APPROACHES ON SELF-HEALING OF AN INTERPENETRATING METAL CERAMIC COMPOSITE

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Abstract: *An interpenetrating metal ceramic composite (IMCC), manufactured via gas pressure infiltration of AlSi10Mg melt into a open porous Al_2O_3-preform, was investigated upon the ability of self-healing. A specific damage is introduced into the IMCC first. Then microstructural investigations are carried out at the damaged samples and for self-healing treated samples. The nature of the interpenetrating structure is used to heat the composite above the solidus temperature of the metallic phase and provide a shape stability by the ceramic phase to melt the metal and fill the cracks formed before. The investigation is systematically compared to the results of the undamaged samples as well as the pre-damaged samples without treatment for self-healing. The microstructural results show a change in crack geometry and therefore the possibility of self-healing. Nevertheless, open questions in process control as well as parameter-optimization require further research to achieve microstructural improvement of the healed samples above the performance of the pre-damaged ones.*

Keywords: Interpenetrating metal ceramic composite (IMCC); self-healing approaches; X-ray computed tomography (CT); microstructural characterization.

1. Introduction

In nowadays engineering success regarding to reduction of greenhouse gases and environmentally compatible implementations in mobility and transportation, light-weight materials for structural application play a key role. By combining light metals and ceramics, improved mechanical properties, as well as wear resistance for a long-lasting use of the composite can be achieved and the reached limits of light weight metals can be outperformed [1]. Metal matrix composites (MMC) reinforced with ceramic particles or fibers have already shown the high potential of this material group in an industrial application like e.g. in piston rings, brakes, engine blocks, connecting rods and propeller shafts [2]. By creating an interpenetrating composite, so that all phases are topologically continuously connected [3], additional advantages can be reached, as different authors stated from the early stage of the development up to latest investigations [1, 4].

In addition to the resource efficient design of components by using superior materials, sustainability, durability, and life cycle extension are becoming increasingly important in nowadays development and research. Based on the biological model of the self-healing ability of living organisms, a new field of possibilities was opened and reached increasing interest in the last decade [5]. The general definition of self-healing, as the ability of a material to heal itself without any external intervention automatically and autonomously is often not the case for manmade materials. Therefore, a differentiation in autonomous and non-autonomous self-healing is made by Gosh et al. [6]. Especially for polymers, many approaches have already been investigated and even autonomous self-healing approaches have shown success, making them highly interesting for industrial applications [7, 8]. To achieve healing in damaged metals, external triggers in form of thermal or electrical energy are required [9] to overcome their strong atomic bonding and the associated diffusion limitation in application environment [10].

In underaged alloys, healing of nanoscale flaws can be achieved by an increase of temperature which enables the diffusion of solute atoms into high stress areas such as cracks to form precipitates [5]. Another approach is the introduction of ceramic capsules containing a low-melting metal into the metallic matrix. In case of damage, the ceramic shell is broken and the metal contained flows into the crack upon heating to its' melting temperature [5].

The above mentioned self-healing concepts were designed and investigated to heal pure metals. Concerning composites, there are generally less ideas to incorporate self-healing due to the more complex structure. However, considering the high costs and complex manufacturing, in composites self-healing is even more beneficial than in conventional materials. Self-healing methods developed to heal composite materials are not to be confused with concepts that require the manufacturing of special composites to achieve healing. The latter include the above mentioned embedding or capsules containing a healing agent. The existing self-healing concepts designed to heal composites are limited to polymeric composites, e.g. to prevent delamination failure in CFRPs [11]. In contrast to that, no research was done with the aim to heal metal-based composites. Considering the big differences of mechanical and physical properties between MMCs and pure metals, the lack of knowledge in the field of self-healing metallic composites makes further investigations necessary.

In this study, first approaches on (self-)healing of an interpenetrating metal ceramic composite are investigated under the condition of a high temperature stable interpenetrating ceramic phase and a "low-melting", near eutectic light-weight aluminum alloy. To heal the internal damage, the influence of heating the metallic phase above the melting temperature is used as an external healing trigger with varying atmosphere, pressure and sample treatment.

The related research question is whether non-autonomous (self-)healing of the interpenetrating composite is possible with the tested approaches by melting the liquid aluminum alloy into the cracks of the sample and which parameters influence the self-healing process. The visibility of microstructural changes will be investigated in the following.

2. Materials and Methods

2.1. Material and sample preparation

To investigate the success of self-healing, an interpenetrating metal ceramic composite is used, based on a porous alumina preform with approximately 74 % open porosity. The preform is firstly evacuated up to a residual pressure of $6 \cdot 10^{-2}$ mbar. Then the metallic phase is infiltrated into the open porosity via gas pressure infiltration technique with an argon gas pressure of 60 bar. Details of the setup are described by the authors in Horny et al. [12]. 2D sections of the morphology are given in Figure 2.

Specimens were extracted from the infiltrated material with a diamond wire saw DWS 250 by Diamond WireTec GmbH&Co. KG and a hollow diamond drill. The cylindrical shaped specimens used for the self-healing approach had a mean diameter of 3.8 mm; a height of approx. 8 mm and parallel polished end faces according to DIN 50134 [13] (see Figure 1 a). For the healing approach, a parallelepiped with dimensions of 30 x 23 x 5 mm³ was cut, surface grinded and finally drilled in the surface extension with a hollow diamond drill with a diameter of 18 mm (see Figure 1 b).

2.2. Experimental procedure

Mechanical characterization

The specimens were systematically pre-damaged via compression testing, carried out in the universal testing machine ZwickRoell Z1464 with a nominal strain rate of 0.06 1/s. The sample strain was captured with a videoXtens optical strain measurement system, also by ZwickRoell.

Microstructural characterization and evaluation

For microstructural characterization, an X-ray computed tomograph (Phoenix Nanotom M) by General Electrics was used. The software components Phoenix data sx2 acquisition and Phoenix data sx2 reconstruction were used to process the data and reconstruct a 3D image of the sample, each also by GE Sensing & Inspection Technologies GmbH. The focus-object distance was 10.5 and 24 mm and the focus-detector distance 300 and 480 mm with a resulting voxel size of 3.5 and 5 µm³ for the self-healing and healing approach, respectively. The beam source was powered with 80 kV and 180 µA. Each sample was scanned three times: In the undamaged state, right after pre-damaging and after the healing attempt.

Evaluation of the CT data was carried out with the Avizo® software by ThermoFisher Scientific. The aim was to compare the CT data of a sample in pre-damaged and healed state concerning changes in crack volume. This was achieved by following two major procedures. Avizo®-specific operations are given in italic letters in the following:

The first procedure was required to align the 3D-data in the coordinate system of the software and therefore ensure comparability of both data sets. By applying *Register Images,* one of the data sets was visually realigned with the second data set serving as reference. The module *Resample Transformed Image* generated a new data set corresponding to this alignment.

The second procedure was separately applied on each data set to obtain the volume fraction values of the crack volumes. First, an *Image Stack Processing* workflow including an *AutoThresholding* module as well as subsequent operations was applied to extract the darker grey values, which are representative for the cracks. To remove artifacts and pores from the resulting data, first *Labeling* and then *Filter by Measure Range* were applied, using the shape function *Shape_VA3d*. The value 1 of this function corresponds to a perfect sphere, whereas higher values represent a less compact object. The resulting crack volume was visualized using *VolumeRendering* and its value was calculated with the module *VolumeFraction.*

Non-autonomous (self-)healing treatment

Temperature induced self-healing was carried out in a conventional furnace by Nabertherm. A pre-damaged sample was heated at 10 K/min to a maximum temperature of 650 °C, hold for 5 min there and then cooled to room temperature with the same cooling rate.

For the pressure-assisted (self-)healing approaches, a gas pressure infiltration setup described by the authors in Horny et al. [12] was used, which enabled the variation of atmosphere, pressure and temperature. The process was modified, as given in Figure 1 c).

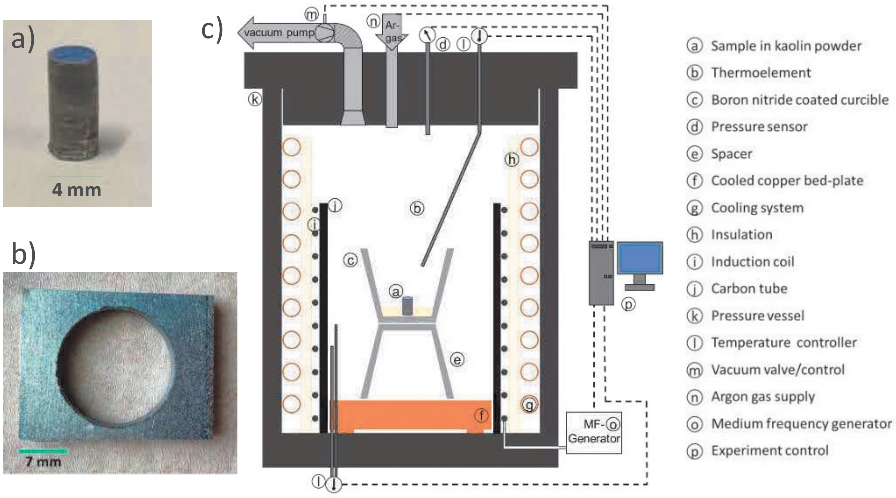

Figure 1. a) Sample geometry for self-healing approaches. b) Sample geometry for alloy supported healing. c) Gas pressure infiltration setup for pressure-assisted (self-)healing experiments.

The sample was put in a crucible with calcined kaolin powder as support material. For accurate temperature control, the thermocouple was positioned as close to the sample as possible. Like during manufacturing of the composite, vacuum of 0.06 mbar was used during heating with a heating rate of 10 K/min. For self-healing approaches under gas atmosphere, a pre-pressure of 5 bar was applied at 550 °C, before the solidus temperature of the alloy was exceeded. Heating then was continued above the liquidus temperature of the alloy, up to 700 °C. Additional gas pressure up to 60 bar was then brought onto the sample. In the end, cooling was carried out, maintaining the atmosphere condition from maximum temperature. For self-healing approaches under vacuum, no pressure was brought up to the sample and the vacuum was held for the whole process.

For the healing approach, additional alloy was put together with the sample into the empty crucible prior to applying the above-described procedure under argon atmosphere.

3. Results

3.1. Temperature induced self-healing approach

When the sample was removed from the setup, an accumulation of alloy on the surface (cf. Figure 2 b, right) could be detected, indicating that the alloy had melted and flowed out of the sample. Quantitative analysis of the corresponding CT-images using the *Image Stack Processing* Workflow revealed a 2.5-fold increase in porosity after the temperature induced self-healing. In addition, the rearrangement of porosity was observed: Some pores were already present in the microstructure before but were no longer visible at this location after the self-healing tests. An example for the pore rearrangement is marked with a blue arrow in Figure 2. The cracks remained mostly unchanged. Figure 2 shows slight changes in the crack thickness that could be found at some locations of the microstructure. The red arrow marks a widening of the crack due to self-healing, while the green arrow points on a crack part that became narrower

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with self-healing. These changes were only observed for cracks at phase boundaries or in metal areas whereas cracks in the ceramic phase remained unchanged by the self-healing experiments.

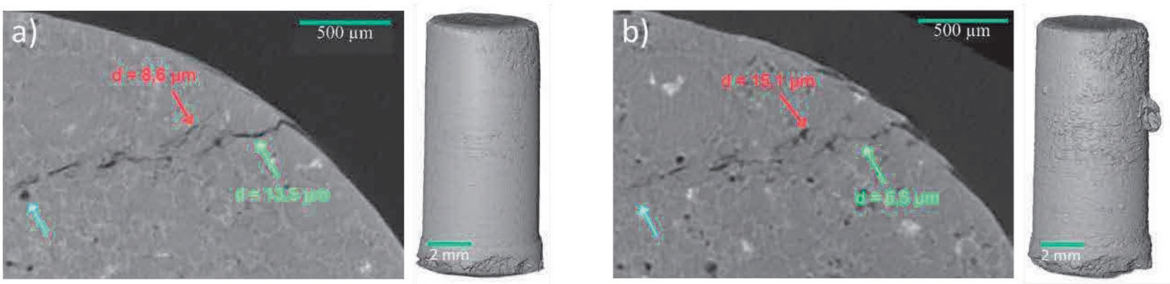

Figure 2. CT-images of a crack and the corresponding samples: a) before and b) after self-healing. The blue arrows pointing on a pore disappearing after self-healing, the red and green arrows marking crack positions that became bigger/smaller due to self-healing.

3.2. Temperature and Pressure induced self-healing approaches

To prevent the alloy from flowing out, the self-healing experiments were extended by additionally applying gas pressure on the sample during self-healing. Different experiments were carried out under vacuum (0.06 mbar), nitrogen or argon atmosphere (40 – 60 bar) as well as varying target temperatures (600 – 700 °C).

Although the self-healing parameters were varied, each sample showed similar microstructural changes analogous to the temperature induced self-healing results in Figure 2.

3.3. Temperature, pressure, and alloy-supported healing approach

After the alloy-supported healing experiments, the sample surface was completely covered in aluminum, indicating full liquefaction of the alloy. The CT-Images shown in Figure 3 reveal that the additional alloy penetrated the crack, resulting in a decrease of crack thickness. Nevertheless, some parts of the crack remained porous after healing. Figure 3 shows the 3D-volume of the crack before and after healing, clearly indicating a strong decrease in crack volume. The quantitative analysis revealed a crack volume of 2.8 vol% before healing, whereas the healed sample had a crack volume of only 0.6 vol%.

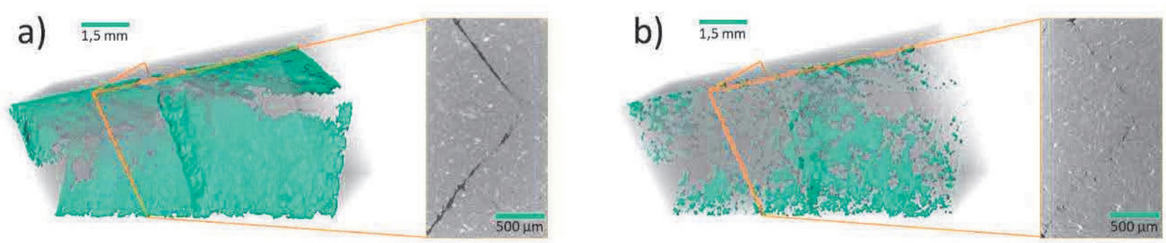

Figure 3. Crack volumes and corresponding CT-images of a) pre-damaged sample with a crack volume of 2.8 % and b) sample healed with additional alloy, showing a crack volume of 0.6 %.

4. Discussion

Firstly, the definition of (self-)healing should be discussed considering its applicability on IMCCs. The differentiation in autonomous and non-autonomous self-healing according to Gosh et al. [6] cannot be transferred to IMCCs because activation by an external trigger is always required for the metallic phase to perform healing. Instead, a more extensive distinction can be made. According to its definition, the term self-healing includes the application of external energy, but not support by additional alloy. Keeping that in mind, the alloy-supported experiments described above can be defined as healing experiments, whereas experiments without additional alloy count to self-healing approaches.

From the observations of the experiments, it can be deduced that IMCCs have a high potential for self-healing due to their interconnected microstructure, which enables to heal cracks both in the metallic and ceramic phase. Blaiszik et al. [14] presented a general formula for the quantitative determination of the healing success, which can be applied to the IMCC:

$$\eta = \frac{f_{healed} - f_{damaged}}{f_{undamaged} - f_{damaged}} \qquad (1)$$

The variable f stands for any property of the material that is changed by the healing attempt. The result η is a percentage value indicating to what extent the material property could be restored by healing. Considering the above presented results of changes in crack thickness, the healing success η for the self-healing approach is 51.8 % for the crack marked with a green arrow in Figure 2. However, as already mentioned, most cracks did not change with the self-healing approach or even became bigger. For example, the widening of the crack marked with the red arrow in Figure 2 results in a negative healing success η of –75.6 %. In contrast to that, the healing approach (see chapter 3.3.) with additional alloy resulted in evenly distributed healing with a healing success η of 78.6 % concerning crack volume. The reason for the increased porosity and little crack changes in the self-healed samples is the outflow of alloy which was prevented in the healing experiments by the additional alloy.

To explain the different observations, it is first necessary to look at the underlying physical parameters. Generally, positive capillary pressure is required for the metal melt to autonomously penetrate cracks [15]. The value of capillary pressure p directly depends on the surface tension σ_L of the metallic melt, contact angle θ between melt and matrix and the capillary radius r, which is the crack diameter in this case [16].

$$p = \frac{2\sigma_L \cdot \cos\theta}{r} \qquad (2)$$

Aluminum alloys and alumina show non-wetting behavior with a contact angle θ between 90 and 180 degrees [16], resulting in a negative capillary pressure. The wetting is inhibited by the naturally existing oxide layer on the aluminum surface [17], which transforms and thus possesses even higher hardness above temperatures of 500 °C [18]. Experiments showed a decrease of contact angle down to 90 ° with decreasing oxide layer thickness, which can be achieved by high vacuum atmospheres, inhibiting the oxide layer growth [17]. Considering equation 1, the capillary pressure can also be altered by decreasing the surface tension of the metal melt, e.g. by using nitrogen atmosphere above 850 °C, resulting in the formation of AlN [16]. Nevertheless, the capillary pressure will always be negative for this material system, making an external pressure required to exceed the capillary resistance of the cracks [16].

This is confirmed by the unsuccessful temperature induced healing attempt, resulting in an outflow of alloy instead of efficient self-healing. However, in contrast to theory, the temperature and pressure induced experiments with varying atmospheres and temperatures also showed no self-healing improvements, indicating that the effect of the gas pressure was not sufficient. As a result, the driving forces for the melt to leave the sample exceed. A scheme of all relevant forces in the (self-)healing experiments is shown in Figure 4.

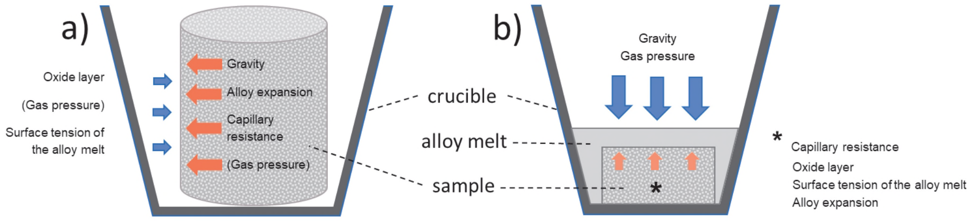

Figure 4. Schematic illustration of all relevant forces acting during the self-healing attempts a) and the healing experiment b).

During the healing experiments, the force mismatch in Figure 4 a) could be changed towards Figure 4 b) as the additional aluminum completely filled the crucible, making the gas pressure act on the upper face of the melt only. As a result, successful infiltration of cracks took place with a healing success that exceeds literature values. Srivastava & Gupta [19] carried out self-healing experiments resulting in a lower healing success of 62.8 %, even though their self-healing approach, a combination of shape-memory alloys and metal melt as healing material, usually is the most effective in the field of self-healing metal concepts. This proves that the healing approach with additional alloy is highly promising. To achieve similar results for the self-healing experiments, further parameter investigation must be carried out.

5. Summary and outlook

In this study, a systematic investigation of approaches on self-healing of an interpenetrating metal ceramic composite was carried out. The general possibility of self-healing was demonstrated, as the metallic phase could be activated by non-autonomous self-healing with thermal activation. The influence of varying the atmosphere and pressure was investigated and in a final step, the addition of aluminum alloy was tested, resulting in a healing success of 78.6 %. In this context, the concept of self-healing was discussed at IMCCs and a distinction was drawn between “healing” and possible “self-healing” concepts. Existing challenges were seen in the surface oxidation of the metallic phase, as well as in the outflow of the liquid metal during the self-healing process. For cracks with surface contact, surface oxidation counteracts the self-healing potential. The outflow of the metallic phase increases the inner porosity and potentially weakens the composites mechanical strength. A further understanding of the influence of surface tension, capillary effects, process parameters and phenomena occurring during healing is required to improve the self-healing potential of IMCCs.

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