JOURNAL OF MATERIALS SCIENCE: MATERIALS IN MEDICINE 12 (2000) 833–835

Determination of quantitative leucite content in pressable ceramics compared to conventional dental porcelains

S. ASSMANN[1], M. ERMRICH[2]*, K. KUNZMANN[3]
[1]Ducera Dental GmbH & Co. KG, Rodheimer Straße 7, D–61191 Rosbach, Germany
[2] Röntgenlabor Dr Ermrich, Am Kandelborn 7, D–64354 Reinheim, Germany
[3]INFRACOR Gmbh, P.O. Box 1345, D–63403 Hanau, Germany
E-mail: roentgenlabor-dr.ermrich@t-online.de

In the present study, the leucite content of two different veneering porcelains and one pressable ceramic have been determined by X-ray diffraction after establishing a standard, using artificial leucite. The results are directly linked to the microstructure and mechanical properties of the materials. The most homogeneous glass-ceramic exhibits the highest relative leucite content and the highest fracture strength.

1. Introduction

The first all-ceramic restoration for dental application was established at the end of the last century. These feldspatic jacket crowns are characterized by a thermal expansion coefficient ($TEC_{25-600\,^{\circ}C}$) of $7 \times 10^{-6}/^{\circ}C$. Because of the corresponding low tensile strength of 35 MPa, the structures tend to crack easily [1]. These problems were solved by a development of Weinstein *et al*. in 1962 [2, 3]. By combining two different components (marked No. 1 and No. 2) the $TEC_{25-600\,^{\circ}C}$ of the final glass ceramic can now be adjusted between 12 to $14 \times 10^{-6}/^{\circ}C$. It is thus possible to fuse this porcelain to metals generally used in dental restorations characterized by a slightly higher $TEC_{25-600\,^{\circ}C}$ of 13.8 to $15.7 \times 10^{-6}/^{\circ}C$. The good mechanical properties of these porcelain-fused-to-metal (PFM) restorations can be attributed to the high strength (combined with a ductile behavior) of the basic metal-frame in combination with the established stresses in the different (glass-ceramic- and metal-) layers [4, 5]. The relatively high thermal expansion coefficient of the porcelain can be attributed to the presence of (tetragonal) leucite ($KAlSi_2O_6$) in component No. 1 of the Weinstein Patent [2, 3]. The tetragonal form has been identified by numerous authors using X-ray diffraction (XRD) methods as the only prevailing crystalline phase in the applied glass-ceramic [6–12]. The $TEC_{25-600\,^{\circ}C}$ of these leucite crystals could be located between 20 and $25 \times 10^{-6}/^{\circ}C$ in contrast to the low value of the pure glass-matrix of 8 to $13 \times 10^{-6}/^{\circ}C$ [9, 13].

Because of the resulting stress-distribution in the material, the fracture strength of the whole glass ceramic generally satisfies the ISO value of 50 MPa [14] in contrast to the above mentioned jacket crowns. By improving the distribution of the tetragonal leucite crystals within the glass-matrix (and thus diminishing the existing flaw sizes [5]), the tensile strength in special porcelains can even exceed 100 MPa [15–17]. In contrast to the Weinstein *et al*. patent [2, 3], the material now consists only of one single component (containing amorphous and crystalline parts). The tensile strength is further improved in the so-called pressable ceramics [18]. This relatively new group of materials is used to form the basic frame of a dental restoration and thus represents one of several possible ways to avoid the use of metal in the latter [19]. Like the ordinary glass-ceramic, it consists of tetragonal leucite embedded in an amorphous phase. The high tensile strength values of up to 180 MPa [18] are generally attributed to a very homogeneous distribution of small leucite crystals in the glass matrix, realized by the processing technique [18]. By applying high temperatures of up to 1200 °C and a pressure of 5 bar, the porcelain is thereby being pressed in the desired form (lost wax technique) [20].

The aim of the present study is to determine why the various glass-ceramics (consisting of two components, consisting of one component, pressable ceramic) yield different values of tensile strength (70, 120 or 180 MPa) [15, 18]. Because of the established stress distribution within the porcelains [4], the strength should be linked to the amount of tetragonal leucite present in the different materials [21]. Furthermore, the homogeneity (and thus the existing flaw size) also has to be taken into consideration when discussing tensile strength values [5]. It is therefore of special interest to determine the fraction of the maximum leucite growth in the different materials examined. The more the theoretically possible leucite forms, the less potassium ions located in the glass matrix. Consequently, the TEC in the latter decreases and the difference in the TEC values of leucite and the glass-matrix should increase, thereby improving (theoretically) the tensile strength of the whole material.

*Author to whom all correspondence should be addressed.

To realize the aims stated above, the leucite content of three selected porcelains was determined, using XRD. The materials are labeled porcelain No. 1 (conventional glass ceramic consisting of two components [15], according to the Weinstein patent [2, 3], porcelain No. 2 (glass ceramic, consisting of only one component [15]) and porcelain No. 3 (Empress pressable ceramic [4]).

2. Experimental procedure

The porcelains were prepared using the following bakes: No. 1: $4 \times 920\,^{\circ}\mathrm{C}$ (1 min); and $2 \times 820\,^{\circ}\mathrm{C}$ (1 min); No. 2: $4 \times 780\,^{\circ}\mathrm{C}$ (1 min); 1st dentine bake with a small cooling rate; and $2 \times 720\,^{\circ}\mathrm{C}$ (1 min); No. 3: Pressing temperature $1180\,^{\circ}\mathrm{C}$ (20 min, 5 bar) and $3 \times 910\,^{\circ}\mathrm{C}, 1 \times 890\,^{\circ}\mathrm{C}$ (3 times dentine bake and gloss).

The X-ray investigations were carried out using the STOE θ/θ-diffractometer, Cu-Kα radiation (U = 40 kV, I = 35 mA), a secondary beam monochromator and a scintillation counter (Fig. 1).

Using the equation of the so-called crystallinity index the leucite content of a partly crystallized sample was calculated using two standard samples, on the one side the starting material as the pure amorphous form and on the other side the 100% crystallized leucite phase. A least-squares fit is applied to all data points of the measured diagram to calculate the crystallinity index X_c, given by the equation:

$$\mathrm{I}(2\theta)_{\mathrm{sample}} = X_c \times I(2\theta)_{\mathrm{crystalline}} + (1 - X_c) \times I(2\theta)_{\mathrm{amorphous}} \quad (1)$$

The X_c values were determined using two different measurements of the porcelain sample itself and both the amorphous and the 100% crystalline sample. The air scatter diagram was substracted before.

In a second way five samples with known crystalline contents (and again the 100% amorphous and crystalline samples) were used to generate a calibration curve. Consequently the crystallinity indexes were determined involving three values (Fig. 2).

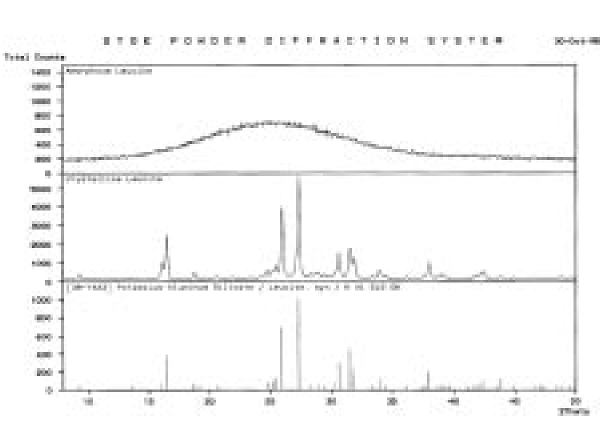

Figure 1 Diagrams of the 100% amorphous sample and the completely crystallized leucite. The leucite phase $KAlSi_2O_6$ (PDF # 38–1423, tetragonal, I41/a) is assigned.

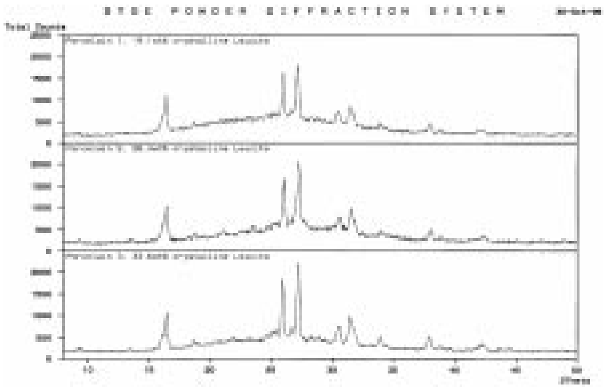

Figure 2 Diagrams of the porcelains.

3. Results and discussion

In dependence on the material, different values of the leucite content are being detected. An increase of the latter (19, 28 and 34 wt %) corresponds to an increase of the fracture strength according to the manufacturer (70, 120 or 180 MPa) [15, 18] for the porcelains No. 1, 2 and 3, respectively (see Table I). When taking the mean composition of the materials into consideration [2, 3, 4, 22], one can calculate the maximum amount of leucite that can theoretically form in the glass ceramics. Dividing the determined leucite content by this value yields the percentage of the theoretically possible growth, realized in the different porcelains. These results are summarized in the context of other important features in Table I.

It can thus be concluded that the fracture strength of dental glass-ceramics is generally improved by the following factors:

- *Homogeneous distribution of leucite crystals (tetragonal form) in the glass matrix:* the better the homogeneity, the smaller the maximum flaw size [5]. The best homogeneity is realized in the pressable ceramic due to the processing technique [18].
- *High quantitative leucite content:* because of the difference in TEC between tetragonal leucite ($20 - 25 \times 10^{-6}/\,^{\circ}\mathrm{C}$ [9, 13]) and the glass-matrix ($8 - 13 \times 10^{-6}/\,^{\circ}\mathrm{C}$ [9, 13, 15]), the compressive stress in the latter is increased with increasing amount of leucite [4]. A high content of leucite is furthermore automatically linked to a high TEC of the whole porcelain [12, 23].
- *High percentage of leucite growth in relation to the maximum growth possible:* the more the theoretically possible leucite grows, the less potassium is located in the glass matrix. The TEC of the amorphous phase is thus decreased and the difference to the (fixed) TEC of tetragonal leucite becomes bigger. The compressive stress in the matrix and consequently the overall tensile strength increases.

4. Conclusions

In the present work it could be shown that the excellent fracture strength and good homogeneity of the pressable

TABLE I Leucite content and material properties of selected dental porcelains. Unless stated otherwise, all data refer to the cited literature

Material	Homogeneity	Maximum fracture strength claimed by the manufacturer according to the cited litreature [MPa]	Measured thermal expansion coefficient ($TEC_{25-500^{\circ}C}$) [$\times 10^{-6}/^{\circ}C$]	Leucite content, determined in this work [wt %]	Theoretical leucite content, calculated according to the composition claimed by the manufacturer [wt %]	Percentage of leucite growth, accomplished in the various ceramics
Porcelain No. 1 Conventional ceramc containing two major components [2, 3, 15]	Average. Regions of high leucite content (component No. 1 of the Weinstein patent 2, 3]) are located next to regions with no leucite at all (component No. 2).	70	13.0	19.1 ± 0.1	48.7	39.92
Porcelain No. 2 Commercially available ceramic, consisting of only one component [15, 22]	Good. It can still be improved in some areas.	120	16.1	28.4 ± 0.2	52.2	54.4
Porcelain No. 3 Empress pressable ceramic [4, 18]	Very good. Small leucite crystals, homogeneous distribution.	180	14.4	33.6 ± 0.4	50.1	67.0

Empress ceramic corresponds to a high percentage of leucite growth in relation to the theoretically possible leucite content. The homogeneous distribution of leucite crystals allows 67% of all potassium ions to reach a leucite crystal by diffusion during the applied heat treatment of the materials examined. The larger the heterogeneous nature of the material, the less potassium ions can take part in the crystal growth because of the lengthened diffusion path. Consequently, only 39% of the possible leucite grows when the material is produced in the conventional way according to the Weinstein patent [2, 3].

It can thus be concluded that the homogeneity should be carefully controlled, when adjusting material properties. High fracture strength in dental ceramics is directly connected to a homogeneous distribution of leucite crystals in the glass matrix.

References

1. W. J. O'BRIAN, ''Dental Porcelains'', Encyclopedia of Materials Science and Engineering, edited by M.B. BEVER, Pergamon Press, Oxford (1996) pp. 1099–1102.
2. M. WEINSTEIN, S. KATZ and A. B. WEINSTEIN, ''Fused porcelain-to-metal teeth'', U.S. Patent no. 3 052 982 (September 11, 1962).
3. M. WEINSTEIN and A. B. WEINSTEIN, ''Porcelain covered metal-reinforced teeth'', Patent no. 3 052 983 (September 11, 1962).
4. G. BEHAM, ''IPS-Empress: Eine neue Keramik-Technologie'', Ivoclar-Vivadent-Report (1990).
5. R. W. DAVIDGE, ''Mechanical behavior of Ceramics'', Cambridge University Press (1979).
6. W. J. O'BRIAN and G. RYGE, *J. Am. Ceram. Soc.* **47** (1964) 5.
7. J. POGGIOLI and J. TULEFF, *Rev. Fr. Odontostomatol.* **17** (1970) 205.
8. C. HAHN and K. TEUCHERT, *Ceramic Forum Int/Ber. Dt. Keram. Ges.* **57** (1980) 208.
9. J. R. MACKERT JR., M. B. BUTTS and C. W. FAIRHURST, *Dent. Mater.* **2** (1986) 32.
10. J. R. MACKERT JR., M. B. BUTTS, R. MORENA and C. W. FAIRHURST, *J. Am. Ceram. Soc.* **69** (1986) C.69.
11. M. M. BARREIRO, O. RIESGO and E. E. VICENTE, *Dent. Mater.* **5** (1989) 51.
12. P. W. PICHE, W. J. O'BRIAN, C. L. GROH and K. M. BOENKE, *J. Biomed. Mater. Res.* **28** (1994) 603.
13. M. A. ROUF, L. HERMANSSON and R. CARLSSON, *Trans. J. Brit. Ceram. Soc.* **77** (1978) 36.
14. ISO 6872, ''Dental Ceramic'', 2nd Edn (1995).
15. O. KOMMA, ''Hydrothermale Dentalkeramiksysteme'', Ducera Dental GmbH & Co. KG (1993).
16. C. RISITO, H. LÜTHY, O. LOEFFEL and P. SCHÄRER, *Schweiz. Monatsschr. Zahnmed.* **105** (1995) 611.
17. M. Y. SHAREEF, R. VAN NOORT, P. F. MESSER and V. PIDDOCK, *J. Mater. Sci: Mater. Med.* **5** (1994) 113.
18. J. K. DONG, H. LÜTHY, A. WOHLWEND and P. SCHÄRER, *Quintessenz.* **43** (1992) 1373.
19. M. A. ROSENBLUM and A. SCHULMAN, *J. Am. Dent. Assoc.* **128** (1997) 297.
20. A. WOHLWEND, ''Verfahren und Ofen zur Herstellung von Zahnersatzteilen'', Europäische Patentanmeldung 0 231 773 (1987).
21. I. L. DENRY, J. R. MACKERT JR., J. A. HOLLOWAY and S. F. ROSENSTIEL, *J. Dent. Res.* **75** (1996) 1928.
22. Duceragold Patent No. DE 4 031 168C2.
23. J. R. MACKERT JR., A. L. EVANS, *J. Dent. Res.* **70** (1991).

Received 19 July
and accepted 22 July 1999