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Stress concentrations and bonding strength in encapsulation-based self-healing materials

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ABSTRACT

In encapsulation-based self-healing materials, filled capsules with the healing agent are embedded in a matrix. But between the capsule and the matrix an interface always exists. The strength of the interface between both components is proved to play a crucial role in the correct working of the self-healing. This paper analyzes numerically the role of the interface bonding strength and the stress concentration around a cylindrical capsule embedded in a homogeneous, isotropic and elastic matrix, which undergoes a uniform and uniaxial far-field stress. Geometry and load condition make it to use a two-dimensional plane strain model. This model is based on a combination of the classical Finite Element Method and cohesive surface techniques implemented in the commercial code Abaqus. Two types of interfaces have been studied: perfect and imperfect bonding. A detailed validation of the model against analytical expressions has been conducted in order to guarantee a correct behavior of the interface elements. The influence of the elastic mismatch between the capsule and the matrix on the stress concentrations has been assessed, as well as the possibility of using capsules with different thicknesses. In order to prevent debonding, a study to provide the optimum combination of material elasticities, capsule thicknesses and bonding strength has been performed. The initiation and propagation of the interfacial crack have been also fully addressed. In that direction, once a crack is initiated, the role of the elastic mismatch and the capsule thickness is also assessed. This model can predict the suitability of the mechanical performance of the interface and whose role is typically underestimated during the preparation process of encapsulated self-healing materials.

1. Introduction

Self-healing materials are receiving a lot of attention nowadays due to their wide range of advantages, for example higher structural reliability under loads, prolongation of the service life, reduction of costs due to less maintenance, less material replacement, among others [1–4].

This work focuses on the so-called encapsulation-based self-healing materials. This family of self-healing materials are produced introducing some small fluid-filled capsules in a matrix. These capsules need to be designed with the adequate shape and material in such a way that they should release the internal fluid when a crack in the matrix reaches the capsule. In an ideal situation, the stress concentrated at the crack tip is fully transferred to the wall of the capsule. Such a high stress undergone by the capsule eventually leads to its sudden breakage. Then, the fluid inside the capsule is released. Depending on the technological application and the size of the crack, the action of the filling of the crack could be driven by the presence of mechanisms like, for instance, capillarity. Finally, if the agent is a substance that cures after certain time, then the crack is finally sealed, and the material is internally repaired, or at least regains partially its original strength.

In connection with the aforementioned process, the aim of this paper is to understand the role that the interface plays during the initial stage of this strategy. When a capsule is embedded in a matrix, an interface always exists between both elements, but the suitability and characterization of the mechanical performance of this interface is typically underestimated during the preparation process. However, an efficient self-healing process depends on the leakage of the healing agent once a crack reaches a capsule, and this is true only as long as the capsule breaks. If the interface is not able to resist the load transfer from the matrix, the capsule simply separates from the matrix. This means that the incoming

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crack would just become an interface crack going around the capsule and continuing its path. This process would leave the capsule intact, and the material would not heal successfully.

The target of the proposed work is twofold: (i) to understand the stress concentration in a solid when its material constituents are not damaged yet, and (ii) to understand how and when an interfacial crack would be initiated because of a lack of bonding. The former case (i) is interesting precisely to find out which constituent would fail first under loading. Then, a question that the proposed model may clarify is to determine in which conditions the capsule or the matrix would become the precursors of internal damage of the solid. In other words, from the design point of view, it would be very interesting to know which combination of material elasticity, capsule size and strength will prevent a premature material failure. Another forthcoming application of this model deals with the interaction between the field stress around the capsule and the field stress at the tip of an arriving crack. As the interest is to break the capsule when a crack approaches it, it is very relevant to know whether the choice of materials will enhance the concentration of stress in the capsule, or conversely, it will prevent the breakage of the capsule because of an unexpected deflection of the crack. Regarding the target of settling the conditions of initiation of interfacial cracks, this proposal may help to provide some valuable clues to experimentalists to get the best self-healing configuration, namely, prior to selecting the materials and start with the fabrication process.

A matrix containing a thin-walled capsule may resemble a solid with a hole. In that sense, the mechanical problem of a plate with a hole has been formerly analyzed theoretically in 2D [5] and in 3D [6]. However, the presence of a capsule, and more particularly, the presence of the interface, complicates the theoretical treatment. A closer problem configuration to this case can be found in the framework of syntactic foams, which are a type of reinforced composite that consists of a matrix that contains hollow microspheres, commonly called microballoons [7]. These foams are technologically very relevant in marine and aerospace structural applications due to their low material density and high specific strength [8]. However, the properties might be affected by the integrity of the interface between the microballoon and the matrix [9]. In that direction, there are some recent works aimed at analyzing the effect of debonding of a matrix-embedded microballoon on the tensile response of the foam. In Ref. [10] an experimental work to assess the debonding of hollow particles embedded in an elastic matrix was conducted. Likewise, a semi-analytical approach based on the Galerkin method was also addressed by Tagliavia et al. [11,12], where two pre-existent interfacial cracks were located at the poles of a spherical microballoon. They studied the influence of the thickness of the microballoons on the overall failure mechanism in a vinyl-ester glass syntactic foam used in marine applications. In Ref. [13], the importance of the bonding strength in a similar system under compression was also treated, where the influence of the microballoon's buckling on the failure mechanism was assessed. Other very important design parameters like the capsule thickness and the combined effect between the bonding strength and clustering have also recently been analyzed [14,15].

On the other hand, in the scenario of having a solid inclusion embedded in the matrix, there is the well-known family of fiber reinforced composites. The problem of debonding in these composites is widely treated in the literature, in particular when an interfacial arc crack is initiated between the fiber and the matrix under different loading modes [16–25]. However, although that problem might share some conceptual characteristics with the problem presented here, the truth is that the flexural capabilities of the capsule incorporate additional complexities [13].

This paper treats the problem of a cylindrical capsule embedded in an infinity matrix. This work contributes with a simulation model that makes it possible to predict the crack initiation at the interface as well as its propagation. In that sense, not only the effect of the elastic and geometric ratios of this heterogeneous system is studied, but also an insight into the stress concentration profiles under perfect and imperfect debonding is also achieved.

A premature debonding of cylindrical capsules in these materials is a phenomenon that is merely inferred by experimentalists (for example, see Ref. [26]). In fact, once the experimental test is finished, it is often too late to find out how and when debonding took place, and more importantly, to find out the real consequences derived by a lack of capsule bonding. For such a reason, the aim of this paper is to provide in advance an extra support to experimentalists to take into account this plausible scenario and how to prevent debonding. To our best knowledge, these issues involving cylindrical capsules are hardly found in the literature, in particular for self-healing applications, neither experimentally nor theoretically. Regarding the methodology proposed in this work, for the sake of the effectiveness and further extension of the problem, a pure numerical treatment has been carried out taking into account that an experimental approach is problematic.

This paper is organized as follows. Section 2 presents a complete description of the model, where the setup and the interaction capsule-matrix is exposed. Likewise, this section also deals with a discussion of the material properties used in the paper and the important part concerning model validation. Section 3 is a block concerning the results assuming a perfect bonding. This section shows the effect of the elastic mismatch and the capsule thickness, as well as a discussion about the applicability of the model. Section 4 aims to reveal the effect the debonding on the mechanical response. Two possibilities of debonding mechanisms and their associated effects are presented. Here the issue of the initiation and tracking of an interfacial crack is also explained. The last part of this section is devoted to a discussion of the applicability of the results to foresee the debonding mechanism under certain configurations of the system. The Section 5 highlights the main features and provides some general hints on possible improvements and further perspectives of this work.

2. Model

2.1. Setup, regions and mesh

The specimen geometry consists in a square plate of length $2L$ containing a central hole of outer radius R_c that houses a capsule with wall thickness t_c. Except for the presence of the capsule, this configuration may resemble the classical problem of a holed plate under a remote tensile stress [27]. Therefore, taking advantage of the symmetry, this analysis has been performed on a quarter geometry, as it is shown in Fig. 1a. The top face of the plate undergoes a uniform far-field stress σ_∞ parallel to the y-axis. The right external boundary is a free-face whilst the left and bottom internal boundaries are symmetry plane faces. For validation purposes with the theoretical case of an infinite plate with a hole (see next section), a large enough domain in comparison with the capsule radius has been set. Specifically, the half-length of the plate was set at $L = 50$ mm and the radius at $R_c = 1.5$ mm. Unless stated otherwise, the capsule wall thickness is $t_c = 0.175$ mm. These sizes for the capsule have been chosen because they correspond with the true dimensions involved in lab tests with self-healing specimens [28]. Fig. 1b shows a schematic detail of the five regions that compose the model. There are two solid regions, the matrix (gray) and the capsule (yellow), and three surface regions: the internal surface capsule (blue), the external surface capsule (red) and internal surface matrix (black).