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Physical property modeling for composites

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Abstract

Recent developments in simulating the physical characteristics of composites are discussed, focusing on three distinct areas. Among these are (i) theoretical approaches to microstructure/property relations; (ii) imaging techniques like X-ray microtomography, which provide high-resolution, three-dimensional microstructural phase information of a composite sample in a non-invasive manner; and (iii) topology optimization, a promising numerical technique that allows for the design of composites with tailored material properties. Current restrictions are discussed, as are the need for future studies.

1. Introduction

Famous scientists and engineers like Maxwell (1873) and Einstein (1905) have grappled with the challenge of calculating the macroscopic or effective physical characteristics of composite media. A heterogeneous material is one that has either distinct regions of various materials (phases) or regions of the same material in distinct phases or states. It is the numerous cases where the'microscopic' length scale (such as the average domain size) is significantly bigger than the molecular dimensions (such that the domains contain macroscopic features) that are the subject of this article. Assigning macroscopic or "effective" qualities to a heterogeneous material is possible under these conditions since the substance may be seen as a continuum at the microscopic scale. There are many examples of heterogeneous media in both natural and artificial settings, such as porous and cracked media, polycrystals, polymer blends, foams, fluidized beds, photographic emulsions, cermets, soils, rocks, blood, and animal and plant tissue. Identifying the relevant characteristics of disordered systems is obviously crucial from a technical perspective.

direct measurement, empirical relations, and approximate as well as rigorous theoretical methods have resulted in a vast body of literature on heterogeneous materials (such as composite and porous media; see Beran, 198; Christensen, 1979; Willis, 1981; Torquato, 1991; and references therein). It is impractical, in terms of both time and money, to do direct measurements on every material sample, covering all potential phase property values and volume fractions. Data correlation is where empirical relations really shine, rather than prediction. For a more comprehensive method, one may compute the properties from the microstructure of the d isordered material and then use this information to quantitatively connect changes in the microstructure to changes in the macroscopic parameters, since effective properties are sensitive to the specifics of the microstructure. This has significant ramifications for the development of materials with custom-tailored characteristics.
In this report, we provide an overview of progress in these three areas:
microstructure/property connections from a theoretical perspective;

topology optimization, a promising numerical technique that allows one to design composites with tailored material properties, and X-ray microtomography, an imaging technique that allows one to obtain high-resolution three-dimensional microstructural phase information of a composite sample in a nonintrusive manner.
Due to the sheer volume of published material, it would be impossible to cover even the most significant advances here. Elastic moduli, thermal and electrical conductivity, thermal expansion coefficients, piezoelectric coefficients, and composite failure characteristics are the physical qualities that will be discussed. We note, however, that the identical methods used to study apparently unrelated features (fluid permeability and trapping rate) have been documented elsewhere (Torquato, 1991). In other words, it is beneficial to

consider all physical features of composites in the same broad context.

First, theoretical regressions

Information Beyond the Volume of Fuction 1.1

For the sake of clarity, we will limit our discussion to composites with just two phases, which we will refer to as phases 1 and 2. The ith phase is defined by a set of physical qualities (elastic moduli, strength, conductivity, etc.) and a constant volume fraction $i, in the case of a statistically uniform system, depending on the physics involved. In terms of microstructure, the volume fraction is the most basic but also most useful piece of data.

The physical characteristics of composites are often predicted by employing outdated methods such as simple combination principles. For any effective property Ke, two common mixing rules are the so-called arithmetic average and the geometric mean.

$$K_e = K_1\$_1 + K_2\$_2 \tag{1}$$

and the harmonic average

$$K_e = \frac{K_1K_2}{K_1\$_1 + K_2\$_2}, \tag{2}$$

where Ki is the phase-i specific variable. The phase volume fractions are the sole quantities included in both the arithmetic average Eq. (1) and the harmonic average Eq. (2). Nonlinear parameters, including strength, are also estimated using mixing rules, in addition to linear properties like elastic modulus and conductivity. It is well knowledge that the arithmetic mean best describes linear features.

Fig. 1. Gray-scale digitized microscope image of a three-phase boron carbide/aluminum composite: white region is Al phase, blackregion is B_4C phase, and gray region is Al_4BC phase. The B_4C and Al phases are the dominant ones.

The effective attribute is often overestimated by Eq. (1) and underestimated by the harmonic average, Eq. (2).

Dispersions of inclusions in a matrix are often approximated using effective-medium or self-consistent approximations (Hill, 1995; Budiansky, 1995), both of which are popular among more experienced practitioners. Basically, you take a regular inclusion and place it within a matrix that already has the mysterious effective characteristic. Because of this, The inclusion shape, together with the inclusion's volume fraction and phase characteristics, must be taken into account when calculating the effective property. Even for moderate to high phase contrast values, self-consistent estimates fail (Christensen, 1979; Torquato, 1998), and they predict, in particular, erroneous percolation thresholds for dispersions. Self-consistent approximations have been demonstrated to be precise for media with a certain kind of topological symmetuy, which is absent in the more common particle and fiber-reinforced composites. Thus, until the phase contrast is sufficiently tiny, self-consistent approximations should not be used to dispersions. Nonetheless, many people keep using it in this context, perhaps due to its attractiveness as a straightforward analytical phrase.

When modeling particle dispersions in a matrix, it is necessary to account for the interconnectedness and disconnection of the matrix and particle phases. To account for these topological characteristics with just volume fraction data, composite-spheres models have been proposed (Hashin, 192; Christensen and Lo, 1979). In the absence of considerable particle clustering, the resulting mathematical formulas for the effective elastic moduli are in excellent agreement with experimental data on dispersions. Microstructural information beyond volume fractions is required when clustering is a concern.

Effective property modeling approaches for increasingly complex microstructures must fail if they rely just on volume fraction data. The micrograph in Figure 1 depicts the microstructure of a three-phase boron carbide/aluminum composite (Torquato et al., 1999), which is rather complicated. While in two dimensions just the boron carbide phase seems to be linked, in three dimensions both phases are interconnected, making it difficult to extract a simple unit cell (as is normally done for dispersions) that describes the system's behavior.

These examples demonstrate that knowledge about the microstructure, beyond that included in phase volume fractions, is crucial to understanding the effective characteristics, and we will refer to this information as higheu-oudeu microstructural information. How can we include this higher-order microstructural knowledge into our property predictions, and how can we properly quantify it? It's obvious that getting these things done will require returning to basics. For linear effective characteristics (elastic moduli, conductivity, etc.), one may take use of rigorous bounding methods and exact system-independent procedures to arrive at accurate microstructural functions. Depending on the physical attribute of interest, these values take the shape of different kinds of statistical correlations with n points.

Limits/microstability uelations

The microstructure (including the correlation functions) is incompletely understood in almost all cases. Given the lack of complete microstructural information, it is natural to try to estimate the effective properties using only partial statistics (lower-order correlation functions), and in particular to establish the range of possible values the effective properties can take (i.e., to determine rigorous upper and lower bounds on the properties). Bounds on effective attributes are often derived using minimal energy concepts.

Improved limits go beyond the information available from the volume fractions and instead rely non-trivially on two-point and high-order correlation functions. Examples of situations where tighter constraints are preferable include the conductivity and elastic moduli of isotropic materials (Hashin and Shtrikman, 192, 193). Both linear (Beran, 198; Milton, 1982; Milton and Phan-Thien, 1982; Torquato, 1991) and nonlinear (Willis, 1991; Ponte Castaneda and Suquet, 1998) materials have improved bounds on a wide range of effective properties in terms of Sn(xn), the probability of finding n points at positions xn x1,..., xn in one of the phases. The elastic moduli and conductivity of different composites have been constrained within three and four-point limits, respectively (Torquato, 1991). Despite this, three
both two- and four-point limits have their uses, however when the contrast between the phase characteristics grows, the bounds diverge. For instance, when phase 2 is superrigid with respect to phase 1, the upper limit often diverges to infinity, even though phase 2 is topologically unconnected. Bounds using such lower-order information are referred to as conventional enhanced bounds, and the explanation for this behavior is because loweu-oudeu Sn do not represent information about peucolating clusteus ou linked routes in the system. However, it should be emphasized that, in high-contrast situations, one of the bounds can still provide a good estimate of the properties, depending on whether or not the system is above or below the peucolation thueshold, i.e., the point at which a disconnected phase becomes connected (Torquato, 1991). Traditional upper and lower constraints on effective qualities have been expressed in terms of several statistical variables such as the point/q-particle function Gn (n = 1+q)

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(Torquato, 1998a) and the surface-void Fsv and surface-surface Fss correlation functions (Doi, 1997a). It is preferable to construct sharper limits in terms of morphological quantities that better capture percolation or connectivity information, although traditional bounds may still be useful in certain cases of great difference. Recently, such limitations in terms of the nearest-neighbor distribution function HP have been defined and calculated for the issue of conduction in particle suspensions (Torquato and Rubinstein, 1991). Torquato and Avellaneda (1991) established constraints on some diffusion parameters for porous media in terms of the pore-size distribution function P(d).

1.1. Micuostuuctuue chauacteuization

The previous section described some of the different types of statistical correlation functions (S_n, G_n, F_{sv}, F_{ss}, H_P, P) that have arisen in rigorous bounds on effective properties. Until recently, application of such bounds (although in existence for almost thirty years in some cases) was virtually nonexistent because of the diAculty involved in ascertaining the correlation functions. Are these different functions related to one another? Can one write down a single expression that contains complete statistical information? The answers to these two queries are in the aArmative.

For statistically inhomogeneous systems of identical d-dimensional spheres, a general n-point distribution function H_n has been introduced and represented (Torquato, 1986b). From the general quantity H_n, one can obtain all of the aforementioned correlation functions and their generalizations. This formalism has been generalized to treat polydispersed spheres, anisotropic media (e.g., aligned ellipsoids and cylinders) and cell models (see Torquato, 1991). We should mention that quantities that are superb signatures of clustering and percolation have been studied and evaluated (Torquato, 1991).

In the last decade or so, considerable progress has been made on the determination of statistical correlation functions from computer simulations (Monte Carlo and molecular dynamics). From a theoretician‘s point of view, simulations may be regarded as ‘experiments‘ that one may test theories against for specific models of heterogeneous media. Computer simulations also offer a means of studying model systems which may be too diAcult to treat theoretically. Obtaining statistical measures, such as H_n, from simulations is a two-step process. First, one must generate realizations of the disordered medium. Second, one samples each realization for the desired quantity and then averages over a suAciently large number of realizations.

Financial market fluctuations (1.2)

The following is a fundamentally and practically interesting question: Is there a way to uigouously connect the material's varied properties? Such correlations between properties would be very helpful in the multipurpose development of composites. I am particularly interested in whether or whether there exists a rigorous relationship between the overall thermal (electrical) response of a composite to an applied thermal (electrical) load and the overall linear mechanical response of the same medium to an applied mechanical load.

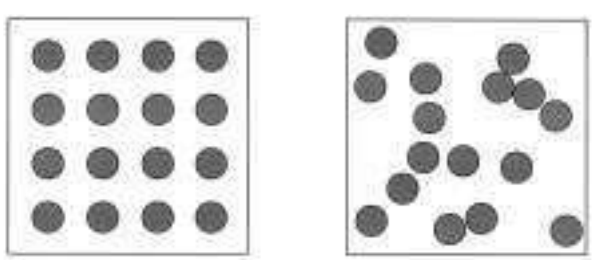

Fig. 2. Two systems at the same volume fraction but the right-most example has greater fluctuations. The failure characteristics ofthese systems can be dramatically different from one another.

Milton (1984) and Berryman and Milton (1988) developed rigorous cross-property constraints connecting conductivity and elastic moduli. After then, the so-called tuanslation technique (Gibiansky and Torquato, 1999) was used to further enhance these findings. Given an accurate determination of one of the effective properties, how precise are these cross-property estimates? Hexagonal arrays of superconducting, superrigid inclusions (phase 2) in a matrix may be used to get precise values for the effective conductivity and effective bulk modulus Ke, which can be used to probe this issue. The actual elastic-moduli data agrees very well with the cross-property limits. Because they do not take into account the fact that the supeuuigid phase is unconnected, the standard vauiational uppeu limits on the effective properties (such as HashinShtrikman) diverge to infinity in this case. In contrast, the upper limit based on cross-property information takes use of the fact that the infinite-continuum phase is unconnected.

The effect of spatial value on composite failure

Failure in composites is a vast topic that goes beyond the scope of this study. For more information on the failure of fiber composites, the reader is directed to the works of Dvorak et al. (1992), Budiansky et al. (1995), Zhou and Curtin (1995), and Christensen (1997); for details on crack propagation in brittle composites, check out Haubensack et al. The applicability of statistical mechanics in characterizing microstructure fluctuations and failure in composites is the narrow topic of this article.
The effect of spatial diversity in the microstructure of composites on the failure characteristics of the heterogeneous materials (see Fig. 2) is of major practical relevance. Several case studies from the published literature are discussed to drive home the idea. Under three-point bending, the strength of borosilicate glass reinforced with SiC fibers was measured by Barsoum et al. (1992). As the local fiber spacing grew, the tension at which matrix fractures began decreased. Therefore, the maximum interfiber spacing is the relevant length scale; in other words, the strength is controlled by the extreme statistics. Since volume fraction information is comparable to average fiber spacing, this means that naïve efforts to estimate the strength using a basic rule of mixing relation Eq. (1) must unavoidably fail. Later on, Botsis et al. (1997) demonstrated that the best correlation with the data was achieved by using a GriAth-type scaling relation for the strength involving the maximum interfiber spacing.
MacHay (1990) conducted an experiment to examine the effects of temperature cycling on cracking in unidirectional metal matrix composites. Matrix fractures and interfacial debonding were traced back to residual strains. More cracking occurred between closely spaced fibers within a row, suggesting a correlation between fiber distribution and cracking. Chung and Weitsman (1994) conducted a theoretical investigation on the compressive strength of unidirectional composites, and they concluded that random fiber spacing

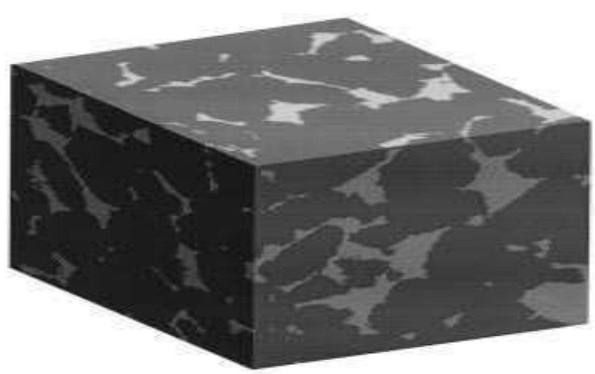

Fig. 3. Fountainebleau sandstone: lighter shade is void and darker shade is solid.

led to significant transverse loadings developing on the fibers. In order to complete the study, the probability density of fiber separation was needed.
These three cases illustrate how even small variations in microstructure may significantly affect how a composite material fails. Although microstructure fluctuations have been studied for quite some time in statistical physics, this research has yet to make its way into the fields of practical mechanics and materials science. Statistics of inteufibeu spacing (Torquato and Lu, 1993) and local volume fuaction fluctuations inside a 'observation' window (Quintanilla and Torquato, 1997) are two such examples th at have been quantified with the use of statistical mechanics. Since these statistical measures account for all statistical moments, they are

directly applicable to the aforementioned three issues.

Microscopic X-Ray Tomography

Sectioning was widely used in the first efforts to characterize the microstructure of heterogeneous materials. Since this method not only destroys the sample but also often causes the sections themselves to be changed during the sectioning process, it is undesirable, particularly in biomedical applications. There was a need for the development of non-invasive methods. Both transmission and scanning tunneling electron microscopy are well-established non-invasive methods at this point, although they only provide a two-dimensional view.
The whole three-dimensional structure of samples has been shown to have a significant impact on the physical characteristics of composites. Noninvasive methods that offer three-dimensional phase information have recently emerged, with examples being X-ray microtomography (Flannery et al., 1987) and confocal microscopy (Fredrich et al., 1995).
In the medical profession, computer-aided tomography (CAT) scans are a typical method of obtaining three-dimensional phase information. However, the achieved resolution is only about

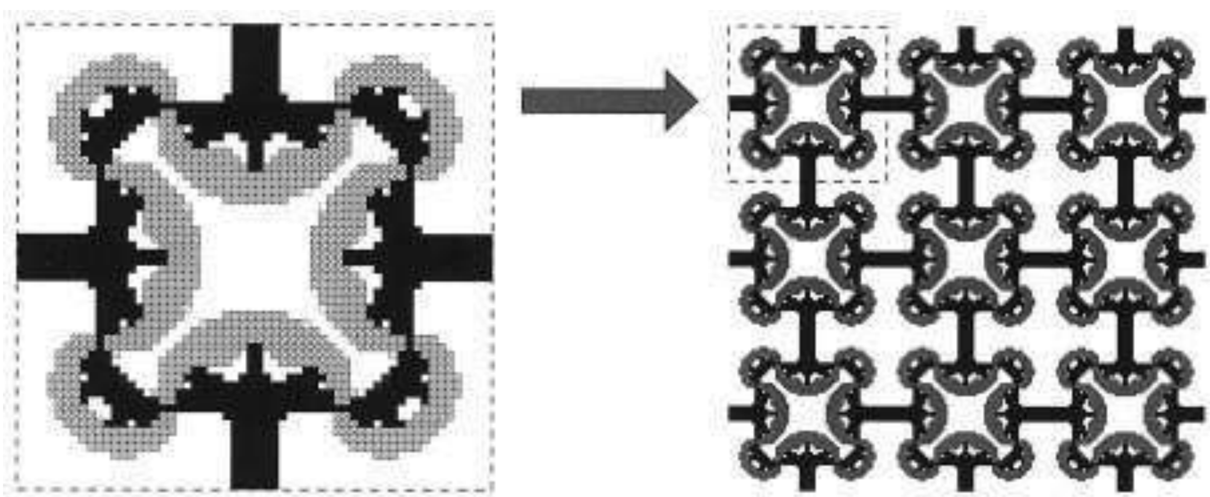

Fig. 4. Optimal microstructure for minimization of effective thermal expansion coeAcient (Sigmund and Torquato, 1997). Whiteregions denote void, black regions consist of low expansion material and cross-hatched regions consist of high expansion material.

~100 µm. Non-destructive examination of three-dimensional structural information is possible with high resolution (1 m) using synchrotron-based X-ray microtomography (Flannery et al., 1987). Microtomographic techniques have recently been used to the investigation of sandstone structure and characteristics (Coker et al., 1999). Microtomography data and reconstructed three-dimensional maps of the materials' X-ray opacity were acquired using beamline X2B at Brookhaven National Laboratory's National Synchrotron Light Source (NSLS).
The digitized picture of sandstone is kept as a 512512512 matrix of voxel values. The sample's electron densities were recorded in these voxels, and after being binned, the two phases are visible as two pe aks in the histogram of electron density values. The sample is then stored as a matrix of bits, with values of 0 (matrix phase) or 1 (void phase) depending on the threshold value used to differentiate the phases. A section through a slab of Fountainebleau sandstone is seen in Fig. 3. The intricacy of the empty space is worth noting. Using the modeling methods described above, the microstructural functions determining the property behavior may be extracted from this picture. This allowed us to calculate the trapping constant and the fluid permeability of the sample.

Optimization of topologies

The topology optimization approach is a novel and exciting tool for the systematic design of composites and smart material systems. A decade ago, Bendse and Hikuchi (1988) created the topology optimization approach, which was at first used for the design of mechanical structures. In addition to its usage in structural design, it has found applications in the design of smart and passive materials, mechanisms, MicroEle ctroMechanical Systems (MEMS), and other areas (Sigmund, 1994; Larsen et al., 1997; Sigmund and Torquato, 1997; Sigmund et al., 1998).

This is a simple statement of the topology optimization problem: allocate resources in a design space to maximize a metric of interest (Bendse and Hikuchi, 1988; Bendse and Hikuchi, 1995). Any subset of these elements may make up the goal function.

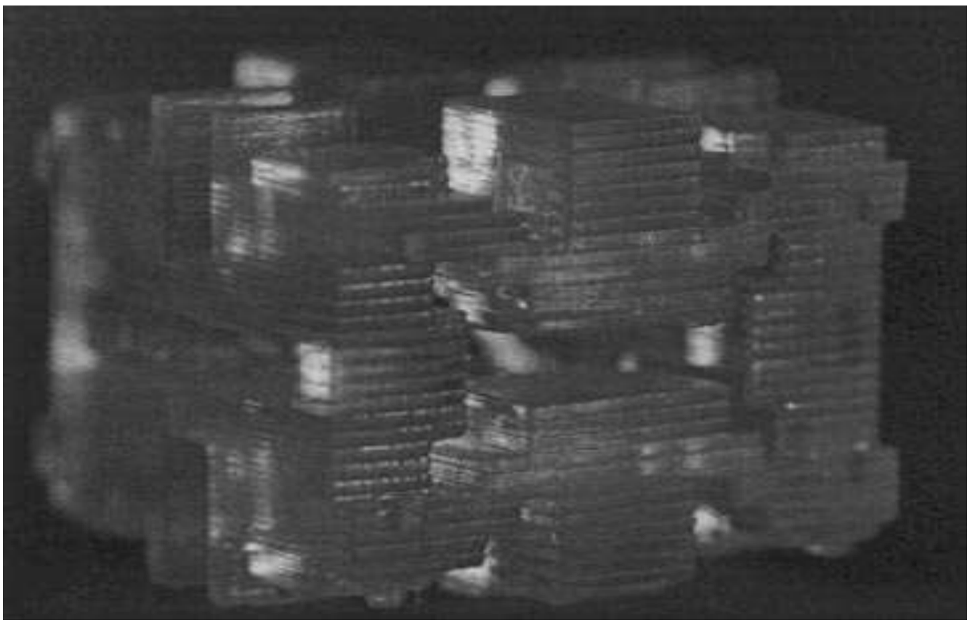

Fig. 5. Prototype of one base cell made by stereolithography. This special porous solid has negative Poisson‘s ratio and, when used as a matrix in a piezocomposite, maximizes hydrophone performance.

effective property tensor that is relevant under specific bounds. The design domain is the periodic base cell, which is discretized into many finite elements at the outset. The goal of the task is to identify the configuration of the basic materials and/or void that will result in the lowest value of the objective function. Based on the sensitivities of the objective function and constraints with regard to design modifications, the optimization technique solves a series of finite element problems before modifying the material type (density) of each of the finite elements (Sigmund and Torquato, 1997).

Extremely elastic, thermal, or piezoelectric composite materials have been designed using the topological technique (Larsen et al., 1997; Sigmund and Torquato, 1997; Sigmund et al., 1998). The technical and fundamental implications of materials having very or unusually low thermal expansion are of interest. Materials with zero thermal expansion are required in buildings, bridges, and pipes that are exposed to extreme temperature swings. 'Thermal' actuators may be made from substances that have a high thermal displacement or force. Contrary to common sense, a negative thermal expansion material will shrink when heated. After being heated, a fastener built from a negative thermal expansion material will slide right into place. It will expand as it cools and fit snugly into the opening. It is possible to create any of the three growth behaviors (Sigmund and Torquato, 1997).

Our discussion of the negative expansion situation, in which a three-phase material consisting of high expansion material, low expansion material, and void area is required, serves to demonstrate the power of the approach. Fig 4 shows The negative expansion behavior is driven by the bimaterial components of the ueentuant cell structure, which bend (into the empty space) and generate substantial deformation when heated, as shown by our two-dimensional optimum design.

Actuators using piezoelectricity may be optimized to provide as much force or displacement as possible. In addition, piezocomposites may be engineered (by embedding a grid of piezoceramic rods in a polymer matrix) to have the highest possible sensitivity to sonic fields. Piezocomposites with superior performance characteristics for hydrophone applications have been developed using the topology optimization technique (Sigmund et al., 1998). The ideal transversally isotropic matrix material has a negative Poisson's ratio in certain directions when designing for maximal hydrostatic charge coeAcient. This matrix material is a composite in and of itself, a novel porous solid. Three-dimensional negative Poisson's ratio materials have been created using an AutoCAD file representing the matrix material structure and a stereolithography method (Sigmund et al., 1998). This example depicts a prototypical cell, which is a polymer-based cellular solid. Fig. 5.

2) The necessity for more study

There will be significant future applications at the interface between solid mechanics and biology. Almost every biological material system is a composite with varying degrees of a single structural characteristic. The machinery of statistical mechanics has not been completely used in the study of composites, and there is presently no formalism to predict the effective characteristics of such complicated multi-scale composites. Characterizing the microstructure of statistically inhomogeneous media (such as functionally graded materials) and the toughness and strength of composites are two examples of outstanding topics where such technologies might be fruitfully utilized.

● Resolution and/or the sorts of materials that may be photographed are currently bottlenecks for current 3D imaging methods. The development of experimental techniques is required to allow the imaging of a broad range of materials and length scales in three dimensions. Reading this data into a computer allows for further analysis and visualization.

Optimal design applications incorporating linear qualities, let alone nonlinear ones, have yet to fully tap the potential of the topology optimization technique, which is still in its infancy. Future optimum design of genuine composite materials (including multifunctional design) will be possible with the help of this computational approach and enhanced production procedures.

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