Contact Mechanics in Dentistry:

A systematic investigation of modern composite materials used for fillings

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Abstract

In the present investigation it is our concern to investigate the stability and reliability of a great variety of new denture materials. Using nanoindentation, these new composite fillings have been tested with respect to their ability to withstand contact loadings.
Producing durable, aesthetic attractive dentures is still a challenging task in modern dentistry. As most of the clinic studies and long time investigations show, the human teeth structure and material composition still seems to be unrivalled by any of the known fillings and dentures. Thus, to copy the physical parameters of the materials a human tooth consists of, is in our opinion the best way to a successful, long lasting substitute of injured and damaged teeth or their parts. Within the present study, 10 different filling materials have been investigated thoroughly according to their basic mechanical parameters like hardness, Young's Modulus and homogeneity by using an UMIS-2000 nanoindenter. In addition, the yield strength has been determined by applying the method of the effectively shaped indenter. The results show that even the most modern composite materials are still very limited in their ability to reproduce the mechanical strength of human teeth. It has also been demonstrated that over-aged composite materials show a significant decrease of the mechanical quality parameters.

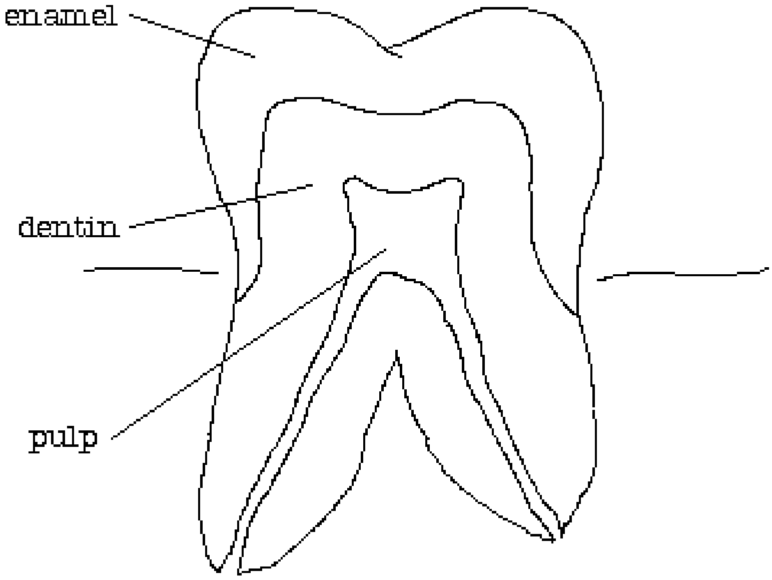

fig.1: Structure of a natural tooth

Introduction

The human tooth is not made of a homogeneous material [1] (fig. 1). Different biological substances - such as cement, dentin and dental enamel - with different mechanical / physical properties make up a complex compound which is adapted optimally to its function (table 1). We find similar values in the studies by Mahoney et al. [2] and Kinney et al. [3]. Even within the dental enamel which is one of the hardest structures in the human body, there are noticeable differences in mechanical parameters [4] (hardness, Young's Modulus) (fig. 2).

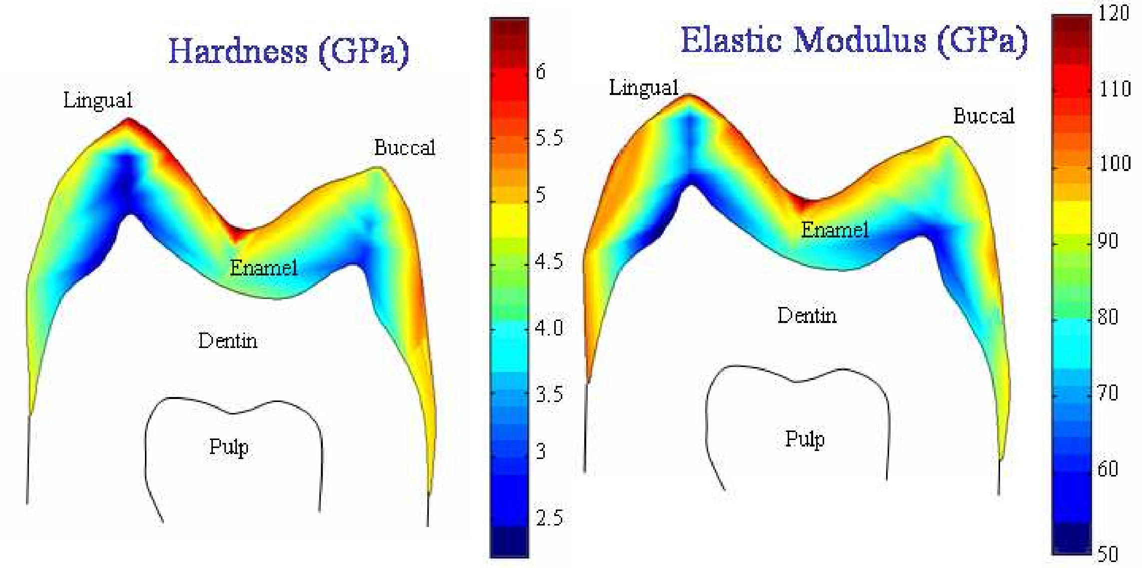

fig.2: Mechanical parameters of a human tooth [2]

Material	Hardness H (GPa)	Young's Modulus (GPa)
Enamel	4.00	85
Dentin	0.83	23

table 1: Mechanical parameters (averaged) of a human tooth (molar)

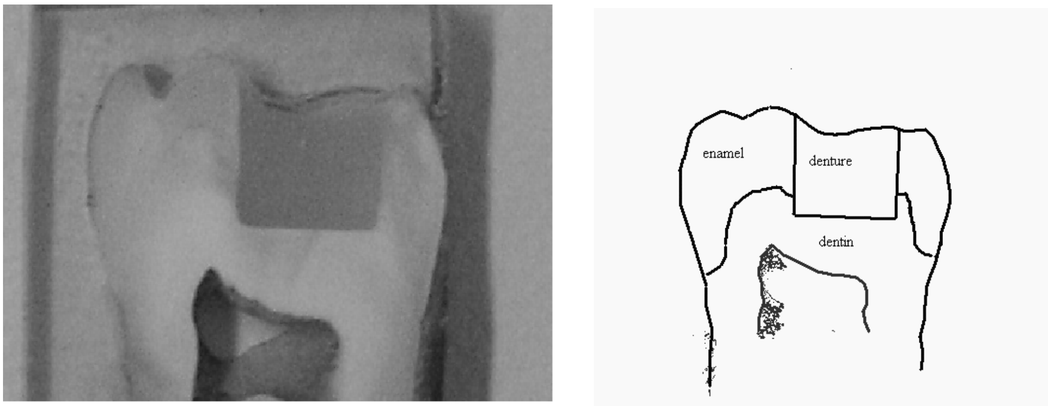

fig.3: Cut through a human tooth with composite filling adjacent to dentin and enamel

All composite materials used for fillings that exist on the market are macroscopically homogeneous. That is why there must be compromises as a defect (caries), which usually affects more than one natural part of the tooth, is filled with such a uniform material (fig. 3). Our investigation does not deal with the structure of every single composite material. This structure can be very complex and in some cases even differ widely. We differentiate microfiller-composites (particles < 2µm) and macrofiller-composites (particles > 10µm) from hybrid-composites, which consists of both of the types above mentioned (fig.4). The point of this practically orientated study, however, is to determine the relevant mechanical parameters in a rather phenomenological manner and to compare them with the natural tooth substances enamel and dentin. For that, we have analysed ten current dental-coloured adhesive filling materials (composite materials). An eleventh sample, an overaged composite has also been investigated. As the human tooth is subjected to contact mechanical loadings, which in fact is its main purpose, we applied indentation experiments in order to determine the resistance of these filling materials. It is clear, that by using indentation with a variety of indenters we can come close to the natural mechanical loading conditions and thus would also – in principle – be able to reproduce the typical failure mechanisms occurring in the repeated contact mechanical stresses a human tooth has to endure during its live.

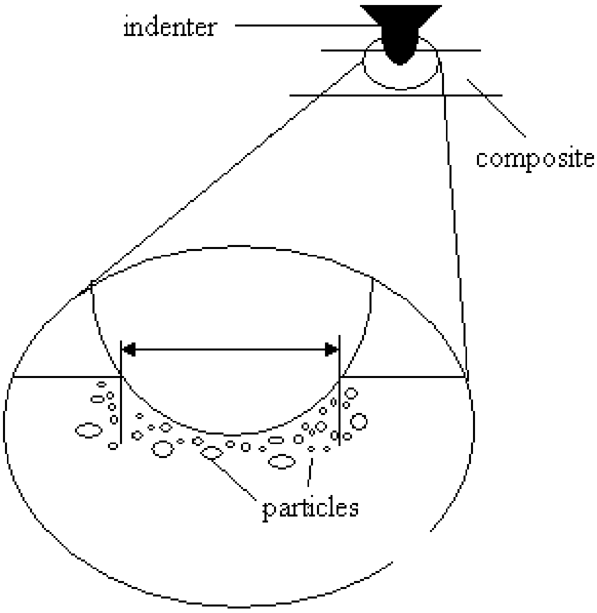

fig.4: Nanoindentation – size of the indenter compared with the particles in the composite

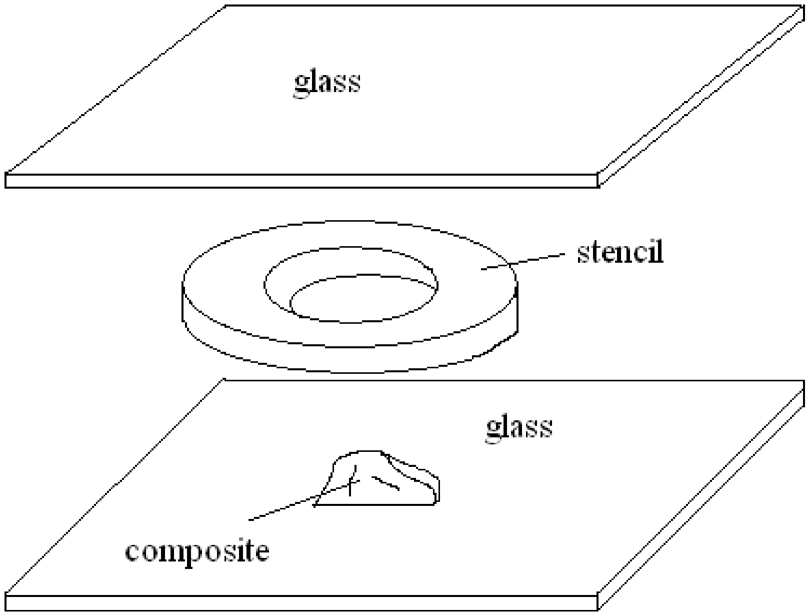

fig.5: Description of the sample preparation

Sample preparation and experimental equipment

All samples have been prepared under the same circumstances. For this, a single portion of the material (advantage: homogeneous material entry without cavities) was applied to a stencil of 1mm thickness (fig.5). This helped to model the sample's shape and enabled a non-adhesive bonding with the composite material. Additionally, the stencil was isolated with vaseline (Vas.album). Two sheets of glass as boundaries made sure that the hard-treating rays of light (wavelength 450-475nm, 900mW/cm^2) had easy access, and allowed the samples to shrink without tensile stresses towards the centre of the volume, respectively towards the source of light. The time for the polymerisation for each sample amounted to 2 x 40 sec. through both sheets of glass and additionally 2 x 40 sec. under a vaseline layer to reduce the oxygen-inhibition-layer.

This procedure is not equivalent to the usual process in a dental practice. It is rather a fact that due to the extended time of polymerisation and hardening of a thin layer from both sides in our laboratory, a higher interconnection of the monomers could be reached. Furthermore, the adhesive bonding of the composite materials with the tooth substance leads to much higher tensile stresses because of shrinking vectors within the polymer due to an overall reduction of the net volume of the filling material.

Finally, we carried out a mechanical treatment to remove any possible partial oxygen-inhibition-layers that might still have existed. The samples have been kept in Aqua.

Nanoindentation

The measurements were carried out with a UMIS-2000 nanoindenter (CSIRO, Australia) at the Institute of Physics of the Technical University of Chemnitz. 30 measurements were done on every sample with a Berkovich indenter at loads of 50mN, 100mN and 300mN. The load was applied in 58s using quadratic load steps to achieve approximately constant displacement rates (see for instance [5]). A hold period of 60s followed for the investigation of creep effects and to reduce creep influences on the unloading curve. Unloading was done in about 45s. A thermal drift correction could not be applied due to the strong viscoelastic behaviour of the materials even after a reduction of the normal load to 10%. However, the thermal drift rate of the instrument, obtained for a large number of measurements on other materials, was typically below 0.05 nm/s. The depth change in a measurement time of 163 s was therefore only about 8 nm in relation to indentation depths between 1.8 µm and 7 µm and a thermal drift had no influence on the results.

The ten measurements per load were averaged before the analysis to reduce the influence of the material inhomogeneity. Single measurements which markedly differed from the other, for instance with a strong kink in the curve caused by a pore, were excluded from averaging. The displacement standard deviation of the remaining curves was used as a measure of the material homogeneity. The minimum contact depth at the smallest load was about 1.8 µm corresponding to a contact area of about 92µm². This is much larger than the size of a single ceramic particle in the fillings (<2µm, see fig.4). Accordingly a load-displacement curve characterizes the deformation behaviour of both: the matrix and the embedded particles. Finally the results for the three different loads were again averaged to get a representative value for hardness and modulus of the material, independent of the measured gradient of the properties. This can be motivated by the fact that the scatter of the single results was in the dimension of the property change along the depth.

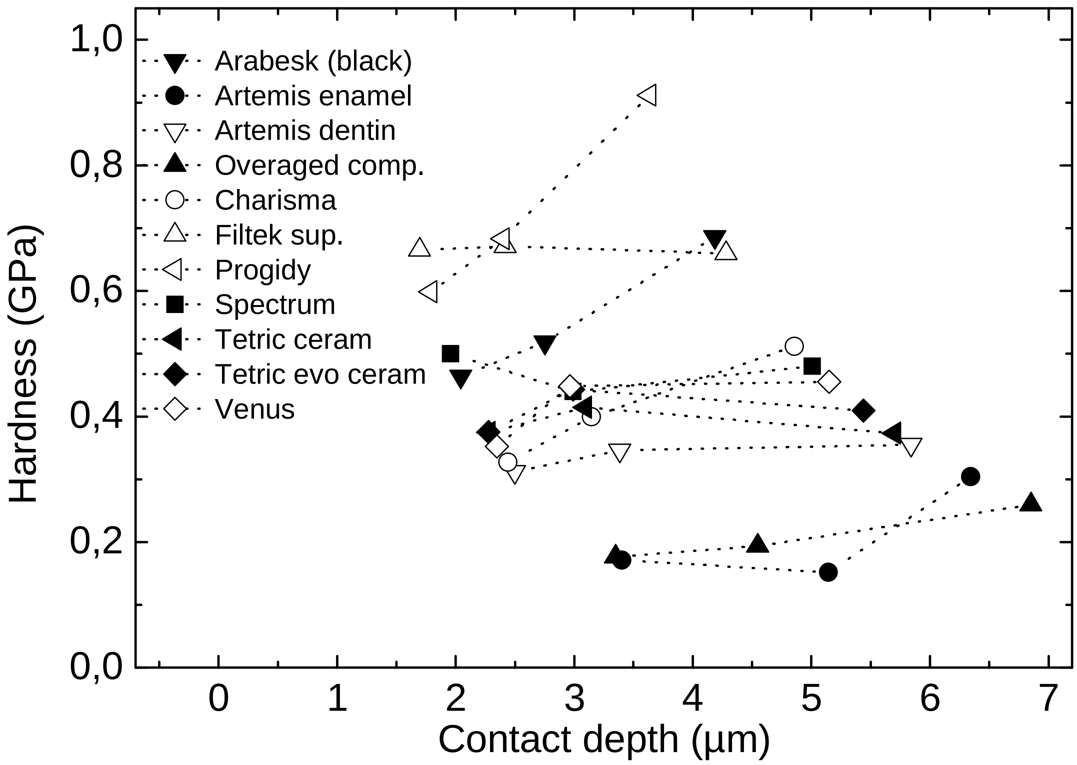

fig.6: Hardness as function of contact depth for the investigated materials. The points for every material belong to loads of 50mN, 100mN and 300mN.

Analysis

Hardness H and Young's Modulus E

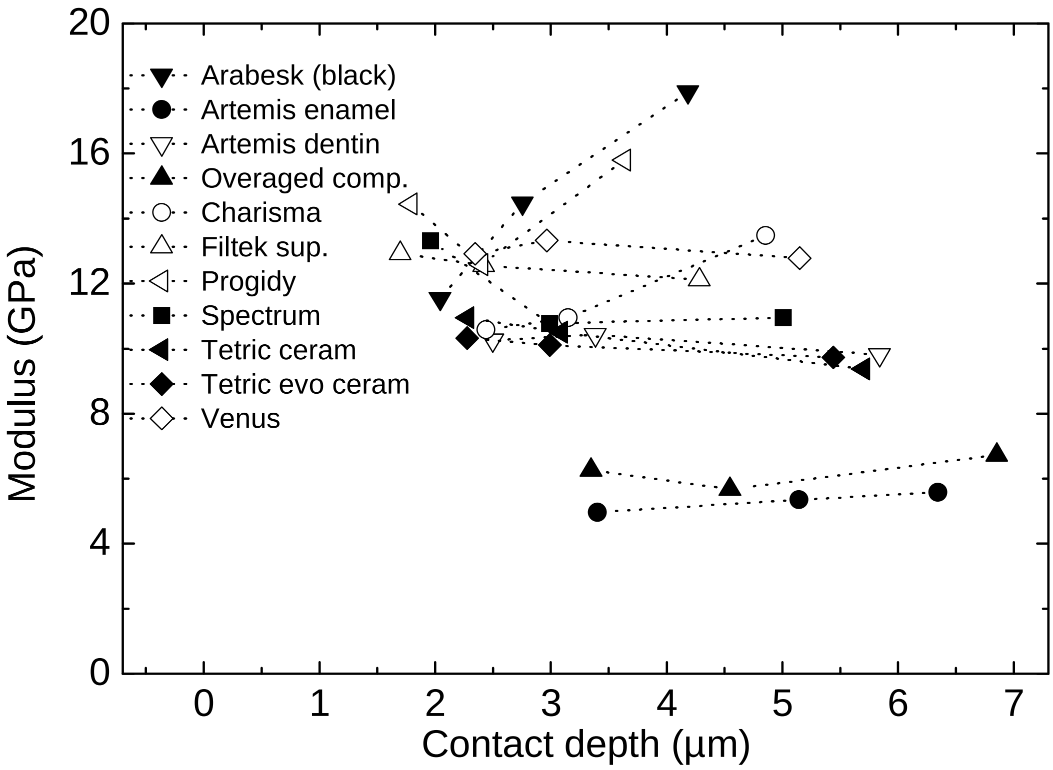

fig.7: Hardness as function of contact depth for the investigated materials. The points for every material belong to loads of 50mN, 100mN and 300mN.

The calculation of hardness and modulus was done according to ISO 14577. The unloading fit range for the fit with a power law was between 98% and 40% F_{max}. The depth dependent results are shown in fig. 6 and 7. A slight hardness decrease towards the surface can be observed. Possible reasons for this effect are particle depletion in the near surface region and residuals of the oxygen inhibition layer (incomplete polymerisation, residual monomers). The average results including error bars (one standard deviation) are given in fig. 8 and 9. The value for human dentin is additional given for comparison.
For mechanical stability and low wear not only high hardness is required. The hardness to modulus ratio (elasticity of the material) is another important criterion. It is well known in tribology that wear is reduced with increasing H/E ratio. Greenwood and Williamson used this ratio in their prominent plasticity index [6]. The ratio is therefore used for a ranking of the investigated materials.

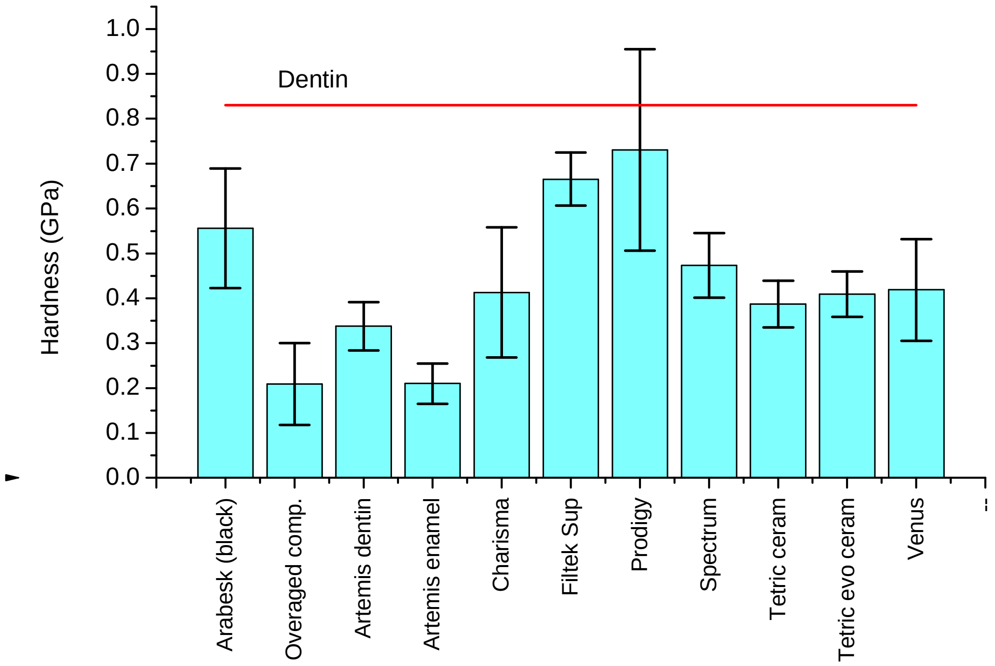

fig.8: Average Hardness of the investigated materials with error bars. The red line depicts the hardness value of dentin for comparison

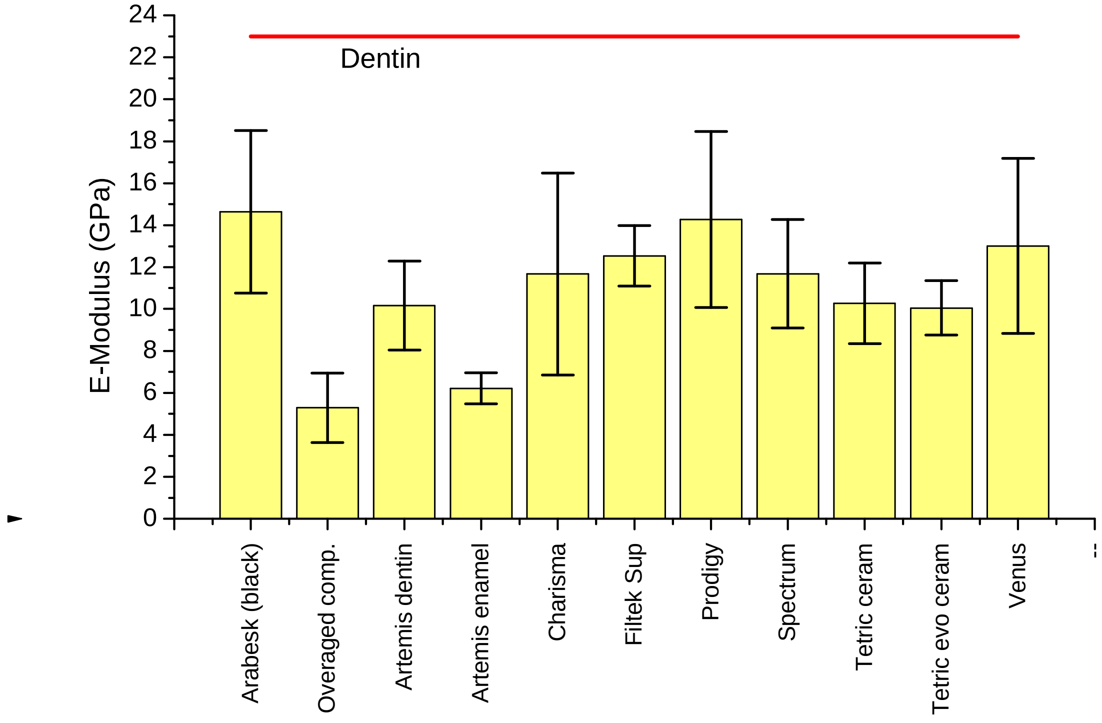

fig.9: Average Young's modulus results with error bars. The red line depicts the Young's Modulus of dentin for comparison

Creep

The creep behaviour of the different materials at maximum load of 300mN is shown in fig.10. Significant differences between the materials can be observed. The depth change during a period of 60s was relatively high with values between 200nm and 600nm. Low creep is desirable for a better mechanical stability of the fillings however, the creep behaviour was not considered in the ranking because it is closely related to hardness.

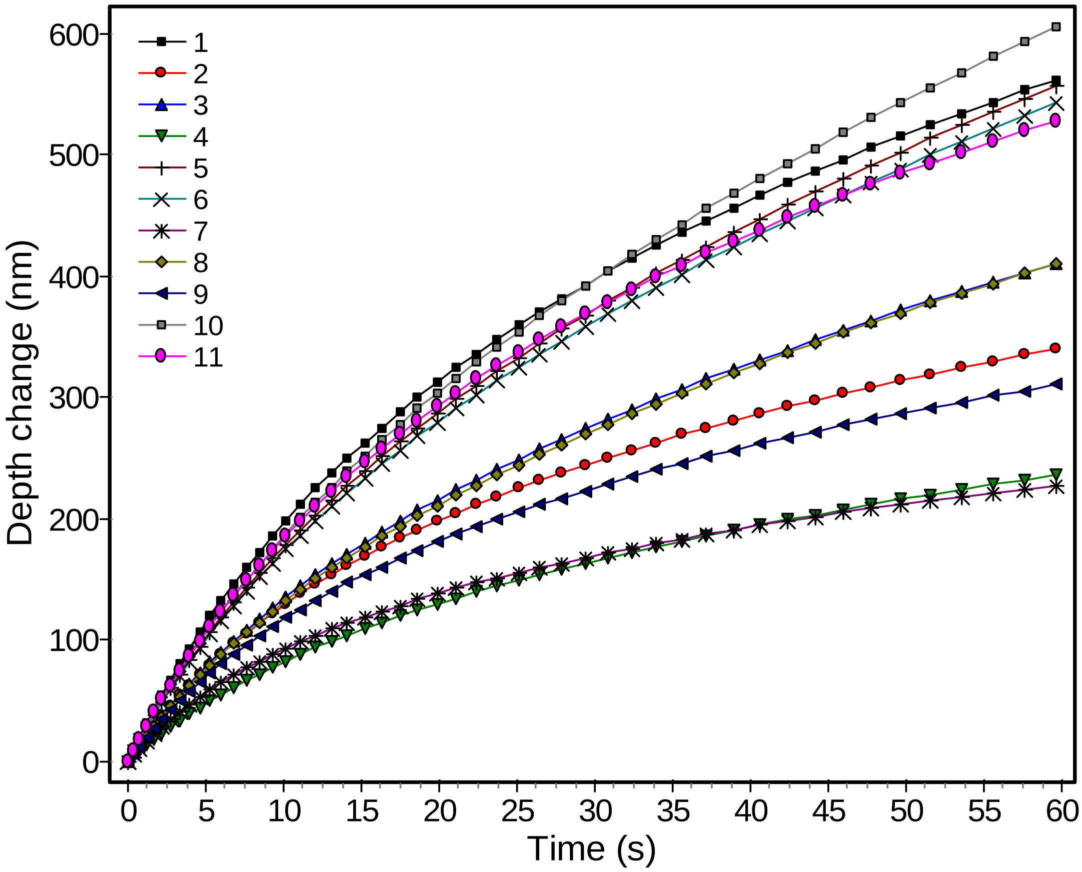

fig.10: Creep behavior of the investigated materials during the hold period of maximum force.

Homogeneity

The standard deviation of the averaged measurements divided by the hardness or modulus average was used as homogeneity criteria (fig. 11). The composites consist of an organic matrix with about 40 vol. % and hard ceramic particles (60 vol. %). In the microscopic dimension the tooth filling materials are therefore inhomogeneous. The organic monomer is responsible for the necessary consistency. After polymerisation a composite material forms which may locally contain different particle density. Further cavities (bubbles) may occur. In an ideal material the ceramic particles are homogeneous distributed without any cavities. Cavities are often the initial point for cracks or bacterial infections. The dentist should therefore try to reduce the volume shrinking of the polymer during the polymerisation to a minimum by an incremental technique (stepwise polymerisation).

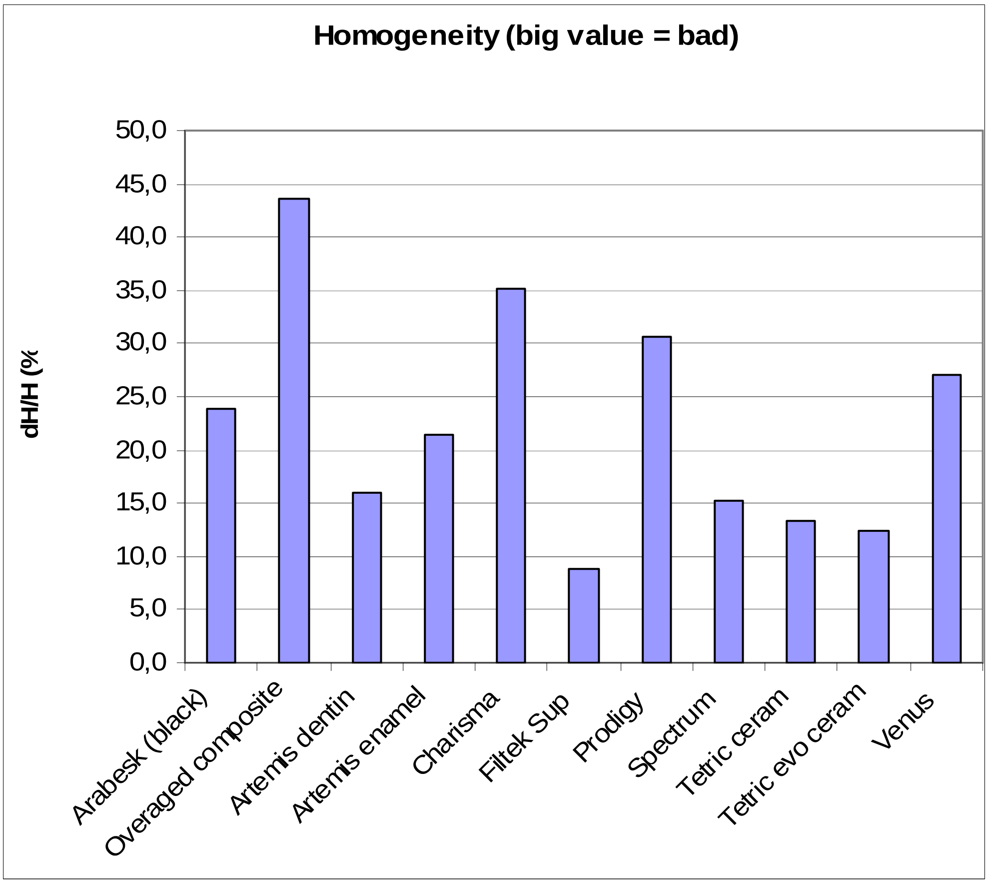

fig.11: Homogeneity of a variety of dental filling materials

Yield Strength

The Yield Strength has been evaluated directly from Berkovich indentation measurements using the method of the effectively shaped indenter [7]. The results are presented as a bar chart in figure 12. It is not surprising that the Yield Strength almost mirrors the hardness behaviour of the filling materials. This can be explained by the help of the extending cavity model of Marsh [8] which gives a relationship between yield strength and the modulus to hardness ratio of a material. For a group of similar materials a relatively constant proportionality factor between hardness and yield strength can be given, the so called constraint factor (e.g. see [9]). This is confirmed for instance by investigations in [10].

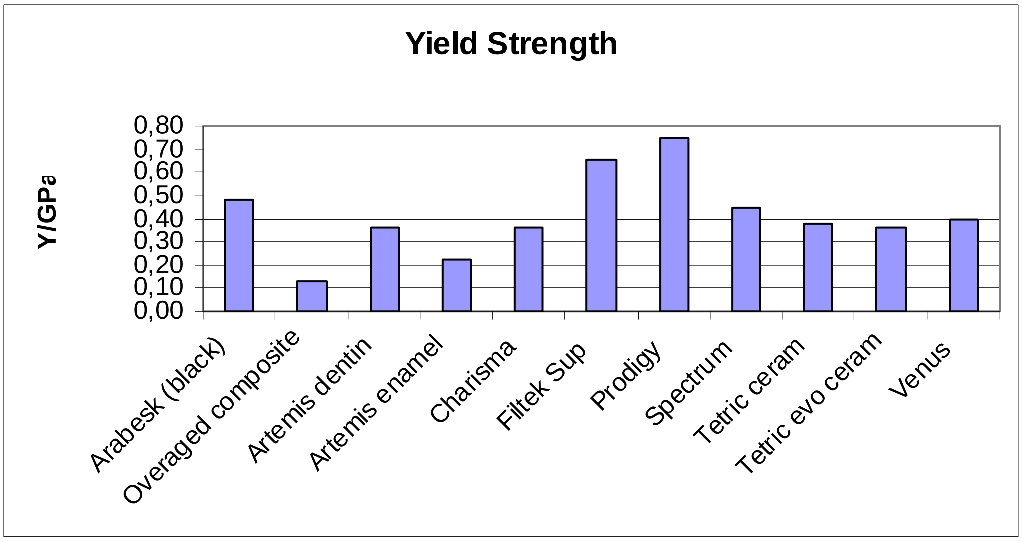

fig.12: Yield Strength of a variety of dental filling materials

Conclusion

A ranking of the investigated materials using hardness, H/E-ratio and homogeneity as criteria allows a benchmarking or "ranking list" (see table 2). Since there is no additional information which would allow to assign weight factors for the three properties we have chosen the simplest case and used weight factors of one. One argument may be that the ranking would not much change if different weights would have been used. Filtek Supreme as well as Prodigy and Tetric evo ceram. perform best although all of them are still far away from the mechanical properties of human tooth material, especially from that of enamel as a by far harder material. The search for more suitable filling materials is still a high demand but the development of such materials is not an easy task and requires beside the know how of the dentists the help of material scientists, physicist, chemists and other experts. Not considered here are other properties which are important for a dentist like aesthetics, processing properties and costs.

Material	Hardness	H/E	Homogeneity	Total	Position
Arabesk (black)	3	6	7	16	5
Overaged composite	11	5	10	26	11
Artemis dentin	9	10	5	24	8
Artemis enamel	10	9	4	23	7
Charisma	6	8	11	25	10
Filtek Sup	2	1	1	4	1
Prodigy	1	2	9	12	2
Spectrum	4	4	6	14	4
Tetric ceram	8	7	3	18	6
Tetric evo ceram	7	3	2	12	2
Venus	5	11	8	24	8

table 2: Ranking list of composite materials for dental fillings. Number one in each column is the material with highest hardness or H/E-ratio and with highest homogeneity, i.e. lowest ΔH/H value. The total column is the sum of the ranking numbers in the single columns and position is the place in the ranking.

The results for the some years overaged composite sample with its bad mechanical properties indicate that a long storage may result in a deterioration of the material. This is an important message for the dentist. However, it is also possible that there was already an improvement in the manufacturing of the materials in the last years.

Outlook

Many aspects still remain to be clarified. The link between artificial and natural material is a well known weak point. Enamel and composite material react strictly water-repellent, whereas the dentin, being a living material, attracts water. The characteristics of the artificial material that have been measured during the present study go much better together with the dentin than with the enamel. This however, must be considered as a disadvantage concerning the live time of a filling, because the mechanical properties of the dentin are not configured for bearing loads appearing at the chewing process. As that only enamel is fitted to withstand these forces. That is the reason why artificial filling materials should be adapted to the enamel. The present inevitable shrinking of the composite material due to the polymerisation even complicates matters as this leads to further tension and stresses. These stresses and strains need to be investigated more closely.

In further studies we should not only rely on the mechanical parameters but should also approach discerningly the boundary layers. We can infer from the study of T. Weihs, John Hopkins University [4] (see fig.2) that there is a continuous adaptation of hardness and Young's Modulus of the dental enamel near the dentin towards the dentin. We find a similar structure in the mechanical parameters of the dentin [3, 11]. The artificial materials misses this adaptation completely! Furthermore, it still remains to be clarified how the filling materials react on temperature changes which inevitably occur during ingestion. The further development of composite materials for dentistry should go to multilayered systems that are noticeable harder and that show a reduced polymerisation shrinking.

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