ISSN 2176-5480

22nd International Congress of Mechanical Engineering (COBEM 2013)
November 3-7, 2013, Ribeirão Preto, SP, Brazil

FUNCTIONALLY GRADED MATERIALS APPLIED TO DENTAL RESTORATIVE SYTEMS - A BIOINSPIRED APPROACH

Bruno Henriques
PPGCEM - UFRN, Brazil; CT2M – Universidade do Minho, Portugal
brunohenriques@dem.uminho.pt

Georgina Miranda
CT2M – Universidade do Minho, Portugal
gmiranda@dem.uminho.pt

Michael Gasik
Aalto Foundation University, Finland
michael.gasik@aalto.fi

Júlio Souza
CT2M – Universidade do Minho, Portugal
jsouza@dem.uminho.pt

António Eduardo Martinelli
PPGCEM - UFRN, Brazil
aemart@uol.com.br

Filipe Silva
CT2M – Universidade do Minho, Portugal
fsamuel@dem.uminho.pt

Rubens Maribondo Nascimento
PPGCEM - UFRN, Brazil
rmaribondo@ufrnet.br

Abstract.
The aim of this work was to study the employment of the functionally graded materials concept on the mechanical performance of dental restorative systems. This was inspired in natural teeth, which are composed of two distinct materials: enamel and dentin with elastic moduli of ~65 and ~20 GPa, respectively. They are bonded by dentin-enamel-junction (DEJ) where the Young's modulus changes linearly from that of enamel to that of dentin, thus reducing dramatically the stress in the enamel when loads are applied and acting as a natural functionally graded material (FGM).
The concept of functionally graded materials (FGM) has been first used in the aerospace industry but has rapidly derived to other fields of interest such as automotive, energetic and finally to medical. The application of such concept to the fabrication of metal-ceramic dental restorations has recently been studied and has shown dramatic improvement in the performance of these restorative systems. A comprehensive study about the influence of a 50%M composite interlayer and a compositionally graded transition (FGM interlayer) at the metal-ceramic interface was performed by FEM analysis. The FEM analysis was performed based on experimental data obtained from Dynamic Mechanical Analysis (DMA) and Dilatometric (DIL) studies of metal/ceramic composites. Results have shown differences on the stress state at metal-ceramic interface region of the functionally graded metal-ceramic systems when compared to those observed for conventional metal-ceramic systems with a sharp transition between metal and ceramic.
The reduction in stress magnitude and smoothness of the stress distribution profile arising in FGM architectures has been suggested to account for performance improvements of metal-ceramic systems in regard to their clinical performance. The findings of this study are in good agreement with the experimental data obtained for metal-ceramic restorative systems and demonstrated elsewhere. The employment of these new FGM restorative systems in dental prosthetics is expected to significantly improve the clinical performance of the novel restorative systems restorations.

Keywords: *thermal residual stresses, denta1 restorations, functionally graded materials, metal, ceramic.*

1. INTRODUCTION

Dental restorations such as crowns and fixed partial dentures (FPD) are designed to restore functionality and aesthetics to failed teeth. The failures of aforementioned restorative systems are undesired occurrences as they often imply money expenditure and discomfort to patients. The main causes to which dental restorative systems failures are

ISSN 2176-5480

attributed are: incorrect selection of materials; the incorrect processing of materials; incorrect design of the component; presence of defects (e.g. cracks and pores) in the prostheses, either in the veneering porcelain or in the substructure; and the interfacial breakdown of the bond between the veneering porcelain and the substructure (metal or ceramic) (Yesil et al., 2009; Özcan M., 2003; Anusavice, 2012; Swain, 2009). Several studies have been addressed to this topic and new solutions and approaches have been proposed in order to strengthen the restorative base materials (mainly the porcelain) and, more importantly, the bond that is established between them (Anusavice et al, 1977a and 1977b; Maira and Padipatvuthikulb, 2010; Carrado and Palkowski, 2010; Özcan and Uysal, 2005; Bieniaś et al., 2009). The bond strength between porcelain and metallic substrates has been reported to increase due to the following procedures: addition of easy-oxide forming elements to noble alloys (Hautaniemi, 1995); coating of reactive metals surfaces (e.g. titanium alloys; CoCrMo alloys) with oxide-controlling elements (Özcan and Uysal, 2005; Bieniaś et al., 2009; Elsaka et al., 2010); use of bonding agents (Yesil et al., 2009; Wagner et al., 1993), among others.

More recently, alternative methods based on Functionally Graded Materials (FGM) philosophy have been proposed aiming the enhancement the overall performance of metal-ceramic and all-ceramic dental restorative systems. Graded dental restorations have been shown to display improved features relative to conventional ones, namely higher resistance to contact and sliding (Suresh, 2001); higher adhesion of porcelain to the substructure (metal or ceramic) (Henriques et al. 2011; Henriques et al. 2012a; Henriques et al. 2012b; Zhang and Kim, 2009); improved aesthetical properties and improved behaviour under fatigue conditions (Henriques et al. 2012b). Another important point to which the FGM design can address is the reduction of thermal residual stresses that remains at the metal-ceramic interface during the cooling cycles after the porcelain firing. These stresses are further magnified when there is a significant difference between the thermal expansion behaviour of the metal and the porcelain. Depending on the thermal residual stress level that remains in the crown and together with those arising from occlusal loads, a catastrophic failure of the restoration can occur. FGMs have been shown to decrease significantly the thermal residual stresses formed at the interface between metals and ceramics in other fields of applications (Gasik, 2005).

In this study, the finite element analysis was used to investigate the effects of the presence of either a composite interlayer or a FGM interlayer on the stress state of the metal-ceramic interface and compare it with that existing in the conventional sharp transition. The properties variation of the FGM was determined directly from experiment by producing and testing homogeneous specimens with a range of compositions. The elastic and dilatometric experimental data sets of the metal-ceramic composite were afterwards modeled and used in the materials properties database of the FEM software.

2. MATERIALS AND METHODS

2.1. Experimental determination of the elastic modulus and the coefficient of thermal expansion

In this study a CoCrMo alloy (Nobil 4000, Nobilmetal, Villafranca d'Asti, Italy) and a dental porcelain (Ceramco3, Dentsply, York, USA) (batch number: 08004925) were used. The chemical compositions of the metallic and ceramic particles are presented in Table 1 and Table 2, respectively. The micrographs of CoCrMo and porcelain particles are shown in Figure 1. The CoCrMo particles display spherical shape and a broad size distribution: D10 = 4.44 μm; D50 = 8.27 μm and D90 = 12.76 μm.

Table 1. Base alloy composition (wt%) (according to manufacturer)

Co	**Cr**	**Mo**	**Si**	**Traces**
62	31	4	2.2	Mn, Fe, W

Table 2. Porcelain chemical composition (wt%)

SiO_2	Al_2O_3	K_2O	SnO_2	ZrO_2	**CaO**	P_2O_5	Na_2O	**Other traces**
41.3	14.5	14.0	11.9	5.8	4.1	4.1	3.0	MgO, SO_3, ZnO, Cr_2O_3, Fe_2O_3, CuO, Rb_2O

The manufacturing of the metal-ceramic composite specimens comprised the following steps: several powder mixtures with different metal/porcelain volume fractions were produced. After weighting, the powders mixtures were blended in a rotary machine at 40 rpm during 10 min. The following mixtures were produced (vol.%): pure porcelain (with 0% metal) and compositions with 20% metal, 40%, 60%, 80% and 100% metal, marked further as "nnM" where nn stays for the percentage of metal phase. Afterwards, the powder mixtures were hot pressed in a graphite die (Figure 2). The hot pressing sequence comprised the following steps: first, the cavity of the graphite die was veneered with ZrO_2 paint to prevent carbon diffusion to specimens. Then the metal-ceramic powder mixture was inserted into the

ISSN 2176-5480

cavity. The hot pressing was performed under vacuum (~10^{-2} mBar) at a temperature of 970ºC and a constant pressure of ~20 MPa. The selected heat rate was 70ºC/min and after a 2 minute stage at 970ºC the induction heating furnace was shut down.

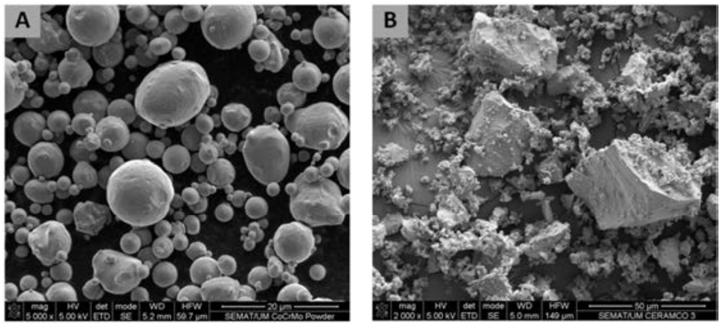

Figure 1. Micrograph of the metal and ceramic powders: A) CoCrMo particles; B) porcelain powders.

Two types of specimens' geometries were processed, rectangular and cylindrical. The dimensions of the rectangular samples used for flexural tests were 36 x 6 x 2.5 mm, while those of the cylindrical samples used for shear tests were Ø4 x 4 mm.

The measurements of the Young's moduli of all materials were obtained by the means of Dynamic Mechanical Analysis (DMA 242 C, Netzsch Gerätebau GmbH, Germany) using a three-point bending sample holder. The coefficient of thermal expansion (CTE) of composites and the monolithic materials was also assessed using dilatometry (DIL 402C, Netzsch Gerätebau GmbH, Germany). Both properties were measured through a range of temperatures starting in 100ºC up to 500ºC. Data was modeled in the software Tablecurve3D (Systat Software, Inc., San Jose, California, USA) and the surface plots of the Young's moduli and the coefficient of thermal expansion are shown in Fig. 2A and Fig. 2B, respectively. The YM variation is given by the Eq. (1) and the thermal expansion coefficient is given by the Eq. (2). The constants of both equations are shown in Tab. 3.

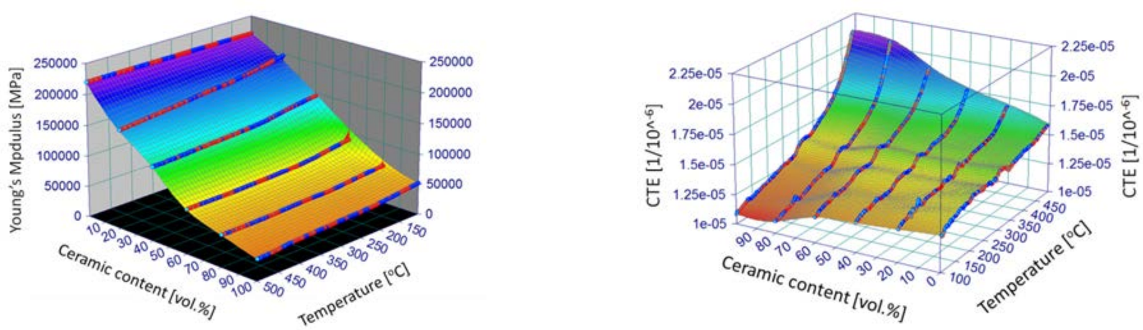

Figure 2. Plot of the mechanical (Young's Modulus) and thermal (CTE) properties of composite materials against temperature variation.

$$\mathrm{YM} = \mathrm{A} + \mathrm{Bx} + \mathrm{Cy} + \mathrm{Dy}^2 \tag{1}$$

where *x* is the temperature (degrees Celsius) and *y* is the ceramic content in the composite (vol. %);

$$\mathrm{CTE} = \frac{\mathrm{A} + \mathrm{Bx} + \mathrm{Cy} + \mathrm{Dy}^2}{1 + \mathrm{Ex} + \mathrm{Fx}^2 + \mathrm{Gy} + \mathrm{Hy}^2 + \mathrm{Iy}^3} \tag{2}$$

where *x* is the temperature (degrees Celsius) and *y* is the ceramic content in the composite (vol. %).

ISSN 2176-5480

Table 3. Constants used in Eq.(1) and Eq.(2).

	YM	CTE
A	234750.25	1.30379e-05
B	-50.803248	-6.6381e-09
C	-1865.4082	-1.6067e-07
D	-19.677519	7.89939e-10
E	0.2101322	-0.00077872
F		-2.0766e-10
G		-0.01095376
H		5.53779e-05
I		-7.3006e-10

2.2. Finite element analysis

The thermal residual stress analysis after cooling the process performed for three types of metal-ceramic interface configurations: sharp transition; 50M50C composite interlayer and FGM interlayer (sigmoid graded transition – Fig.3). In this work, cylindrical-shape specimens with a diameter of 4.5mm and a height of 8 mm were analyzed (Fig. 4). This geometry is widely used in metal-ceramic bond strength tests between dental restorative materials. For the analysis of the sharp interface between metal and porcelain, the specimen was considered to be composed by half metal and half porcelain with 4mm height in each side (Fig.4). The specimens used to analyze the effect of either the 50M composite interlayer or the FGM interlayer was considered to have a 0.4mm interlayer between the metal and the porcelain sides. The variation in the volume fraction of porcelain across the FGM interlayer is shown in Fig. 3 and is given by the following function:

$$f_{porc.} = \frac{100}{1 + e^{-\frac{(z-0.0002)}{0.00005}}} \quad (3)$$

,where *z* is the distance (in meters) to the metal surface.

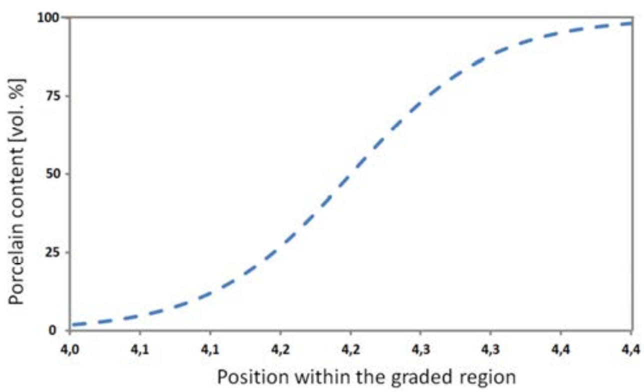

Figure 3. Variation of the volume fraction of porcelain throughout the FGM interlayer region for the Eq. (3).

The finite element analyses were conducted using the commercial finite element software COMSOL Multiphysics 4.3a (Comsol Inc, Los Angeles, USA) adapting triangular elements. In order to simplify the study, the computational model was considered to be axisymmetric for the study of thermal residual stresses after cooling (Fig.4).

The computational procedure consisted in cooling the specimens from a temperature of 500ºC to 100ºC, during 1000s in increments of 100s. The maximum principal stresses at the interfacial region were taken for each model after the cooling time elapsed.

The convergence analysis was performed in order to examine the sensitivity of the results to the size of the mesh. The obtained results for the tested meshes (with five different refinement levels) are plotted in Fig.4. Analyzing the mesh refinement one can find that the difference in maximum principal stresses, after refinement level 4 is

ISSN 2176-5480

approximately 7% in the sharp transition model and less than 3% and 1% in 50M50C composite interlayer model and the FGM interlayer model, respectively.

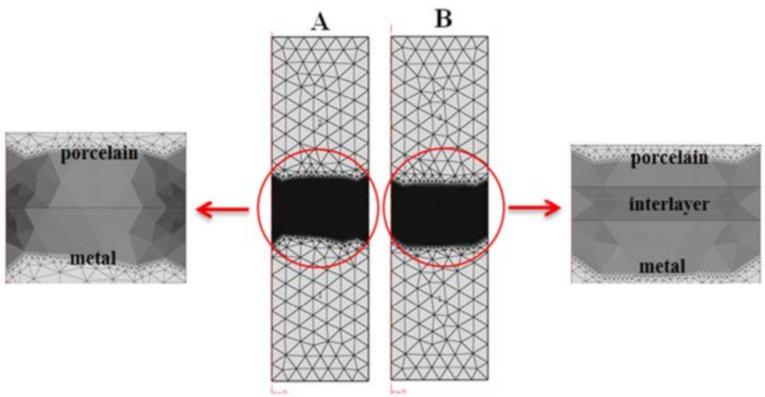

Figure 4. Finite element models with localized mesh refinenent at the interfacial region: A) Sharp transition; B) 50%M composite interlayer and FGM interlayer

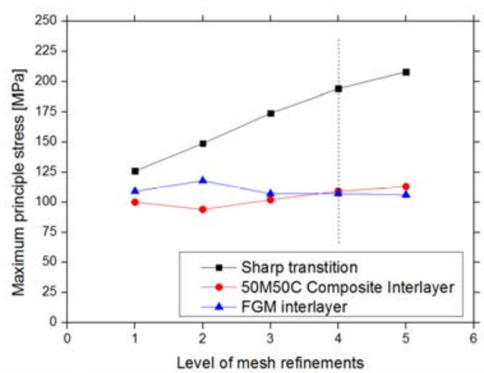

Figure 5. Mesh convergence study performed for the three models

The evolution of the coefficient of thermal expansion and the Young's modulus for the different metal-ceramic interface designs used in this study are shown in Fig. 6.

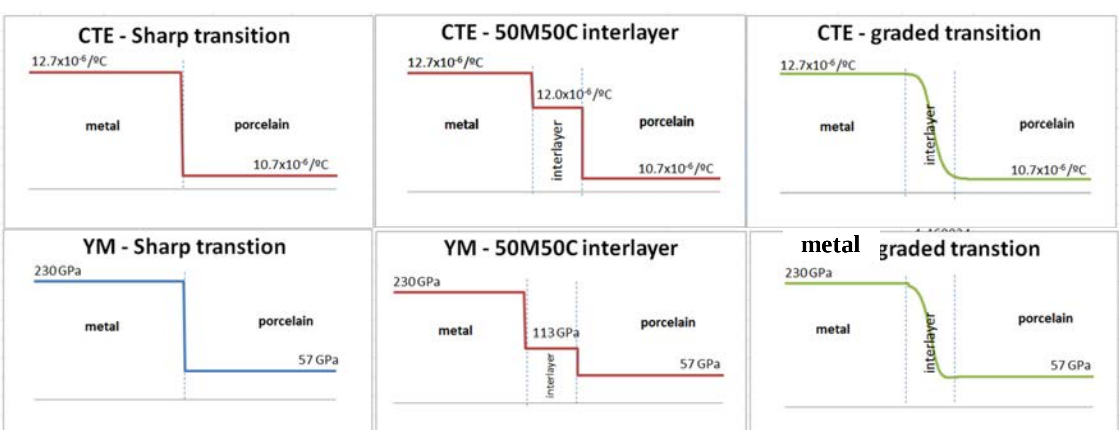

Figure 6. Evolution of the coefficient of thermal expansion (CTE) and Young's moduli (YM) across the metal-ceramic interfaces for the different specimens' configurations studied.

ISSN 2176-5480

3. RESULTS AND DISCUSSION

The results of the simulations providing information on the thermal residual stresses remaining in the specimens after cooling are shown in Fig. 7. Thermal residual stresses can have a critical impact in the clinical performance of dental crowns. Tensile stresses located at the veneer porcelain can lead to failure when high occlusal forces are superimposed. Moderate compressive stresses are beneficial to ceramics, while excessive ones can be detrimental to the clinical performance of the restorations.

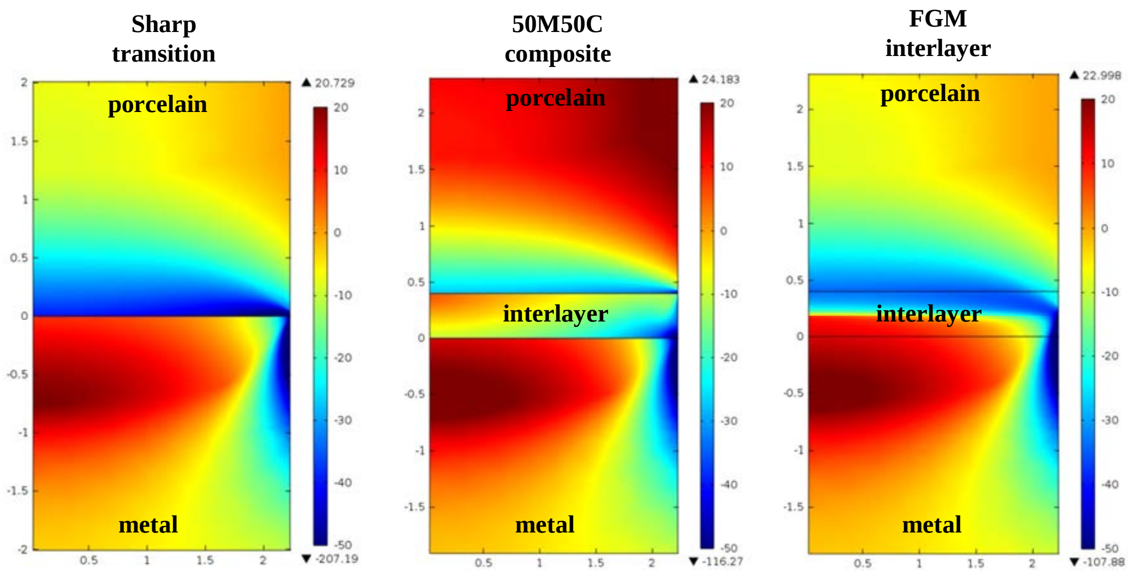

Figure 7. Maximum principal stress distribution in the different metal-porcelain models [MPa]: (a) sharp interface; (b) 50M50C composite interlayer; (c) FGM interlayer.

In Fig. 7 are plotted the maximum stresses obtained in the simulations for the three models: sharp transition, 50M composite interlayer and the FGM interlayer. It must be highlighted at this point that all models had identical nominally dimensions and the only difference between them was the interface design. The results presented in Fig. 8 show clear differences in the stress distribution and magnitude of principal maximum stresses installed in the different models. Models with interlayers, either 50M or FGM, showed lower maximum stresses than those shown by the model with conventional sharp transition. Stress reduction was 43% and 50% for 50M composite interlayer and FGM interlayer, respectively, when compared to sharp transition. These findings are in well agreement with those reported by Huang et al. (2007). They reported ~30% reduction in maximum principal stresses, originated by contact induced deformations, when functionally graded layers (FGM architecture) between the crown materials and the joins that attach them to dentin were applied. Huang et al. (2007) and Niu et al. (2009) have also shown that the functionally graded architectures, aiming at mimicking the functionally graded structures of the dentin-enamel-junctions (DEJs) in natural teeth exhibited higher critical loads resistance over a wide range of loading rates.

The presence of the interlayers has also shown to introduce a wide compressive stress field at the porcelain side adjacent to the interface, which is helpful in preventing the porcelain to crack and ultimately to fail.

Figure 8 shows the evolution of the maximum stresses along the outer surface of the models, at the end of the cooling process. This is a critical direction as cracks are likely to start at these specific locations. Arman et al. (2009) have shown that residual and transient stresses concentrate at the cervical margin of the crown restorations. The stress distribution profile at this location show clear benefits of FGM design vs. sharp and composite designs especially in the smoothness of the profile (no stress derivatives singularities have been detected for this component). The composite interlayer and the FGM interlayer promoted a significant decrease in the stresses magnitudes observed at the interface.

From the practical point of view, the application of a 50M composite interlayer between the metal and ceramic is a simpler process than the application of a FGM interlayer (Henriques et al., 2012b). Results have shown that, despite the FGM interlayer had displayed the better stress distribution profiles among the three models simulated in this study, the 50M composite interlayer may also introduce a significant improvement in the clinical long tern success of the restoration. Therefore, the 50M interlayer may be seen as a more immediate solution to be employed in today's dental restorations.

ISSN 2176-5480

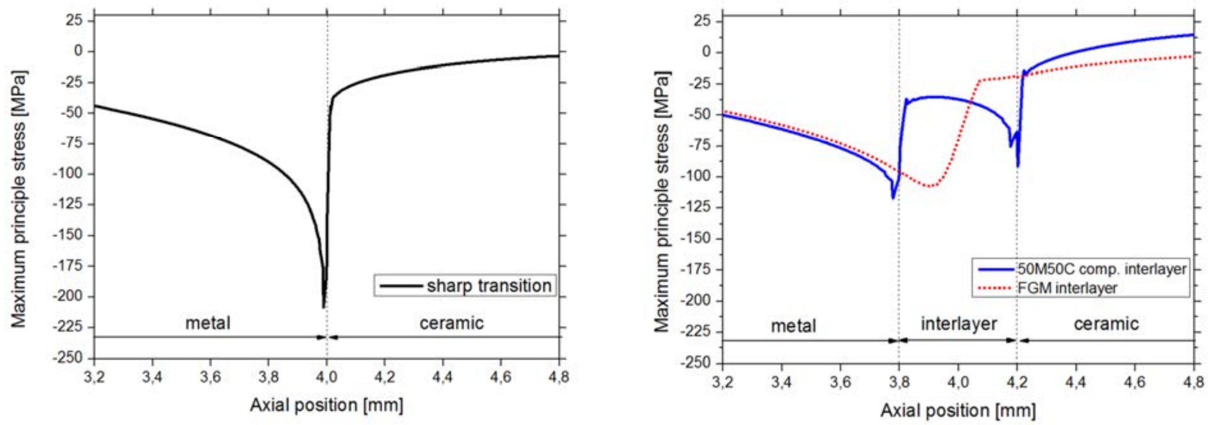

Figure 8. Maximum principal stress profiles at the outer surface of the models (r =2.25).

Figure 9 shows the transient stresses at the locations where maximum residual principle stresses were detected in the end of the cooling process. The three show similar stress profiles evolution. The compressive stresses start to form after 300s and increase monotonically until stabilizing between 800s and 900s, where the maximum stresses are reached.

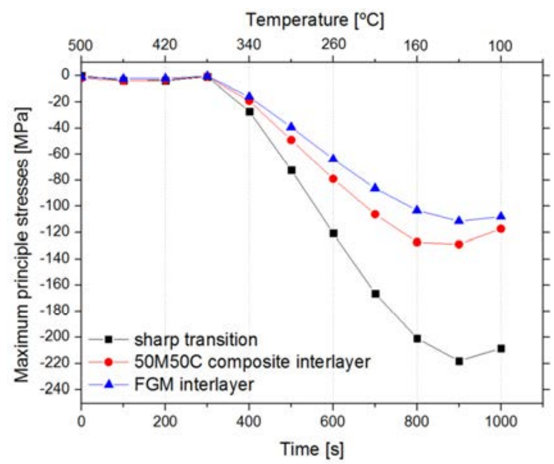

Figure 9. Evolution of stresses with time (and temperature) measured at the site where maximum principle stresses were measured (outside surface).

4. CONCLUSIONS

This paper is aimed at studying the influence of the presence of a 50M50C composite interlayer and a FGM interlayer on the thermal residual stresses upon loading at the metal-ceramic interfacial region. The finite element simulations were conducted using materials data sets obtained experimentally and showed that interlayers can contribute to significant reductions of maximum thermal residual stresses arising in the specimens after cooling. The FGM interlayer provided the smoothest stress profile, with no stress derivatives singularities in both situations. This study focused on a 2D analysis where the properties changed unidirectional along the interface. Further analysis involving the 3D geometries enclosing the complexity of real crowns should be later conducted.

5. ACKNOWLEDGEMENTS

The authors acknowledge the Portuguese Foundation for Science and Technology (FCT) and CAPES for the financial support. Assistance by MSc. M. Friman (Aalto Univeristy Foundation) in DMA and DIL experiments is also acknowledged.

6. REFERENCES

Anusavice, K.J., Horner, J.A., Fairhurst, C.W., 1977. "Adherence controlling elements in ceramic-metal systems. I. Precious alloys", *Journal of Dental Research*, Vol. 56, p.1045

Anusavice, K.J., Horner, J.A., Fairhurst, C.W., 1977. "Adherence controlling elements in ceramic-metal systems. II. NonPrecious alloys", *Journal of Dental Research*, Vol. 56, p. 1053

Anusavice, K.J., 2012. "Standardizing failure, success, and survival decisions in clinical studies of ceramic and metal–ceramic fixed dental prostheses", *Dental Materials*, Vol. 28, p. 102–111

Arman, Y., Zor, M., Gungor, M.A., Akan, E., Aksoy, S., 2009. "Elastic-plastic finite elements analysis of transient and residual stresses in ceramo-metal restorations", *Journal of Biomechanics*, Vol. 42, p. 2104-2110.

Bieniaś, J., Surowska, B., Stoch, A., Matraszek, H., Walczak, M., 2009. "The influence of SiO2 and SiO2–TiO2 intermediate coatings on bond strength of titanium and Ti6Al4V alloy to dental porcelain", *Dental Materials*, Vol. 25, p. 1128–1135.

Carrado, A., Palkowski, H., 2010. "Microstructural and Mechanical Investigations on Porcelain-Fused-to-Metal in Multilayer System". *Advanced Engineering Materials, Vol.* 12(4), p. b122-b127.

Elsaka, S.E., Hamouda, I.M., Elewady Y.A., Abouelatta, O.B., Swain, M.V., 2010. "Influence of chromium interlayer on the adhesion of porcelain to machined titanium as determined by the strain energy release rate", *Journal of Dentistry*, Vol. 38(8), p. 648-654.

Gasik, M., 1998. Micromechanical modeling of functionally graded materials. *Computational Materials Science,*Vol. 13: 42-55.

Hautaniemi, J.A., 1995. "The effect of indium on porcelain bonding between porcelain and Au-Pd-In alloy", *Journal of Materials Science: Materials in Medicine*, Vol. 6, p. 46-50.

Henriques, B., Soares, D., Silva, F., "Optimization of bond strength between gold alloy and porcelain through a composite interlayer obtained by powder metallurgy", *Materials Science and Engineering A*, Vol. 528, p. 1415-1420.

Henriques, B., Gasik, M., Soares, D., Silva, F., 2012a. "Experimental evaluation of the bond strength between a CoCrMo dental alloy and porcelain through a composite metal-ceramic graded transition interlayer". *Journal of the Mechanical Behaviour of Biomedical Materials*, Vol. 13, p.206-214.

Henriques, B., Felix, S., Soares, D., Silva, F., 2012b. "Shear bond strength comparison between conventional porcelain fused to metal and new functionally graded dental restorations after thermal-mechanical cycling", *Journal of the Mechanical Behaviour of Biomedical Materials*, Vol.13, p. 194-205.

Henriques, B., Soares, D., Silva, F., 2012c. "Microstructure, hardness, corrosion resistance and porcelain shear bond strength comparison between cast and hot pressed CoCrMo alloy for metal-ceramic dental restorations", *Journal of Mechanical Behaviour of Biomedical Materials*, Vol. 12: 83-92.

Henriques, B., Soares, D., Silva, F., 2012d. Influence of preoxidation cycle on the bond strength of CoCrMo-porcelain dental composites. *Materials Science and Engineering C*, Vol. 32(8), p.2374-2380.

Henriques, B., Soares, D., Silva F., 2013. "Hot Pressing effect on the bond strength of a CoCrMoSi alloy to a dental porcelain", *Materials Science and Engineering C, Vol.* 33(1): 557-563.

Huang, M., Wang, R., Thompson, V., Rekow, D., Soboyejo, W.O., 2007. "Bioinspired design of dental multilayers", *Journal of Materials Science: Materials in Medicine*, Vol. 18, p. 57-64.

Maira, L., Padipatvuthikulb, P., 2010. "Variables related to materials and preparing for bond strength testing irrespective of the test protocol", *Dental materials*, Vol. 26, p. e17–e23

Niu, X., Rahbar, N., Farias, S., Soboyejo, W., 2009. "Bio-inspired design of dental multilayers: Experiments and model", *Journal of the Mechanical Behaviour of Biomedical Materials*, Vol. 2, p. 596-602.

Özcan, I., Uysal, H., 2005. "Effects of silicon coating on bond strength of two different titanium ceramic to titanium" *Dental Materials*, Vol. 21, p.773–779

Özcan, M., 2003. "Fracture reasons in ceramic-fused-to metal restorations", *Journal of Oral Rehabilitation*, Vol. 30, p. 265-269

Swain, M.V., 2009. "Unstable cracking (chipping) of veneering porcelain on all-ceramic dental crowns and fixed partial dentures", *Acta Biomaterialia*, Vol.5, p.1668–1677

ISSN 2176-5480

Suresh, S., 2001. “Graded materials for resistance to contact deformation and damage”, *Science,* Vol. 292, p. 2447-2451.

Wagner, W.C., Asgar, K., Bigelow, W.C., Flinn, R.A., 1993. Effect of interfacial variables on metal-porcelain bonding. *Journal of Biomedical Materials Research,* Vol. 27, p.531-537.

Yesil, Z.D., Karaoglanoglu, S., Akyıl, M.S., Seven, N., 2009. “Evaluation of the bond strength of different bonding agents to porcelain and metal alloy”. *International Journal of Adhesion & Adhesives,* Vol. 29, p. 32–35

Zhang, Y., Kim, J.W., 2009. “Graded structures for damage resistant and aesthetic all-ceramic restorations”, *Dental Materials,Vol.* 25, p.781–790.

7. RESPONSIBILITY NOTICE

The authors are the only responsible for the printed material included in this paper.