Composites Science and Technology 162 (2018) 146–155

Contents lists available at ScienceDirect

Composites Science and Technology

journal homepage: www.elsevier.com/locate/compscitech

Identifying interphase properties in polymer nanocomposites using adaptive optimization

Yixing Wang[a], Yichi Zhang[a], He Zhao[a], Xiaolin Li[c], Yanhui Huang[d], Linda S. Schadler[d], Wei Chen[a], L. Catherine Brinson[a,b,*,1]

[a] Department of Mechanical Engineering, McCormick School of Engineering and Applied Science, Northwestern University, 2145 Sheridan Road, Evanston, IL 60208, USA
[b] Department of Material Science and Engineering, Northwestern University, 2220 Campus Drive, Evanston, IL 60208, USA
[c] Theoretical and Applied Mechanics Program, Northwestern University, 2145 Sheridan Road, Evanston, IL 60208, USA
[d] Department of Material Science and Engineering, Rensselaer Polytechnic Institute, 110 8thStreet, Troy, NY 12180, USA

ARTICLE INFO

Keywords:
A: Nanocomposites
B: Interphase
C: Finite element analysis (FEA)
C: Adaptive optimization

ABSTRACT

To predict the properties of nanocomposites, computational models have demonstrated that the interphase behavior can be expressed using the matrix properties with a modified (shifted) frequency. The amount of the shift necessary for a given sample data set can be determined by achieving the best fit between the predicted curve from a computation model and the experimental data through trial-and-error. However, with the complexity of experimental data and expensive computational costs, a manual process to solve this inverse problem is impractical to handle many experimental data sets. This difficulty hinders investigation of the underlying principles behind nanocomposite interphase. In this work, we present an adaptive optimization approach that accelerates the search for interphase properties in polymer nanocomposite data sets by solving the inverse problem using global optimization. The objective is to minimize the difference between the predicted bulk property of a nanocomposite with that from the experiment data. A Gaussian Process (GP) model is built as a surrogate of the objective function with quantification of prediction uncertainty. An adaptive sampling strategy is applied to effectively navigate the complex search space by iteratively selecting the next sampling point based on an expected improvement function. The surrogate model and the optimal solution evolve until the desired objective is achieved. The approach is tested on both the simulations of dielectric and viscoelastic properties in nanocomposites. Our work provides insight into identifying the interphase properties for polymer nanocomposites using adaptive optimization and demonstrates the potential of data-driven approach for achieving a deeper understanding of the interphase properties and its origins.

1. Introduction

Polymer nanocomposites have attracted great interest in recent years because of their potential as tailored materials with enhanced properties [1–3]. Recent experiments have shown that polymer nanocomposites are able to achieve significant improvement in dielectrical, thermal, mechanical and other physical properties compared with their parent polymer systems [4–9]. More importantly, these outstanding properties can be achieved at low filler loadings such that the polymer system does not sacrifice the advantages of easy processability [10,11].

One of the reasons for the enhancement in properties is the large interphase region that results from both the strong chemical and geometric interactions between the particle surface and the polymer segments near the particle and the high surface-to-volume ratio of the nanoinclusions. For example, if a composite sample contains 5 wt% of 40 nm particles perfectly dispersed, the resulting total interfacial surface area is about $3.5 m^2/cm^3$. As illustrated in Fig. 1, due to the interacting cooperative nature of the macromolecular network, the interphase area extends beyond the layer of matrix chains directly bound to the filler surface resulting in the significant extension of the interphase domain into the matrix [12].

In order to understand the behavior of the interphase and make accurate predictions of nanocomposite properties, efforts have been made to measure the interphase thickness and its mechanical or dielectric response [13–16]. Although direct measurements of the interphase are limited because of challenges in experimental visualization at

* Corresponding author. Department of Mechanical Engineering, McCormick School of Engineering and Applied Science, Northwestern University, 2145 Sheridan Road, Evanston, IL 60208, USA.
E-mail address: cbrinson@northwestern.edu (L.C. Brinson).
[1] Dr. Brinson is currently Professor of Mechanical Engineering and Materials Science at Duke University, Durham NC, USA.

https://doi.org/10.1016/j.compscitech.2018.04.017
Received 12 September 2017; Received in revised form 14 March 2018; Accepted 11 April 2018
Available online 28 April 2018
0266-3538/

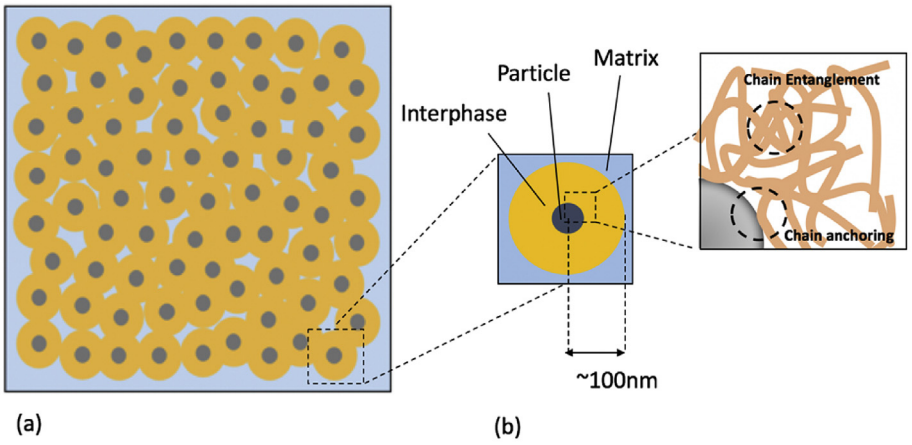

Fig. 1. Interphase configuration. (a) Schematic of the interphase regime in a nanocomposite sample. (b) Schematic showing the extended interphase structure. Yellow chains directly anchored to or chemically interacting with nanofiller. Blue chains impacted by filler indirectly through interaction with yellow chains. Cooperatively interacting chains propagate interphase zone to persist to the order of 100 nm from the particle surface. Note: figures not to scale.

the nanoscale, recent studies focus on measuring the local elastic and viscoelastic properties in different polymer microdomains by correlating thin film and nanocomposite data, providing adequate evidence that the local polymer properties are significantly altered in the vicinity of polymer surface [17–22]. The actual thickness of the interphase may vary depending on specific polymer-filler system and has a wide range values from several nanometers to hundreds of nanometers. In particular, an atomic force microscopy (AFM) -based method that directly measures the mechanical properties of polymers adjacent to a substrate with nanometer resolution shows a gradient of mechanical properties extending approximately 100 nm from confining surfaces [20,23].

To predict properties of nanocomposites, continuum mechanics methods are often employed in which three phases must be considered: polymer, particle and interphase. Micromechanical models [24], such as Halpin-Tsai, Mori-Tanaka and the self-consistent scheme have been applied to predict the thermomechanical behavior of nanocomposites [25–28]. A variety of analytical models have been developed to analyze the dielectric behaviors including the Bruggeman model [29], Lichtenecker model [30] and Todd-Shi model [31]. Given experimental limitations to measure interphase properties directly, one approach to determine the interphase properties is inversely through tuning the parameters in finite element analysis or micro-scale model constitutive equations using the bulk composite properties from experiments [31–34]. Importantly, in order to capture the dispersion state or the morphology information of the fillers, multiscale simulations are often necessary. For example, multiscale approaches have been applied to study the viscoelastic properties in polymer nanocomposites [35–37].

FEA simulations can accommodate complicated non-homogeneous material systems with explicit configuration of all relevant material phases. This makes FEA a good option for to analyze behavior of nanocomposites and include both important nanofiller dispersion as well as interphase properties. We have developed finite element models for analyzing the thermal and mechanical [32,38–40] and dielectric behavior [33,41] of polymer nanocomposites and investigated the impact of the interphase on the corresponding properties. We have shown that in some cases, the interphase properties can be described by shifting factors based on the pure matrix properties, which can be well represented by the Prony Series as a parametric expression of multiple relaxation times and strengths [38,41]. Given experimental data for both the pure matrix and bulk nanocomposites properties (either dielectric or thermomechanical) the necessary interphase properties can be determined from a trial-and-error based iterative tuning procedure by matching simulated results from FEA with experimental data. However, there are several disadvantages for this trial-and-error based manual fitting. First, this process can be very time-consuming given the complexity of experimental data and computational cost of the FEA (a single FEA model for one sample with ~10 k elements requires 30 min to run on typical server). Since many manual iterations are often required, where the optimal shifting factors are guessed based on the previous outputs, this tedious manual tuning process prohibits efficient investigation of the correlation between shifting factors and material constituent characteristics.

In this work, we present a combined FEA and optimization approach to accelerate the search of optimal interphase properties given experimental data of bulk property for the composite. Our objective is to find the optimal interphase properties that minimize the difference between simulations and experiments, and to do so with an automated procedure. We adopt an adaptive global optimization approach that incorporates Gaussian Processing (GP) modeling [42,43] and sequential sampling strategy [44] to efficiently find the global optimal solution. Our proposed method can accurately find the optimal shifting factors given experimental data in tens of iterations, which significantly eases the computation costs from simulations. We demonstrate our method by finding both dielectric and mechanical properties of the interphase based on composite property data. This method is an efficient and reliable tool to determine interphase properties and can facilitate future work of uncovering the relationship between interphase properties and material constituents.

2. Methodology

Our goal is an optimization of interphase properties for a single sample, for which we have a) constituent properties, b) composite properties, c) microstructure information. We seek to determine the interphase properties that will yield the composite properties (b) from (a) and (c). We assume the interphase properties can be represented by shift factors with respect to matrix properties (defined in more detail below). Thus, we seek to find the shift factors that will optimally match the composite data via an automated procedure applying adaptive optimization. Based on experience and literature data we begin with reasonable bounds for the shift factor values, and the space defined by the n-dimensional factors (n = 5 for dielectric case [41], n = 2 for mechanical case [36]) varying in these bounds defines the search space for the adaptive optimizer. The key components of the adaptive optimization method are summarized in Fig. 2: (1) The empirical bounds of shift factors are used to set the range to sample initial training sets of shift factors from design of experiments (DOE) using Optimal Latin hypercube (OLHC). For each set of shifting factors, the FEA model is run and outputs the simulated result. Then the objective function is formulated as the difference between the experimental data and the simulation using mean square error (MSE); (2) A surrogate model, in our case, a Gaussian Process (GP) model, uses the training data to learn the relationship between the objective (Difference F) and features (shifting factors), with uncertainties.; (3) adaptive optimization (selector) provides the most promising candidate points for the new simulation and augments the initial shift factor set from DOE. In our study, new