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Thermo-mechanical behaviour of ceramic metal brazed assemblies

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Abstract: This study investigates the effects of residual stresses developed during brazing on the performance of brazed ceramic metal joints. The thermal expansion gradient between ceramic and metallic materials leads to the development of such stresses during the cooling phase of the brazing process, which consequently reduce the strength of these composite structures. Here, the objective is to compare the failure behaviour of various assemblies observed during experimental tests and obtained through numerical simulations. In order to get a representation consistent with the physical mechanisms involved, these simulations must account for the brazing phase giving rise to residual stresses before applying in use solicitations to brazed joints. This paper focuses on the tensile strength of ceramic metal joints, for which two brittle failure modes (within the ceramic or at the interface) are observed during the experimental tests.

Keywords: ceramic–metal assembly, residual stresses, thermo-mechanical analysis, experimental tests, numerical simulations

1 INTRODUCTION

The association of materials with dissimilar thermal, mechanical, electronic, and chemical behaviour is especially interesting for high-technology applications (medical, aircraft, spatial, electronic or nuclear). Among joining processes, brazing is one of the most efficient and widespread to produce ceramic to metal assemblies. However, the difference between the thermal expansion coefficients (CTE) of the base materials leads to the development of residual stresses within the assemblies during the cooling phase of the brazing process. Such residual stresses clearly reduce the strength of the brazed joints and can cause cracking inside the ceramic and/or debonding at the interface, even during the brazing process itself [1–4].

The prediction of these failure mechanisms under any kind of solicitation is an important task for both practical and fundamental applications. Especially,

associated problems often limit the use of ceramics in spite of their attractive high-temperature properties. In this way, many analytical and numerical approaches have been proposed to estimate the residual stresses field induced by the brazing process [5–12].

This paper aims at determining through numerical simulations these residual stresses within standard tensile specimens of two different ceramic metal assemblies. Then, these features are taken into account to determine the damage development and the consecutive bond strength of such composite structures under standard tensile tests. In particular, this approach stands out by the introduction of the brittle behaviour of the ceramic metallization. Numerical and experimental results are finally compared and discussed.

2 EXPERIMENTAL AND NUMERICAL TECHNIQUES

The tensile test is generally used by the SCT Company for tension and vacuum testing of metallized ceramic seals. Indeed, this quite severe solicitation leads to

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a representative idea of their in-use performance. Accordingly, the objective is to compare in what follows the mechanical response of ceramic–metal assemblies (i.e. after the brazing process) during such a test obtained by experiments and simulations.

2.1 Materials and brazing conditions

In this case, the ceramic part of assemblies is an alumina with a purity of 97.7 per cent which exhibits linear elastic brittle behaviour. Its elastic properties, which do not depend on temperature for the temperature range of the brazing process (20–850 °C), have been identified through ultrasonic tests at room temperature (Table 1). If the failure of alumina material depends on the solicitation applied, the behaviour under tensile solicitations is purely brittle with a tensile strength around 250 MPa [13].

Two metals have been joined to the ceramic:

- (a) stainless steel AISI 304L;
- (b) Kovar (nickel–cobalt–ferrous alloy).

The filler metal (or braze alloy), which is introduced between base materials to produce the joint, is in both cases a silver–copper eutectic alloy. All these metallic materials exhibit elastoplastic behaviour depending on temperature. The evolution of their yield stress

Table 1 Mechanical and thermal properties of materials according to temperature: alumina (A), filler metal (FM), stainless steel (SS), and Kovar (K)

	Temperature, T (°C)						
	20	100	200	300	400	500	600
Young's modulus (GPa)							
A	330	330	330	330	330	330	330
FM [9]	79	79	79	79	79	79	79
SS	193	191	183	–	168	–	148
K	138	138	138	138	138	138	138
Poisson ratio							
A	0.26	0.26	0.26	0.26	0.26	0.26	0.26
FM [9]	0.33	0.33	0.33	0.33	0.33	0.33	0.33
SS	0.22	0.27	0.27	–	0.31	–	0.37
K	0.32	–	–	0.32	–	0.32	–
Yield stress (MPa)							
FM [9]	247	183	87	40	27	13	6
SS	247	243	169	148	136	133	125
K	345	–	–	223	–	217	–
Tensile strength (MPa)							
SS	973	774	667	635	633	589	517
K	517	–	–	462	–	385	–
Tensile failure strain							
SS	0.48	0.44	0.38	0.34	0.35	0.34	0.34
K	0.34	–	–	0.42	–	0.36	–
Coefficient of thermal expansion ($\times 10^{-6}/^{\circ}\text{C}$)							
A	–	6.2	7.4	7.8	8	8.1	8.2
FM	–	16.7	19.2	20.2	20.4	20.7	20.5
SS	–	18.4	20.3	20.9	21.3	21.4	21.7
K	–	5.9	5.2	5.1	5.1	6.2	7.8

with temperature given in Table 1 is obtained from reference [12]. Plasticity damage within these metallic parts has been neglected because tensile tests rather show a brittle mode of failure of specimens. Finally, note that CTE of base materials and filler metal according to temperature have been identified through dilatation test (Table 1).

If the attraction of the filler metal by the metallic part occurs naturally, it is necessary to apply formerly on the ceramic surface a low thickness metallization to increase wettability by the filler metal at the liquid state and allow capillary attraction (molybdenum process [14]). Assuming a brittle linear elastic behaviour, nano-indentation tests on the ceramic metallization part of brazed assemblies have been performed to determine the elastic properties of such material. The Young's modulus mean value is about 230 GPa [15] and the Poisson ratio is equal to 0.22. Besides, the tensile strength of this brittle material has been deduced from standard tensile specimens on ceramic-to-ceramic assemblies, leading about to 100 MPa.

The brazing process comprises heating the assembly of materials to a suitable temperature above the liquidus temperature of the filler metal. Then, the filler metal solidification that occurs during the cooling phase to room temperature leads to the final seal of the composite structure. Such procedure is realized in a vacuum atmosphere to protect the metallic part from oxidation and the heating (around 4 °C/min) and cooling (around 2 °C/min) rates are defined to protect the brittle parts from thermal shock. Base materials and filler metal are not submitted to any metallurgical transformation during the cooling phase; moreover, there is only thermal loading on the ceramic–metal assembly during the brazing process.

The microstructure of a ceramic–metal assembly after brazing is depicted in Fig. 1. Three domains:

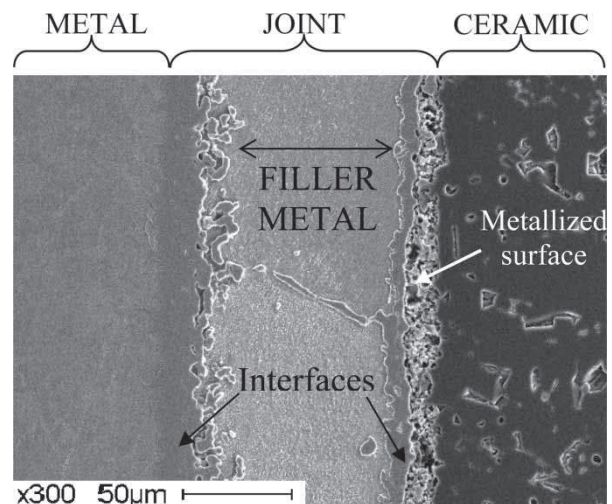


Fig. 1 Microstructure of the ceramic to metal brazed assembly

metal, joint, and ceramic are observed. The joint is composed of the filler metal, the metallized surface of the ceramic, and the interfaces generated at high temperature by diffusion. Finally, note that the figure shows the very tiny thickness of the metallization layer after brazing, with a value of about $15\ \mu\text{m}$. This value is close to the mean metallization thickness measured by the SCT Company after the molybdenum process and before the brazing process.

2.2 Tensile test

The experimental procedure follows the standard test method described by the American Society for Testing and Materials (ASTM F19-64) [16]. The specimen is composed of two identical washers of filler metal, which join a metal washer situated at the centre of the specimen with two identical ceramic components. The geometrical dimensions of the ceramic component are shown in Fig. 2, the metal washer thickness is equal to $0.3\ \text{mm}$ and the filler metal washer thickness $0.06\ \text{mm}$.

The gripping device used to apply the load (rate of $2\ \text{mm}/\text{min}$) allows a self-aligning of the test specimen with the axis of the test machine heads. The results dispersion for the identification of the tensile strength can be significantly reduced with this care.

2.3 Numerical model

Numerical simulations have been performed with the finite-element ABAQUS code. The model is defined in accordance with the standard specimen geometry defined in Fig. 2 and with metallization thickness

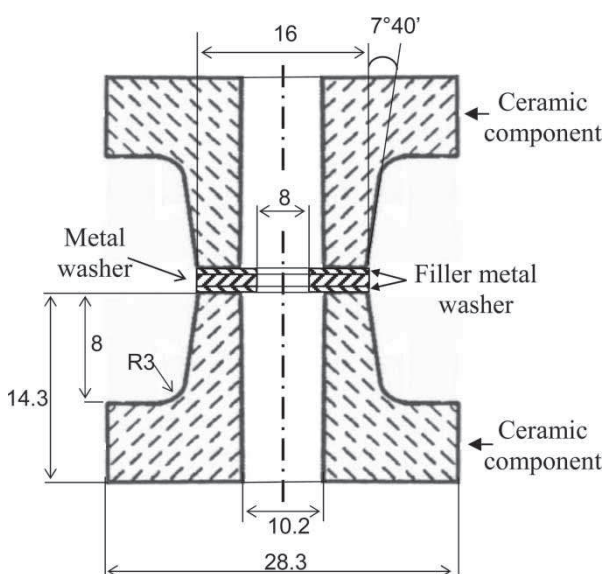


Fig. 2 Tensile specimen geometry of ceramic metal seal (distances in millimetre)

observed after brazing in Fig. 1. The simulation is divided into two parts: the first step is the numerical simulation of the brazing process and the second one is the simulation of the tensile test.

For both models:

1. The structure is two-dimensional axisymmetric and symmetric with the median plane.
2. The mesh of the assembly has been done with an eight-node quadratic axisymmetric quadrilateral element. According to the failure modes observed, the main residual stresses are essentially located near the interfaces and the free side surface, the element size is then taken lowest on these zones (Fig. 3). A converging study on maximum principal stresses has shown that 5210 nodes leads to an accurate estimation of the mechanical response after brazing process and during the tensile test.
3. The interfaces between the base materials, filler metal, and metallization are supposed to be perfect.

For the brazing process model:

1. The starting point of the simulation is the solidification of the filler metal during the cooling phase of the brazing process and the simulation ends when the ceramic-metal assembly temperature is equal to the room temperature.
2. There is no mechanical loading on the ceramic-metal assembly.
3. The thermal loading is the cooling cycle from the filler metal solidus temperature to room temperature with the uniform cooling hypothesis: the temperature of all the nodes is assimilated to the furnace temperature according to time.

For the tensile test model:

1. The stress state within the specimen is equal to the residual stress state given by the brazing process mode.
2. A negative vertical displacement is applied on the lower surface of the specimen (Fig. 3(b)).
3. As the ceramic part is only concerned by tensile solicitations, a simple Rankine criterion is used to detect crack initiation. This criterion states that a

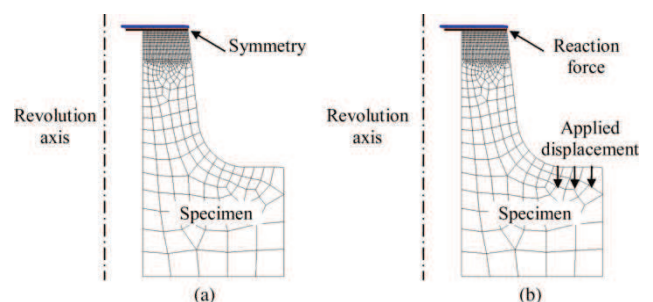


Fig. 3 Mesh of the specimen: (a) brazing process and (b) tensile test

crack forms when the maximum principal tensile stress exceeds the tensile strength of the alumina and leads to a loss of the material stiffness in the related principal direction.

- The metallization layer exhibits a negligible thickness in comparison with other materials; accordingly, its damage evolution is analysed with the cohesive zone formulation of ABAQUS introduced by Needleman [17]; especially, the brittle failure of such an interface occurs when the normal stress reaches the tensile strength of the considered material.

3 RESULTS

3.1 Experimental results

Two different types of assemblies depending on the choice of the metal have been tested, one with the stainless steel and the other one with the Kovar. Five specimens for each type of assembly were prepared and tested for this study. Figure 4 shows the evolution of the load applied on the specimen according to the displacement of the grip for each sample.

It is observed that the load increases slowly during the first part of the curve that represents the self-aligning of the grip and also the compression of the elastomer (split TFE-fluorocarbon) washer. Then a linear increase takes place until the brittle failure of the sample that occurs within the ceramic part

near the bond interface for the alumina–stainless steel specimens and within the ceramic metallization for the alumina–Kovar specimens (Fig. 5). Although the linear coefficient of the elastic response is quite the same for each specimen, the applied load that leads to the failure is quite different between the five samples. The average of these five values is equal to 10.7 kN for the alumina–Kovar assembly and 12.2 kN for the alumina–stainless steel assembly with a standard deviation of 1 kN in both cases. The tensile strength deviation of the brittle materials (alumina and metallization) could explain such deviation of the tensile strength measurements on the assemblies.

3.2 Numerical results

The stress evolution during the brazing process and during the tensile test within two types of specimen has been studied.

3.2.1 Brazing process

In this case, the base materials and the metallization remain in the elastic regime at the end of the brazing process. For the mechanical reliability of assemblies, it is essential to examine residual stresses developed within brittle materials. As an illustration, it is chosen to focus here on the ceramic part for which Fig. 6 shows the spatial distribution of the maximum principal stress (failure governed by the Rankine criterion). Especially, the maximum value is located for

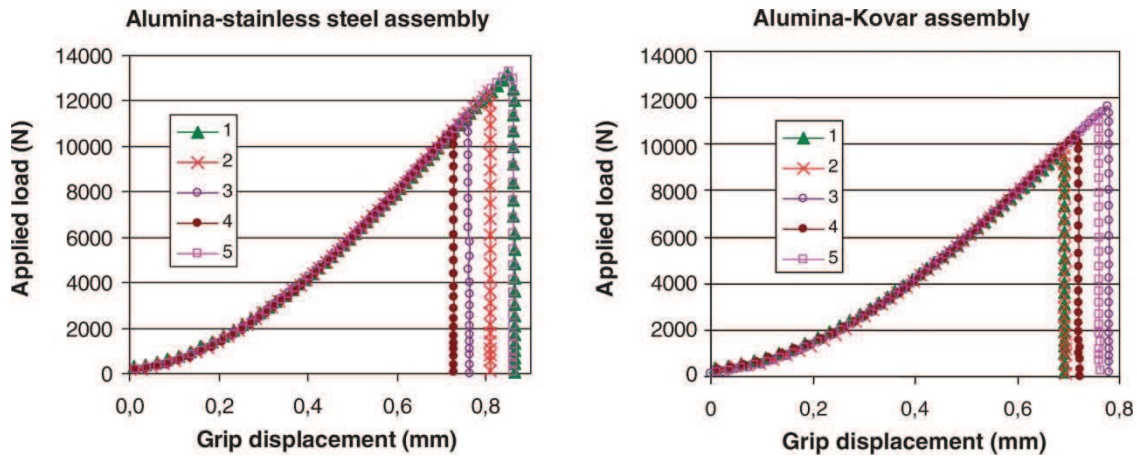


Fig. 4 Tensile tests on ceramic metal assemblies

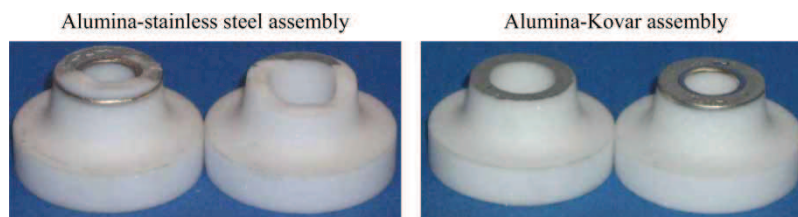


Fig. 5 Tensile specimens after failure

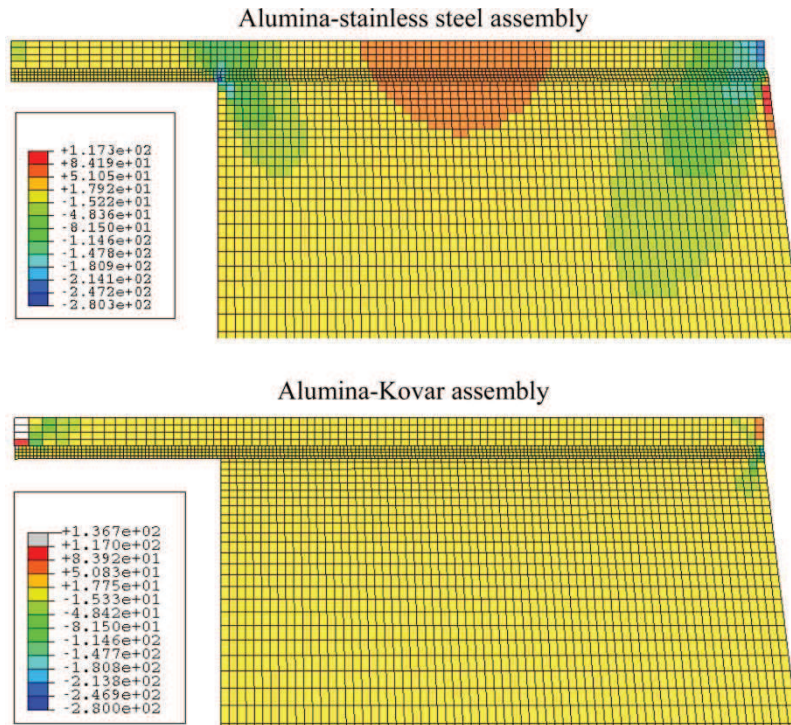


Fig. 6 Maximum principal stress distribution within the specimen after brazing

both types of assemblies on the radial free edge of the specimen near the ceramic filler metal interface. Nevertheless, its intensity during the brazing process clearly differs according to the metal of the assembly (Fig. 7): residual stresses are negligible for the alumina–Kovar assembly since CTE of base materials are very close; on the contrary, the thermal expansion of the stainless steel is much bigger than alumina and leads to greater values of residual stresses. Finally, note that the above conclusions can be done also

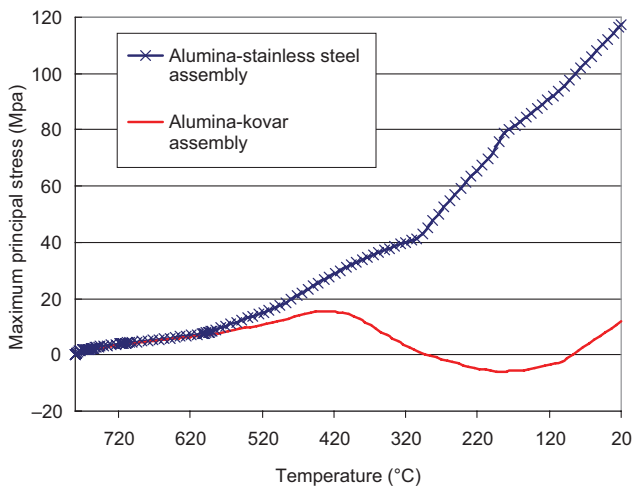


Fig. 7 Evolution of the maximum value of the maximum principal stress during the brazing process for each type of specimen

for the case of the metallization layer with a normal stress (which controls the failure in this zone) inferior to 60 MPa.

3.2.2 Tensile test

Figure 8 shows the failed elements of the specimen at the end of the tensile test (after global failure) for each kind of assemblies. A failed (respectively active) element is such that the failure criterion (Rankine for ceramic or normal stress for metallization) has been reached (respectively not reached). In accordance with experimental observations, numerical results clearly establish a marked difference in the failure mode depending on whether the coefficient of thermal expansion of the ceramic and metal are matched or not: for similar coefficients (alumina–Kovar), failure occurs within the metallization layer, whereas if coefficients are very much different (alumina–stainless steel), failure concerns the ceramic part close to the interface. The influence of residual stress near the ceramic metal interface explains this failure type variation. In this sense, it demonstrates the importance of considering residual stresses within numerical simulations and good predictive ability of the model to account for the assembly failure mode: without residual stresses, stainless steel assemblies would have behaved as Kovar assemblies, namely with a failure at the interface (within the metallization layer).

Concerning the quantitative analysis, Fig. 8 shows for the two types of assemblies the evolution of the

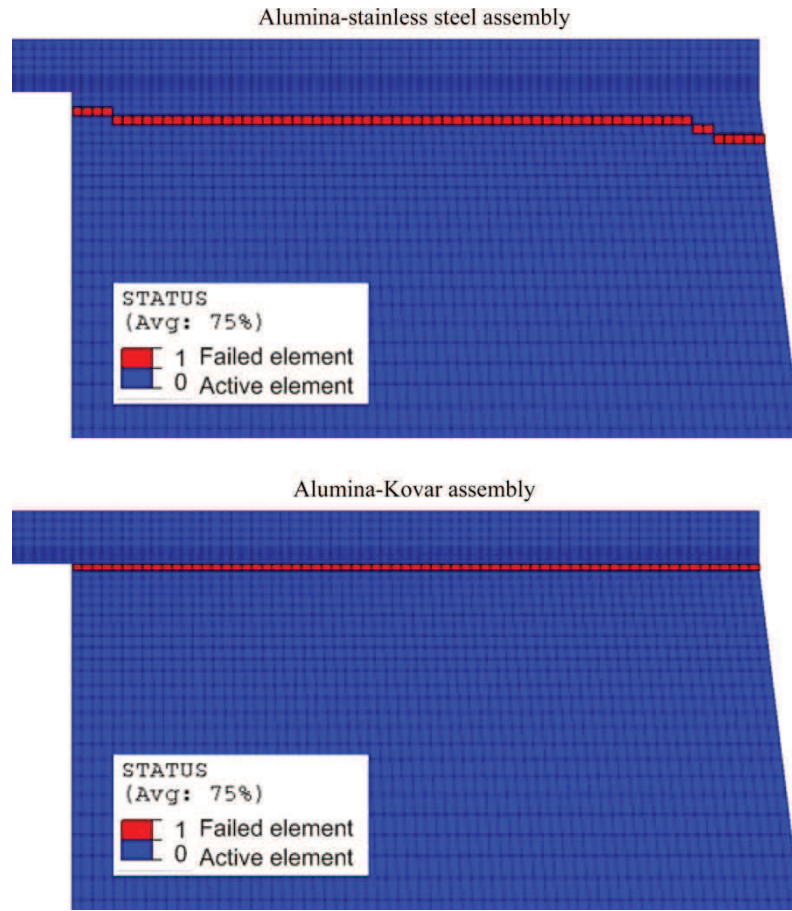


Fig. 8 Status elements for each type of assembly at the end of the tensile test

load measured at the central symmetry on the specimen according to the displacement applied at its bottom. The tensile strength obtained through simulation is then equal to 11.1 kN for the alumina–Kovar assembly and 10.2 kN for the alumina–stainless steel assembly, contrary to tests results which show stronger bounds for alumina–stainless steel. If the strength magnitude is correctly reproduced (around 11 kN), these slight differences between experiments and predictions could be explained by uncertainties on some materials properties given by literature data, especially for the strength of alumina and metallization. Moreover, such brittle materials often exhibit an important scatter in their properties that can be attributed due to a heterogeneous initial defect distribution. In this sense, the direct characterization of materials considered here and the coupling of the approach with reliability methods [18] would surely allow a more realistic estimation of the ceramic metal strength.

Finally, note in Fig. 9 that finite-element simulations lead to a non-linear response of the assembly before failure, contrary to the almost linear result in experiments (Fig. 4). It seems that such discrepancy comes from the difference in the application of the mechanical loading in both cases. In the numerical model, the

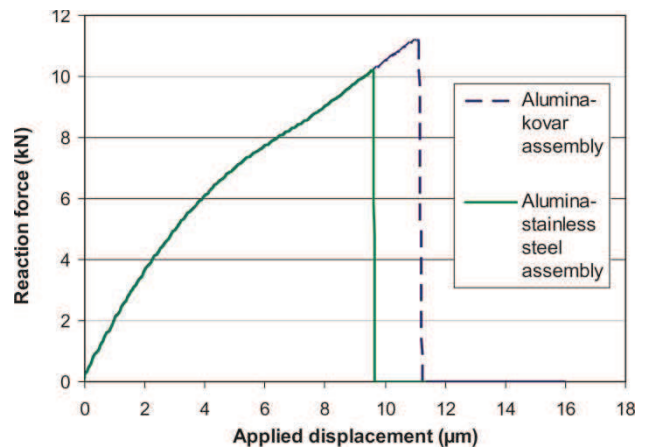


Fig. 9 Reaction force according to the applied displacement

solicitation is directly applied to the assembly and induces the development of some plastic deformation within metallic materials. On the contrary, it is difficult to avoid additional displacements in testing conditions due to the experimental device (especially due to the elastomer washer located between grip and specimen), as shown by the difference in displacement

magnitude in Fig. 4 (expressed in millimetres) and Fig. 9 (expressed in micrometres); in this way, plastic deformation during experimental tests may be hidden by such supplementary components.

4 CONCLUSION AND PERSPECTIVES

In view of the design of joints in engineering structures, the numerical model developed in this work represents an essential tool to study the mechanical behaviour and the reliability of ceramic metal brazed assemblies. Indeed, the brazing process model allows a consistent estimation of the residual stresses developed within such structures during the cooling phase, which remains crucial to get a realistic representation of their in-use behaviour.

Results obtained in this work are really promising since the numerical tool is able to correctly predict the experimental failure mode of various assemblies whether the coefficient of thermal expansion of metal and ceramic is matched or not. Nevertheless, if the tensile strength magnitude is reasonably reproduced, the quantitative agreement could be improved in the future by an experimental characterization of the properties of brittle materials considered here (alumina and metallization) including notably their potential scattering. Moreover, it may be interesting to complete this work by simulating the damage growth within the ceramic material and to compare this to the crack pattern generated during experimental tests.

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