from those aimed at in the case of more usual applications such as light-weight reinforcing fillers (see page 2, lines 48 to 50, of the patent in suit). Accordingly D5 does not deal with the problem of improving the water resistance of the microbubbles without deteriorating their strength, nor with the

problem of producing the microbubbles over a broad density range in high yields and in an easy way. It also contains no information from which the skilled person could have inferred that a high R_0/R_2O would be beneficial. In these circumstances and taking into account the fact that the skilled person was warned by D7 against using compositions having too a high R_0/R_2O ratio (see point 5.1 above), the board is not convinced that the teaching of D5 would have encouraged the skilled person to increase the R_0/R_2O ratio to the claimed extent in the glass of D3 in order to solve the existing problem.

- 5.3 The appellant also put forward arguments starting from D5 as the closest prior art instead of D3. In the board's judgement, however, D5 cannot be considered as the closest prior art. As already pointed out above, D5 is directed to a very different purpose and use of the microbubbles (ie as laser target for laser-induced thermonuclear fusion), whereas the microbubbles of D3, like those of the patent in suit, are used in particular as light-weight fillers. The technical problems dealt with in D5 are accordingly different. Furthermore, D5 contains no description of how the microbubbles with high R_0/R_2O ratios were manufactured and only refers in broad terms to the "permeation" method of D24. The selection of D5 as the closest prior

art mainly because of composition similarities with the claimed solution would therefore, in the board's view, have been the result of an inadmissible ex post facto analysis of the relevant state of the art.

5.4 The other documents cited during the opposition and appeal proceedings do not contain additional information which could hint at the claimed solution, when considered alone or in combination with the teaching of the documents already taken into account.

5.5 It follows from the above that the subject-matter of claim 1 according to the main request also meets the requirement of inventive step set out in Articles 52(1) and 56 EPC.

6. Claim 1 being allowable, the same applies to the dependent claims 1 to 5 whose patentability is supported by that of claim 1. Claim 6, which is directed to a free-flowing mass of particles containing at least 70 wt% of the microbubbles of any of the preceding claims, derives its patentability from that of claim 1.

Order

For these reasons it is decided that:

The appeal is dismissed

The Registrar:

The Chairman:

S. Hue

R. Spangenberg