Algorithm-driven design of fracture resistant composite materials realized through additive manufacturing

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Abstract
Fracture, the breakdown of materials as cracks advance, is one of the most intriguing materials phenomena; it can happen even to very tough biological tissues including tendons, skin, bone, and teeth, materials whose critical physiological functions can be compromised by structural irregularities. It has been suggested that creating composites by mixing heterogeneous constituents of contrasting material properties can yield designs that can better adapt to stress concentration, leading to synthetic materials with higher toughness than their constituents. Here, an optimization algorithm is used to assess material fracture resistance in the presence of a crack. The analysis is further extended through experiments that involve the use of additive manufacturing. Optimal solutions are composed solely of soft and stiff material elements, and are compared to various benchmarks. Multi-material three-dimensional-printing (3D-printing) is used to create material samples. Experimental results and mechanical testing show that an algorithmic design coupled with 3D-printing technology can generate morphologies of composites more than 20 times tougher than the stiffest base material, and more than twice as strong as the strongest base material. Direct comparison of strain fields around cracks shows excellent agreement between simulation and experiment. The results suggest that the systematic use of microstructure optimization to generate enhanced fracture resistance constitutes a new materials design paradigm.

1. Introduction
One tissue engineering strategy to treat bone loss due to impact or disease is designing reliable scaffolds that are made of synthetic materials with desired mechanical functions and properties [1,2]. Current classes of scaffolds use materials such as hydroxyapatite, alumina, and calcium phosphates, all of which have low resistance to fracture and high brittleness [3,4]. Cracks that originate in these structures can quickly propagate and cause catastrophic material failure due to the high stress and strain concentration around the crack tip. Using homogeneous synthetic materials to make the entire product may not be efficient, as it requires materials to be tougher than bone, which is difficult to achieve. It has been suggested that by creating a heterogeneous material by mixing different materials of inferior material properties, these designs can better adapt to the stress concentration, leading to a material with higher toughness than its constituents [5,6]. Therefore, understanding methods to design these novel heterogeneous materials that can better resist fracture compared to their constituents offers new opportunities to efficiently create fracture-resistant composites for many biomedical applications [1,2,4,7].

Fracture is one of the most intriguing materials phenomena and has long been the subject of extensive studies in applied mechanics. Indeed, “materials by design” is an emerging trend that allows researchers to tune materials according to function. Optimization of material form and composition is an excellent pathway to designing new structures. Specifically, topology optimization has evolved into an efficient design tool to meet objectives such as compliance, stiffness, and thermal expansion coefficient [8–17]. Due to the complexity of the solutions achieved, topology optimization in the past has often been limited to theoretical work [11,15,18]. Recent advances in additive manufacturing now allow users to physically create specimens with the optimized designs [19–28]. However, little research thus far has focused on fracture properties, especially on composite toughness optimization in the presence of cracks. Biological composites, such as nacre and bone, tend to have an array of toughening mechanisms at different length scales and have been shown to have superior toughness compared to their own con-
stituents, which has led to a surge in biomimetic composites using various techniques such as freeze casting, biomineralization, extrusion, and additive manufacturing [29–32]. Toughness amplification relying on biomimicry has been demonstrated in the literature, but these studies have not further optimized beyond their baseline biomimetic design, and have not rigorously explored algorithm-driven solutions [30,31,33–39].

The hypothesis formulated here is that by redistributing soft and stiff materials, the composite toughness can be amplified using an algorithm-driven approach. In a homogeneous material, stress concentrates around a crack, leading to failure of the structure before distant regions in the material have been subjected to high loads. By redistributing soft and stiff materials, the stress field can be altered to mitigate the stress concentration at the crack tip. Here, an optimization algorithm for toughness developed by the authors is validated with experiments using additive manufacturing to test this hypothesis and provide proof of concept [40]. The model material problem has an edge crack with tensile loading under mode I failure. For this topology optimization problem, a modified greedy algorithm [41,42] is used that searches for an optimum distribution of soft and stiff material under the constraint of constant volume fraction. The materials are chosen to have equal toughness in order to study geometric, not material, effects of soft and stiff material distribution on composite toughness. The algorithm works by picking an initial random geometry and then switching all elements one by one and checking if the toughness improves. If toughness does not improve after switching each element in every iteration, the simulation terminates. The simulation technique is summarized in the simulation methods section.

This paper is organized as follows. Section 2 outlines the experimental method used to validate the in silico algorithm, ranging from additive manufacturing to mechanical testing. Section 3 describes the experimental results, comparing the performance of the 3D-printed optimum designs to those of the benchmark designs. Finally, the conclusion is used to summarize the study findings, discuss limitations, and offer suggestions for future research.

2. Material and methods

The research reported here employed simulation, additive manufacturing, and mechanical testing, sequentially. Additive manufacturing was used to create the geometries generated by the preceding simulation. Mechanical testing of the 3D-printed geometries was performed to compare and analyze results from simulation and experiments. This procedure of 3D-printing and testing was adapted from reference [31].

2.1. Simulation

The simulation step optimizes a random initial geometry with soft and stiff building blocks, which is described in further detail in reference [40] and briefly summarized in this section. The model problem has geometry with an edge crack with x-direction tensile loading under mode I failure. The optimization method used is a modified greedy algorithm and receives as an input a random initial population made up of soft and stiff elements. From this initial pattern, the objective function is evaluated in order to select the next-best solution for the system. At each iteration, one element at a time is replaced by its opposite material; for example, if the element at hand is a stiff material, it will be switched to a soft material. This procedure continues for all elements in the system, and the corresponding objective function with constraint is computed for each switch. From the collection of switches, the switch that attains the highest objective is kept and used for the next iteration. If the kept objective value is higher than its previous iteration, which for the first iteration is compared to the random initial configuration, it feeds back to replacing each element one at a time. This loop continues until the geometry has converged. Convergence occurs when the new objective value is not higher than its predecessors, which prompts the program to exit and output the final geometry as its final iteration (Fig. 1(a)).

The objective function seeks to optimize the toughness modulus. The toughness modulus is defined as the area under the stress–strain curve for a material; it can be understood as the energy needed to fracture a system. The objective function is solved for using a finite element method, with four node elements, each with two degrees of freedom. The finite element method is used to obtain the stress and strain fields from displacements for different material geometries. Elastic stress mismatch does not pose a problem here because of the sufficiently low stiffness ratio used for soft and stiff elements. A linear elastic model is used because it is assumed that the dominating mechanisms are controlled by linear elastic mechanisms, as suggested by experimental evidence in additive manufactured materials [31,43]. A mesh size of 40 × 40 is used and the symmetry of the problem reduces the number of elements needed to optimize by twofold. The failure criterion used is strain at the crack tip; once the crack tip strain reaches the failure value for the element type, the objective function is determined. Material parameters referenced in [40] will be used in this study to obtain optimum solutions that will be compared to various benchmark solutions.

A schematic of how the toughness ratio between the current geometry and initial geometry (T/T0) increases with iteration number is shown in Fig. 1(b). After every iteration, the toughness increases from that of the initial random geometry, through several intermediate geometries, to the final solution. In addition, the stiffness is overlaid in Fig. 1(b) to show that no sacrifice in stiffness is made in the process of toughness optimization.

2.2. Benchmark and optimized geometries

The base material benchmark geometries used are 100% volume fractions of stiff and soft material, respectively. The benchmark geometries with 20% volume fraction soft material are random (Rnd), minimum toughness (Min-T), and an arbitrary design (MIT). The combination of base materials at 20% volume fraction soft material forms a random geometry. From a random geometry, the other geometries can be generated. The minimum toughness (Min-T) design was created from an initial random geometry with the objective of minimizing toughness in the simulation. The arbitrary geometry (MIT) was created by selectively forming the soft material into a desired design, such as the MIT logo. Similarly, the two optimized geometries (Opt-1, Opt-2) were created from two different initial random geometries with the objective of maximizing toughness using the proposed optimization approach. Displacement boundary conditions was used, with a crack size of 20% of the sample length. All the geometries are shown in Fig. 2(a).

2.3. 3D-printing

The heterogeneity in material composition in the optimized geometries makes it difficult to create them with traditional subtractive manufacturing. Hence, this study utilized recent advances in additive manufacturing, particularly multi-material polyjet printing. The benchmark and optimized geometries from simulation was first converted into stereolithography (.stl) files. A separate file was created for each material phase of the generated geometries. The .stl files were used in conjunction with a Stratasys Object Connex500 multi-material 3D-printer. The two bulk materials that were used are proprietary photopolymers, VeroMagenta and TangoBlackplus, which have contrasting material
properties. The stiffness of the Veromagenta is approximately 1000 times greater than that of the Tangoblackplus. The properties of the photopolymers are listed on the Stratasys company website and also verified here by testing with dogbone samples [31,37], as per ASTM D638 for Veromagenta and ASTM standard D412 for Tangoblackplus. The composites were printed with a material jetting technology that allowed two distinct materials to be printed simultaneously. The resolution of the printer is on the order of 10 micrometers in all directions. Several copies of the benchmark and optimized composites were printed for mechanical testing and investigation of stress-strain response. These material samples are shown in Fig. 2(b).

2.4. Mechanical testing

The samples were prepared for testing after printing. An edge notch was cut a fifth of the sample length in the y-direction by a horizontal mill and sharpened by a razor blade. The sharp notch promotes the crack propagation from the crack tip. The dimensions of the sample were 76 by 76 by 2 mm in addition to 25 mm-wide buffer regions for the grips on the two opposite sides. For secure gripping of the specimens in the testing apparatus, four aluminum tabs were attached over the grip areas with epoxy glue. The samples were tested in an Instron 5582 Testing Machine with 100 kN static load cell under displacement boundary conditions. Furthermore, the samples were clamped in place with steel grips. The displacement rate in the experiment was 2 mm/min for all samples tested. The testing procedure in this study was not meant to emulate a standard procedure as outlined in ASTM; instead, this approach is in line with the computational modeling scheme employed in this study and allows for true comparison between the benchmark and optimized composites. Stress-strain curves were obtained from mechanical testing. The toughness modulus is defined as the area underneath the stress-strain curve and is a measure of fracture resistance [31,37]. The samples were assumed to have failed once the load dropped by 20% or more. Due to the assumptions made in the model concerning the constitutive relationship of the base materials, it was not expected for the model to give accurate quan-
2.5. Digital image correlation

The samples were sprayed on one side with black and white paint for digital image correlation (DIC) before testing. DIC was carried out using the VIC-2D software created by Correlated Solutions [44]. The main components of the DIC include a user interface that allows different frames of the deformation at different times to be viewed, and also a camera pointed at the specimen while it is loaded in the Instron testing machine. Tape was placed on the tabs before spraying to prevent paint from getting on the aluminum tabs, which might cause slipping during mechanical testing. The DIC measures displacements between the black and white sprayed dots while the sample undergoes deformation, thus permitting visualization of the strain field at each instant of time.

3. Results and discussion

3.1. Mechanical properties: stress-strain relations

The geometries obtained from simulation were manufactured using a Stratasys Connex 3 3D-printer. Representative stress-strain response curves for all materials are depicted in Fig. 3(a). The stress-strain response of the softer materials is shown in a separate plot in Fig. 3(b). The softer materials exhibit low stress failure but high strain failure. The failure strain for Soft material is greater than 50 times higher than for the base Stiff material. The Stiff material, on the other hand, has greater than 50 times higher failure stress than the base Soft material. This shows that these two materials exhibit contrasting failure responses. Both the MIT and Rnd samples have very similar stress-strain responses as compared to the Stiff material, but their stiffness is diminished due to the addition of soft elements. The same applies to the Min-T sample, which has a much higher stiffness compared to the Soft base material owing to the increased amount of stiff elements in the Min-T samples. The optimized geometries performed very similarly, as they were produced from the same optimization algorithm, albeit with different initial random configurations. Both optimized geometries have similar stress-strain responses, as indicated in Fig. 3(a). Additionally, the area underneath the optimized geometries is much greater than with the other samples. Therefore, the results demonstrate that the optimized composites have a marked improvement of toughness and strength (maximum stress) over the benchmark geometries.

The optimized and benchmark geometries are compared through the introduced parameter of “toughness amplification.” Toughness amplification is a ratio of the average sample toughness divided by the average toughness of the homogeneous Stiff sample \( T/T_{\text{ref}} \). The toughness amplification and measured strength of all the printed samples are shown in the bar chart in Fig. 3(c) along with
error bars, which represent ±1 standard deviation. The two optimized geometries have on average a more than 20-fold increase in toughness modulus compared to the Stiff homogeneous sample. The increase in toughness modulus of the optimized geometries compared to the other samples is of the same order of magnitude (~10-fold). It is believed that the optimized solutions, which both have very similar geometries, have this high toughness amplification due to the soft material localizing around the region of the crack tip, absorbing as much energy as it can before failure. As predicted from simulation, the minimum toughness (Min-T) sample performed the worst among other benchmark samples with a 20% soft volume fraction. For the Min-T sample, once the crack reaches the soft material, it follows the path of the soft material to failure, which contributes to its low toughness modulus. Rnd performed similarly in toughness amplification to Soft base material, as its increase in failure stress compared to Soft base material is compromised by its inferior failure strain. The toughness modulus combines information from both these material attributes. The Rnd sample actually has higher toughness than stiff base material even though the soft material distribution is not optimized, with soft elements spread out in a less efficient and less scrupulous manner throughout the composite. Any soft elements around the crack tip are better than no soft elements when reducing the stress concentration at the tip. This result suggests that a random sample with some soft material distribution in a stiff material will perform better than a homogeneous stiff material. However, it also shows that a random sample still cannot outperform optimized geometries with the same constraints.

Fig. 3(d) depicts performance in terms of strength for the various samples. The results show that the strength of the optimized samples is enhanced, surpassing the stiff and random samples by approximately two-fold, and the other samples by orders of magnitude. It is interesting to note that the addition of soft materials to a strong material using an algorithm-driven approach has led to a stronger material. It is also worth noting that Stiff, Rnd, and MIT performed very similarly in strength, which may be justified by the dominance of the failure mechanism of the stiff material. The weakest samples are Min-T and Soft, which makes sense because the failure mechanism of the soft material dominates in both these cases.

3.2. Digital image correlation comparison with computational results

For the homogeneous and optimized samples, DIC was used to visualize experimental strain fields and compare them with simulation results, shown in Fig. 4. It can be seen from the strain field of the homogeneous material that a strain concentration exists at the crack tip because the stresses at the crack tip are magnified due to the presence of the notch. The stress at the crack tip surpasses the critical stress of the specimen, and consequently the crack propagates through the material from the crack tip. This phenomenon is exhibited when the stiff bulk material fails catastrophically. For the
optimized composites, the strain field is much more distributed and the entire specimen works in concert to resist fracture. The strain is no longer concentrated at the crack tip; instead, it is highest in the locations where there is soft material that can withstand the highest strain. The simulation and the experimental images show good agreement of the stress and strain delocalization of the optimized materials. Moreover, there is stress and strain delocalization in the optimized materials that is not manifest in the homogeneous material. Harnessing this insight, it becomes possible to optimize for toughness before manufacturing for a wide range of engineering applications, such as airplane wing composite designs, or scaffolds and implant devices for biomedical applications.

3.3. Discussion

In this study, optimized designs constrained with a volume fraction of 20% soft material were generated. Introduction of soft elements is necessary to alleviate the stress concentration at the crack tip, resulting in a higher strength and toughness compared to the stiff base material, as shown in Fig. 3. At the same time the designed composites cannot be completely made of soft material, as this will recover the Soft base material with inferior strength, stiffness, and toughness, as shown in Fig. 3. A 20% volume fraction is chosen because it is desired for this composite design to strike a balance between structural integrity (stiffness) and fracture resistance (toughness); a 20% volume fraction of soft material satisfies this requirement [43].

The approach presented here has demonstrated that optimized geometries with increased toughness and strength can be created through algorithm-driven design. Initial tests were run with varied volume fraction (10–30%) with no significant change in the optimized topology. In all cases there is still localization of soft material at the crack tip to alleviate stress concentration. As the volume fraction decreases, the soft material is still allocated around the crack tip by the algorithm, but sparsely. As the volume fraction increases, the soft material is still around the crack tip, with the excess amount of soft material distributed further from the crack tip, which can be detrimental to composite toughness. Indeed, these findings suggest an optimum volume fraction, determination of which is outside of the scope of the current work, but a direction for future research. Future work can include the volume fraction as a variable in the optimization problem to find the ideal volume fraction, especially for applications where materials are scarce.

An elastic response is assumed in the material models for both the stiff and soft phases. Expanding this research to more ductile materials that exhibit a plastic response can be done by considering a more complicated constitutive material model. These inelastic models can include parameters such as yield, hardening, and flow behavior during plastic response and can be incorporated in future work. Additionally, the model shown here considers perfect adhesion at the interface between the two phases. This assumption is made based on the additive manufacturing method (discussed in Methods section) used which cures the two material phases simultaneously in situ, guaranteeing sufficient interfacial adhesion between the two phases. Moreover, this is validated by the experiments where it is observed that the samples do not fail at the interface.

During the simulation process, different benchmark geometries are compared to the generated optimized geometries. The mesh size of the samples is kept constant during simulation in order to compare all geometries. The given mesh discretization is chosen to minimize computational cost, as optimization can become very expensive as the number of finite elements increases. Additionally, the element size is chosen to be compatible with the resolution of the printer. Different geometries can be compared to each other to distinguish if a better design and response can be obtained only if their mesh sizes are the same. Nonetheless, the results are sensitive to the grid size used and this may be another parameter to tune in future work. The focus of this work has been to show a proof of concept and experimentally validate that better designs for fracture resistance can be generated through an algorithm-driven approach. This search for a better design, not necessarily the optimal, is valid for engineering applications and adopted in literature [45].

Starting from different initial random material topologies, there may be different optimal designs. Especially as mesh size gets larger, it becomes more difficult to reach a global optimal design due to the increasing number of possibilities. As with many heuristic optimization approaches (simulated annealing, greedy, genetic, etc.), there can never be a guarantee of a global optimum, because sometimes it is possible to obtain multiple local minima [10,46,47]. Instead, optimization comes with the assurance that the obtained solution is definitely better than the starting configuration. In terms
of an optimization problem the results show that multiple solutions are possible to improve the objective function. Experiments have shown that even though the geometries are slightly different, both optimized geometries under the same constraints performed better than homogeneous and random samples and actually performed similarly to each other with respect to the introduced toughness modulus metric.

This study has shown a method to tune and optimize material properties such as fracture toughness through an algorithm-driven approach. The optimum solutions of the algorithm were 3D-printed, mechanically tested, and analyzed. Results from simulation and experiment clearly demonstrate that introducing soft elements into a stiff element field causes a redistribution of strain at the crack tip under load. This larger strain footprint alleviates the stress concentration in the sample and leads to a composite with toughness that surpasses its constituents. This property-oriented approach can be applied to a wide range of engineering problems, such as airplane wing designs, at the design stage rather than further along in the process, thereby cutting expenses and saving time for manufacturers. However, it is important to remember that this problem is solved with an a priori given set of boundary conditions. This may not be possible in some applications where loading conditions are unknown. It is left to future work to investigate geometries and microstructure that can obtain high fracture toughness under any loading condition to be suitable for those applications.

4. Conclusions

In summary, this paper reports the optimization of a composite structure based on soft and stiff building blocks for a model problem of a single crack. With a redistribution of soft and stiff elements, the optimized composite morphology achieves a better design by altering the stress field and maximizing the toughness modulus. The 3D-printed optimized geometries, along with benchmark geometries, are used through subsequent testing to obtain the stress-strain responses for each of the specimens created. The results show that the optimal geometries had a higher toughness and strength than the benchmark geometries. Additionally, a direct comparison of strain fields around cracks is reported, showing excellent agreement between simulation and experiment. Moreover, a more delocalized strain field leads to the optimized designs having a higher toughness than the homogeneous designs. This new insight sheds light on the possibility of tuning material properties before production, which can be vital when it comes to designing fracture-resistant composites for various engineering applications in the aerospace, biomedical, and automotive industries. This work demonstrates the efficacy of the synergy between algorithm use and additive manufacturing as verified by testing, and exemplifies a materials-by-design process.

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