Microstructure Modelling and Ultrasonic Wave Propagation Simulation of A206-Al$_2$O$_3$ Metal Matrix Nanocomposites for Quality Inspection

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Abstract

Ultrasonic testing is a promising alternative quality inspection technique to the expensive microscopic imaging to characterize metal matrix nanocomposites. However, due to the complexity of the wave-microstructure interaction, and the difficulty in fabricating nanocomposites of different microstructural features, it is very challenging to build reliable relationships between ultrasonic testing results and nanocomposites quality. In this research, we propose a microstructure modelling and wave propagation simulation method to simulate

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ultrasonic attenuation characteristic for A206-Al2O3 metal matrix nanocomposites (MMNCs). In particular, a modified Voronoi diagram is used to reproduce the microstructures and the numeric method elastodynamic finite integration technique (EFIT) is used to simulate the wave propagation through the generated microstructures. Linear mixed effects model (LME) is used to quantify the between-curve variation of ultrasonic attenuation from both experiment and simulation. Permutation test is employed to quantify the similarity of the quantified variation between experiment and simulation. This research supports the experimental results through the simulation approach and provides a better understanding of the relationship between attenuation curves and the microstructures.

**Keywords:** microstructure modeling; metal matrix nanocomposites; ultrasonic attenuation; acoustic wave; numeric simulation

1. **Introduction**

A206-Al2O3 metal matrix nanocomposites (MMNCs), where lightweight A206 alloys (93.5%-95.3% Al, 4.2%-5.0% Cu) are reinforced with nanosized Al2O3 particles, have been intensively studied recently because of their significantly enhanced mechanical properties, such as high strength, ductility, long fatigue life, and excellent hot tearing resistance [1-6]. It can be fabricated by dispersing Al2O3 nanoparticles into the molten A206 using ultrasonic cavitation assisted casting technologies [7, 8]. Well dispersed Al2O3 nanoparticles in A206 have strong nucleation potency and can significantly reduce the grain size of the primary Al phase and break the Al2Cu intermetallic network [1, 3], thus leading to significantly reduced hot-tearing susceptibility and enhanced mechanical properties. To facilitate the scale-up production, a fast yet effective quality inspection technique is critically important to ensure the quality of nanoparticle dispersion and morphology modification. Currently, the standard quality inspection method is to use microscopic images of nanocomposites microstructures, which are very time-consuming and costly to obtain. It is highly desirable to develop alternative simpler and effective quality inspection techniques.

Ultrasonic testing is one of the most popular nondestructive evaluation techniques. It has been intensively investigated and widely used in size measurement, flaw detection [9], structural health monitoring (SHM) [10], and materials and biological tissue characterization [11-15] etc. Ultrasonic attenuation is one of the most commonly used ultrasonic parameters in the ultrasonic testing applications. It refers to the decaying rate of the acoustic wave as it propagates through materials, which can be measured using the spectral ratio analysis technique [16], as shown in Fig.
1, where the two successive echoes reflected from the back wall of the sample are extracted and ratio of the spectrum amplitude is used to calculate the attenuation curves. The ultrasonic attenuation is highly dependent on the material properties and microstructural features, e.g., elastic constant, grain size, grain boundaries, inclusions, porosity and dislocations. Therefore, ultrasonic testing is promising to be an economical and effective method to characterize the microstructural configurations and material properties.

![Ultrasonic Testing Illustration](image)

**Fig. 1.** Illustration of the ultrasonic testing using ultrasonic attenuation curves [3].

Recently, Wu *et al.* [3] discovered an important relationship between acoustic attenuation profiles and the microstructural characteristics of A206-Al₂O₃ nanocomposites. For nanocomposites with satisfactory microstructures (i.e., small grain size, dissolved Al₂Cu phase and well dispersed Al₂O₃ nanoparticles), the between-curve variation of attenuations measured at different locations is much lower than that of bad quality nanocomposites. This study provided useful guidelines to establish a new quality inspection technique for A206-Al₂O₃ MMNCs. However, there still exist several issues that need to be addressed in order to develop a reliable quality inspection method: 1) there are multiple microstructural features (e.g., grain size, Al₂Cu morphology) affecting the variation of ultrasonic attenuation. However, due to the complexity of the interaction between the microstructural configuration and wave propagation, how each feature contributes to the variation is still unknown; 2) the nanocomposites samples and experimental data are quite limited because of the high experimental cost and the difficulty in fabricating samples with planned microstructural features, which makes it difficult to build a quantitative relationship between the attenuation curve and microstructural features.
For the reasons given above, numerical simulations of ultrasonic wave propagation in A206-Al$_2$O$_3$ MMNCs are needed to support the experimental tests by generating alternative data under different microstructural features, leading to a better understanding of the relationship between the microstructural configurations and attenuations. Numerical simulation of ultrasonic wave propagation have attracted intense interest for its promising in solving problems that may be inaccessible to direct experimental study or too complicated for theoretical analysis. It allows easy control of each experimental parameter independently, which enhances the understanding of wave propagation in complex systems. Indeed, using simulation to help explain the complex relationship between process parameters and process outputs has been widely used in the manufacturing science [17-19]. The most common techniques used to solve the wave propagation equations include the finite difference methods (FDM) [20-22], the elastodynamic finite integration technique (EFIT) [23], the finite element method (FEM) [24, 25], and the spectral finite element method (SFEM) [26]. Acoustic wave simulation has gained more popularities in many areas in recent years for the progress in computational power and availability. For instance, assessing the stability of an implant is difficult due to the complex heterogeneous nature of bone in ultrasonic bone and biological implant characterization. The use of numerical simulation enable researchers to understand the wave propagation phenomena occurring in prototype titanium cylindrical implants and to investigate the sensitivity of the ultrasonic response to variations of the biomechanical properties of surround tissues, which are determinant for the implant stability [15, 22, 27]. Another example is the area of structural health monitoring (SHM). SHM for the detection of damage in aerospace materials is an important engineering area. Experimental signals of complicated flaw geometries may be difficult to interpret. With the help of numerical simulation, scientists are able to investigate ultrasound scattering from flaws in materials and to develop optimized experimental SHM techniques [28]. Ultrasonic wave propagation simulation has also been applied in materials characterization [29, 30], however, very limited simulation work has been done on lightweight alloy based nanocomposites.

In this study, the simulation approach to model the microstructural features of A206-Al2O3 MMNCs in 2D space is developed and the ultrasonic wave propagation on the generated nanocomposites is simulated to study the relationship between the microstructural properties and ultrasonic attenuations. To simulate the MMNCs microstructure, a Voronoi diagram is first generated, and then the edges of the generated diagram is modified to describe different
morphologies of Al2Cu intermetallic phase. In the wave propagation simulation, the EFIT is selected for the following reasons: 1) EFIT naturally requires staggered spatial and temporal grids, which leads to stability; 2) boundary conditions are easily incorporated into EFIT; 3) the mathematical analysis is straight-forward and leads to equations that are easy to implement in any programming language. The simulated acoustic attenuations are consistent with the experimental measurements, which then can be used to further investigate the relationship between the microstructural properties and ultrasonic attenuations and to develop statistical quality control methods for scale-up production.

The rest of this paper is organized as follows. In Section 2, the Al2O3 nanoparticle based morphology modification mechanism is first introduced. Then the microstructure of A206-Al2O3 MMNCs is simulated based on the microscopic images and the morphology modification mechanism. In Section 3, the EFIT is briefly introduced. The simulation and experimental results are presented in Section 4. The statistical similarity testing between the simulation results and the experimental ultrasonic measurements is given in Section 5. Section 6 presents the conclusion and briefly introduces the future research directions.

2. Modelling and Simulating Microstructure of A206-Al2O3

In this section we first introduce the microstructural features of the A206 alloys and Al2O3 reinforced nanocomposites, and the morphology modification mechanisms. Based on these features, we propose a new microstructure modelling method. Three experimental samples are used in this paper to show the microstructural features and measured attenuations curves: the A206 alloy, the A206-Al2O3 MMNCs with 1wt.% and 5wt.% of Al2O3 nanoparticles. These samples are fabricated using the ultrasonic cavitation based casting technology [3]. The experimental setup of ultrasonic processing in the casting of A206–Al2O3 MMNCs consists of a resistance heating furnace, an ultrasonic cavitation based processing system (Misonic Sonicator 3000) with a niobium probe of 12.7 mm in diameter and 92 mm in length, a temperature control system and a gas protection system. A graphite crucible with an inner diameter of 88.9 mm and a height of 101.6 mm was used for melting. The ultrasonic probe vibrates with the operating frequency of 20 KHz and power of 4.0 KW. A206 alloy was first melted in the graphite crucible under the protection of argon gas with temperature controlled at 700°C. The γ-Al2O3 nanoparticles with a diameter of 50 nm were then added into the molten melt with ultrasonic cavitation turned on for 15 minutes. Then the molten melt was heated up to 740°C and poured into a steel permanent mold with a preheated
temperature of 400°C. The casted samples are polished for ultrasonic testing. The attenuations were measured using the Olympus Epoch 1000 series NDT device using transducer D785-RP with a nominal central frequency of 2.25 MHz.

2.1. Microstructures and Morphology Modification

The left panel in Fig. 2 shows the representative optical micrographs (top) and polarized light micrographs (bottom) of pure A206 and A206-1wt.% Al₂O₃ nanocomposite [3], and the right panel shows the simulated microstructures. The pure A206 alloy exhibits large dendritic primary α-Al surrounded by continuous θ-Al₂Cu phases. The θ-Al₂Cu phases are distributed along the boundaries of primary aluminum grains and have the morphology of long continuous network. For the nanocomposites with 1wt.% Al₂O₃ nanoparticles, the α-Al dendrites becomes small equiaxed crystals and the intermetallic θ-Al₂Cu phases turn to be smaller, thinner and much less continuous. It indicates that the Al₂O₃ nanoparticles can reduce the grain size of α-Al phase and break or refine the θ-Al₂Cu phase.

![Experimental Microstructure vs. Simulated Microstructure](image)

Fig. 2. Microstructures for pure A206 and A206-Al₂O₃ MMNCs. Left panel: experimental micrographs. Right panel: simulated microstructures.

The formation mechanism of the continuous network of θ-Al₂Cu in A206 and the morphology modification mechanism by Al₂O₃ in A206-Al₂O₃ nanocomposites have been well studied [1, 4, 31, 32]. For the pure A206 alloys, due to the high percentage of Al content, the primary α-Al phases nucleate first and then grow to large dendritic structure during the solidification process.
The Cu solute is pushed out of the α-Al phases into the remaining liquid phase due to the high super-cooling of the θ-Al2Cu nucleation. As the temperature decreases and the content of Cu increases in the remaining liquid, the θ-Al2Cu phase is finally able to nucleate and grow between α-Al dendrites. At last, the θ-Al2Cu phase will form a layer in-between the α-Al dendrites, which is called the divorced eutectic microstructure.

For the A206-Al2O3 nanocomposites, however, the eutectic formation mechanism is modified with the existence of Al2O3 nanoparticles. Similarly, the primary α-Al phases first nucleate and grow in the melt, pushing most of the Al2O3 nanoparticles and Cu to the remaining liquid. The Al2O3 nanoparticles have good nucleant potency and they can serve as effective nucleation sites for θ-Al2Cu to nucleate and grow before the remaining liquid reaches the eutectic composition. While the θ-Al2Cu phases are growing, the liquid surrounding the θ-Al2Cu is enriched with Al due to the depletion of Cu. Consequently, the α-Al phase nucleates and grows on the edges and tips of the θ-Al2Cu, which blocks the growth of θ-Al2Cu. Finally, the partially divorced eutectic phase is formed and both α-Al phase and θ-Al2Cu phase are refined.

2.2. Microstructure Modelling Using Voronoi Diagram

To achieve successful simulations of the ultrasonic wave propagation and reproduce the comparable attenuation curves, the key step is to generate microstructures that can sufficiently capture the microstructural features of A206-Al2O3 MMNCs. The most common method to generate polycrystalline material structure in the computational materials science is the Voronoi diagram or Voronoi tessellation [33-36]. It assigns the same number of points to the space as the desired number of grains, and the space is subsequently divided into many polyhedral based on the closeness to these points. Fig. 3 shows a representative Voronoi diagram where the space is partitioned to 20 cells based on the 20 randomly generated points.
Based on the micrographs of A206-Al2O3 MMNCs (Fig. 2 left panel) and the morphology modification mechanism, we know that the intermetallic network is broken and become thinner and less continuous with the introduction of Al2O3 nanoparticles. The extent to which the intermetallic phase is modified is positively correlated with the amount of Al2O3 nanoparticles [3]. To model this microstructural feature, we first generate a Voronoi diagram with an appropriate number of grains \( N \), and then modify the edges by: 1) randomly selecting some edges, 2) shortening these selected edges, and 3) randomly assigning the width of the remaining edges. After these operations, we obtain a modified Voronoi diagram with edges denoting the intermetallic \( \theta \)-Al2Cu phase and the inner space denoting the primary \( \alpha \)-Al phase. The rationale of this strategy to model the nanocomposites for ultrasonic wave propagation simulation is based on the following considerations and simplifications: 1) It is known that when the grain or inclusion size is less than 1/1000 of the wavelength, its scattering effects on the acoustic wave are negligible [37]. Since the sizes of the dispersed nanoparticles are significantly smaller than the ultrasonic wave length (2~3mm), we neglect the wave scattering by nanoparticles and do not consider nanoparticles in the microstructure simulations; 2) The acoustic scattering arises at the boundaries between grains or inclusions due to the change of material properties. The neighboring \( \alpha \)-Al grains have slightly different material properties because of their different crystal orientations. Therefore the boundaries between \( \alpha \)-Al grains can scatter acoustic waves and contribute to ultrasonic attenuations. However, their scattering effects are negligible compared with those between \( \alpha \)-Al grain and \( \theta \)-Al2Cu phase. Therefore, to simplify the modeling process, the grain boundaries between \( \alpha \)-Al grains are not considered.

To implement the first two operations in the edge modification process, we introduce another two parameters: 1) \( \alpha \), the percentage of edges in the Voronoi diagram to be dissolved, and 2) \( \beta \), the percentage of length left after dissolving. For example, if there are total 100 edges in the diagram, \( \alpha = 0.3 \) and \( \beta = 0.7 \) means 30 edges are randomly selected and each of the selected edges is dissolved to 70% of its original length. Fig. 4 (b) shows a result of applying \( \alpha \) and \( \beta \) to the initial Voronoi diagram in Fig. 4 (a). A well fabricated nanocomposites is featured by small grain size and short intermetallic phases. Therefore we can select large \( N \), large \( \alpha \) and small \( \beta \) to model the microstructures of the good samples. For the experimental microstructures, the thickness
of the intermetallic is not constant. To model this, we add random widths to each edge after the edge dissolving step (Fig. 4 (b)) using the following way (shown in Fig. 4 (d)). We first select \( m \) points with equal interval for each edge, and then assign two points for each selected point along two sides at the same horizontal location with uniformly distributed distances in the vertical direction. After that these assigned points are connected to form a polygon and finally the space within each polygon is used to denote the intermetallic phase. In this width assigning step, the number of middle points \( m \) and the distribution parameters of the random distance can be changed to capture various microstructures. Note that we do not keep the amount of intermetallic phase constant in the morphology modification process. The reason is that the dissolved part of the intermetallic phase is very small in size and has negligible effects on the acoustic attenuation. For simplicity, we do not consider this aspect in the microstructure modeling process. In summary, the microstructure generation procedure is listed in Table 1.

Table 1. The microstructure generation procedure.

<table>
<thead>
<tr>
<th>Input ( N )</th>
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<tbody>
<tr>
<td>1) Generate ( N ) random points.</td>
</tr>
<tr>
<td>2) Partition the pace using ordinary Voronoi diagram based on ( N ) generated points.</td>
</tr>
<tr>
<td>3) Index all edges in the diagram.</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Input ( \alpha, \beta )</th>
</tr>
</thead>
<tbody>
<tr>
<td>4) Randomly select ( \alpha ) percentage edges among all edges.</td>
</tr>
<tr>
<td>5) Shorten the selected edges to ( \beta ) percentage of their original lengths.</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Input ( m, u_l ) and ( u_u )</th>
</tr>
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<tbody>
<tr>
<td>6) Evenly select ( m ) middle points for each edge</td>
</tr>
<tr>
<td>7) Generate one point above each selected point with vertical distance following uniform distribution ( U(u_l, u_u) ).</td>
</tr>
<tr>
<td>8) Generate one point below each selected point with random distance following uniform distribution ( U(u_l, u_u) ).</td>
</tr>
<tr>
<td>9) Connect the generated points to form polygons.</td>
</tr>
<tr>
<td>10) Fill polygons with black color representing the ( \theta )-Al(_2)Cu phase</td>
</tr>
</tbody>
</table>

Fig. 4 shows the overall microstructure modeling process and Fig. 2 (right panel) shows four generated microstructures to simulate pure A206 and A206-Al\(_2\)O\(_3\) MMNCs. In Fig. 5, we present
several simulated microstructures with different parameters $\alpha$, $\beta$ and $N$. Typically, a small $N$ and a large $\beta$ will be chosen for pure A206 and a large $N$ and a small $\beta$ for A206-Al$_2$O$_3$. By adjusting these parameters, we can generate microstructures that are similar to the observed microstructures. For example, the modified Voronoi diagrams are visually similar to observed microstructures in Fig. 2. The similarity can also be roughly quantified. In Fig. 2, the microstructure of pure A206 (Fig. 2a and 2c) and the composite with 1wt% Al$_2$O$_3$ (Fig. 2b and 2d) are simulated using parameter combinations $N=800$, $\beta=0.9, \alpha=1.0$ (Fig. 2e and 2g) and $N=1200$, $\beta=0.7, \alpha=1.0$ (Fig. 2f and 2h), respectively. Through a simple image processing and measure of the optical image, we found that the average grain sizes are $\sim1936 \ \mu m^2$ and $\sim1309 \ \mu m^2$ and the percentage of the dark phase that corresponds to Al$_2$Cu are 10.75% and 5.13% for pure A206 and the composite with 1wt% Al$_2$O$_3$, respectively. In the corresponding simulated microstructure, the average grain sizes are $\sim1849 \ \mu m^2$ and $\sim1156 \ \mu m^2$ and the percentage of the dark phase are 10.02% and 5.05%, respectively. We can see that these measures are close between the observed and the simulated microstructure.

The simulated microstructures will be used as the input in the wave propagation simulation. In the next section, we introduce the EFIT, the acoustic attenuation simulation in details.
Fig. 4. The microstructure modeling process: (a) initial Voronoi diagram, (b) after edge dissolving step controlled by $\alpha$ and $\beta$, (c) after assigning random thickness to each edge, (d) the random thickness assigning process.

3. Wave Propagation Simulation using EFIT

The EFIT is a very stable and efficient numerical scheme to model wave propagation in homogeneous and heterogeneous, isotropic and anisotropic elastic media. It was first developed by Fellinger et al [23], and since then it has been widely used to explore elastic wave behaviors in a variety of applications [28, 38]. The EFIT uses velocity-stress formalism on a staggered spatial and temporal grid. It discretizes the following first-order equations:

$$\int\int\int_{V} \frac{\partial}{\partial t} p(\mathbf{r}, t) dV = \oint_{S} \mathbf{n} \cdot \mathbf{T}(\mathbf{r}, t) dS + \int\int\int_{V} f(r, t) dV, \quad (1)$$

$$\int\int\int_{V} \frac{\partial}{\partial t} S(\mathbf{r}, t) dV = \oint_{S} \text{sym}\{\mathbf{n} \cdot \mathbf{v}(\mathbf{r}, t)\} dS + \int\int\int_{V} \mathbf{h}(\mathbf{r}, t) dV. \quad (2)$$

$p$ is the momentum density vector, $\mathbf{T}$ the stress second rank tensor, $\mathbf{S}$ is the strain second rank tensor, $\mathbf{v}$ is the particle velocity vector, $f$ is the source of force density, $\mathbf{h}$ is the source of deformation rate second rank tensor, $\mathbf{n}$ is the outward normal unit vector of $S$ and $\text{sym}\{\mathbf{n} \cdot \mathbf{v}(\mathbf{r}, t)\}$ denotes the symmetric part of the dyad $\{\mathbf{n} \cdot \mathbf{v}(\mathbf{r}, t)\}$. More detailed explanation of (1) and (2) can be found in [39].
In our research, we employ the existing code Visco-Elastodynamic Finite Integration Wave Solver (VEFIT) [40], which is written in C with interface with MATLAB. The VEFIT uses EFIT equations (see Appendix A for details) to solve wave propagation in media. The VEFIT requires a user-defined phantom (i.e., a 2D geometry which can be homogeneous or inhomogeneous), the phantom parameters (i.e., the density of material, the normal and shear velocity of the ultrasonic wave in the media, the bulk viscosity and shear viscosity) and the transducer parameters including the position and size of transducers and the excitation signals as the inputs. The outputs include the stress, the velocity, the acceleration at any selected locations, and the transducers outputs recording the velocity received by transducers at each time step. The transducer outputs will be used to calculate the acoustic attenuation. Example of input phantom, the wave propagation and the transducers output generated by VEFIT are presented in Fig. 6.
In Fig. 6, the transducer output shows the waveforms of initial pulse and received echo. The two waveforms are extracted using a rectangular window with the same size. The frequency spectra are obtained by performing the fast Fourier transform (FFT) on the extracted signals. The attenuation can be calculated using the spectral ratio analysis technique [41] as:

$$A(f) = \frac{1}{2d} \ln \left( \frac{S_1(f)}{S_2(f)} \right),$$  \hspace{1cm} (3)

where $A(f)$ is the attenuation coefficient at frequency $f$, $d$ is the thickness of the media, $S_1(f)$ is the frequency spectra calculated using FFT on the extracted signals. $S_1(f)$ is calculated from the incident wave $S_1(t)$ and $S_2(f)$ is from the first bounced back wave $S_2(t)$.

![Fig. 6. Examples of input phantom, wave propagation snapshots and transducer output by VEFIT.](image)

The overall simulation procedure is presented in Fig. 7. The material properties in the phantom need to be determined. For Al-Cu alloy A206, the main chemical compositions are Al (93.5%-95.3%) and Cu (4.2%-5.0%). The acoustic properties are calculated based on its elastic properties i.e., Young’s modulus, Poisson's ratio and density, which are found in [48, 49]. The phantom parameters are summarized in Table 2. In the wave propagation simulation, the transducer is placed in the middle of the left side of the microstructure, as shown in Fig. 6. The size of the
microstructure is selected as 1.2mm × 1.2mm and the size of the transducer is selected as one sixth of the length of left side of the microstructure. The central frequency of the excitation signals is set to be 2.25MHz, the same as used in the experiment [3]. The boundaries are specified to be absorbing in the top and bottom sides and reflective in the left and right sides. In the next section, the simulation results will be discussed and compared with experimental data.

Table 2. Phantom parameters of Al2Cu and Al

<table>
<thead>
<tr>
<th></th>
<th>Density (g/mm³)</th>
<th>Normal velocity (m/s)</th>
<th>Shear velocity (m/s)</th>
<th>Bulk viscosity</th>
<th>Shear viscosity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al2Cu</td>
<td>0.00436</td>
<td>5945</td>
<td>2892</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Al</td>
<td>0.0027</td>
<td>6420</td>
<td>3040</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

4. Simulation and Experimental Results

In the first set of simulation, we fix α = 1.0, i.e., we dissolve all edges of the initial Voronoi diagram. The number N of cells ranges from 800 to 1600. N = 800 is approximately the number of grains in pure A206 in the space of 1.2mm × 1.2 mm. N = 1600 is roughly the number of
grains in A206 nanocomposites of the same dimension size with 5wt.% Al₂O₃ nanoparticles. β is chosen from 0.9, 0.7 and 0.5. For every combination of N and β, 20 microstructures are randomly generated for wave propagation simulation. Since the Voronoi diagram is regenerated for each simulation, we expect different attenuation curves for each replication.

![Experimental Measurement vs Simulation Results](Image)

**Fig. 8.** The comparison of experimental attenuation curves and the simulated attenuation curves with different simulation parameters (attenuation units: dB/mm, frequency unit: MHz).

Fig. 8 shows the experimentally measured attenuation curves from [3] and the simulated attenuation curves using different microstructural parameters. In Fig. 8, (a), (b) and (c) show experimental attenuation curves measured at 20 randomly selected locations on each sample using the Olympus Epoch 1000 series NDT device with transducer D785-RP of 6mm in diameter and of 2.25MHz in nominal central frequency. Based on the experimental results, Wu et al [3] stated that there are three sources that influence the material homogeneity and cause the between-curve variations, the large α-Al dendrites, the long and continuous intermetallic Al₂Cu phases, and the non-fully dispersed Al₂O₃ clusters. Well dispersed Al₂O₃ nanoparticles can enhance the nucleation of both α-Al and Al₂Cu phases, resulting in more homogeneous materials. In the simulation we do...
not consider Al₂O₃ clusters. Only the grain size of α-Al phase and morphology of Al₂Cu phase influence the attenuation curves. From Fig. 8 we can see that increasing N or decreasing β can reduce the variation of attenuation curves, which is consistent with the experimental results, for that larger N or smaller β indicates a more homogeneous material. Besides, the trend and mean value of the attenuation curves are also quite similar to the experimentally measurements. For the pure A206 alloy, the attenuation decreases with frequency, while for the Al₂O₃ reinforced nanocomposites, the attenuation is more severe for acoustic wave of higher frequency. Therefore, the simulation approach is capable of reproducing the characteristics of the attenuation measurements.

To investigate the influence of parameters α and β on the attenuation curves, we fix N = 1200 and run the simulation with different α and β, as shown in Fig. 9. From the simulation results we observe the following phenomenon: 1) For a fixed β, as α increases from 0 to 1, the attenuation curves tend to be more uniform. This is what we expect. Since α controls the percentage of edges being dissolved, more edges dissolved as α increasing result in the more homogenous microstructures; 2) For a larger value β, increasing α will change the attenuation curves less significantly. In the extreme case, if there is no dissolving at all, i.e., β = 1.0, then the change of α will not influence the attenuation curves. Similarly, for a smaller α, the change of β can hardly influence the attenuation curves; 3) For fixed α or β, the decreasing of β or increasing of α will result in the down shift of the attenuation curves. It is because the decreasing of β or increasing of α for fixed α or β will reduce the amount of the intermetallic phase in the microstructure, thus reducing the wave scattering effects. In the next section, we will investigate the similarity of the between-curve variation between experimental and simulated attenuation curves.
5. Statistical Comparison of Experimental and Simulated Attenuation

Both the experiment and simulation show that the between-curve variation can be used to measure the homogeneity of the A206-Al₂O₃ MMNCs. Therefore it is important to quantify this variation and compare it between experiment and simulation for future statistical quality control tool development. From Fig. 8 and Fig. 9 we can see that all the attenuation curves of each sample share the similar characteristics (e.g., slope, intercept). On the other hand, the variation from curve to curve also exist for each sample. Therefore, it is natural to select the linear mixed-effects model [42] to describe the population-level features and also model the variation among replicated attenuation curves.

Let $y_{ij}$ denote the acoustic attenuation coefficient for $i$-th curve at $j$-th frequency $f_j$. $a$ and $b$ represent the fixed intercept and slope of the regression line respectively. The linear mixed effects model with first order polynomial in the fixed effects can be written as:

$$y_{ij} = a + bf_j + a_i + b_if_j + \epsilon_{ij}, i = 1,2,\ldots, n, j = 1,2,\ldots, m,$$  \hspace{1cm} (4)
where \(a_i\) and \(b_i\) are random effects of the intercept and slope for \(i\)-th curve with the assumption 
\[
\begin{bmatrix} a_i \\ b_i \end{bmatrix} \sim N(0, \Sigma), \quad \Sigma = \begin{bmatrix} \sigma_1^2 & \rho \sigma_1 \sigma_2 \\ \rho \sigma_1 \sigma_2 & \sigma_2^2 \end{bmatrix},
\]
and \(\epsilon_{ij} \sim N(0, \sigma^2)\) is the error term which is used to model the measurement error or model inadequacy, and is assumed to be independently and identically distributed for all attenuation curves. In this model there are two parts, fixed effects and random effects. Fixed-effects term \(a + bf\) is the conventional linear regression part used to describe the population-level mean attenuation curve. The random-effects term \(a_i + b_if\) is associated with individual measurement \(i\) and is used to describe its deviation from the mean attenuation curve. Note that we can alternatively use higher order polynomial in the linear mixed effects model. However, this may result in over-fitting issue. The attenuation curves in Fig. 8 and Fig. 9 show a good linear relationship with frequency \(f\), therefore first order polynomial is sufficient in the model fitting.

The model parameters can be estimated using maximum likelihood estimation (MLE) method. Suppose \(y_i = (y_{i1}, y_{i2}, ..., y_{im})\). Denote \(\beta = (a, b)^T\), \(b_i = (a_i, b_i)^T\), and \(\theta = (\beta, \sigma^2, \Sigma)\), then the likelihood function is
\[
L(\theta | y_1, ... y_n) = p(y_1, ... y_n | \theta) = \prod_{i=1}^{n} p(y_i | \theta) = \prod_{i=1}^{n} \int p(y_i | \theta, b_i) p(b_i | \theta) \, db_i
\]
where
\[
p(y_i | \theta, b_i) = (2\pi \sigma^2)^{-m/2} \exp \left( -\frac{1}{2\sigma^2} \| y_i - X_i(\beta + b_i) \|^2 \right)
\]
\[
p(b_i | \theta) = (2\pi)^{-1/2} |\Sigma|^{-1/2} \exp(-b_i^T \Sigma^{-1} b_i/2)
\]
\[
X_i = [1, 1, ..., 1; f_1, f_2, ..., f_m]^T
\]
By integrating out \(b_i\) we can get
\[
L(\theta | y_1, ... y_n) = \prod_{i=1}^{n} \left( 2\pi \right)^{m/2} |X_i\Sigma X_i^T + \sigma^2 I|^{-1/2} \exp \left( -\frac{1}{2} (y_i - X_i \beta)^T (X_i \Sigma X_i^T + \sigma^2 I)^{-1} (y_i - X_i \beta) \right)
\]
By maximizing \(L(\theta | y_1, ... y_n)\) with respect to \(\beta\) and \(\Sigma\), we can obtain the MLE estimated model parameters. The optimization details can be found in [43].

Table 3 shows the fitting results for the experimental measurement of A206-5wt.%Al2O3 and attenuation curves shown in Fig. 8 (c3). We select the attenuation curves in Fig. 8 (c3) here as an example due to its visual similarity to the experimental measurements of A206-5wt.%Al2O3. From
this table we can see that the fitted results for the simulation data are quite close to the experimental data.

Table 3. Fitting results for experimental attenuation curves of A206-5wt.% Al₂O₃ and attenuation curves shown in Fig. 8 (c3). “Lower” and “Upper” are the lower and upper bound of the 95% confidence interval.

<table>
<thead>
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<th>Simulation data shown in Fig. 8 (c3)</th>
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<tr>
<td></td>
<td>Fixed Estimate</td>
<td>S.E</td>
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<tr>
<td>a</td>
<td>-1.00</td>
<td>0.024</td>
</tr>
<tr>
<td>b</td>
<td>0.55</td>
<td>0.011</td>
</tr>
</tbody>
</table>

The histograms of the fitted random effects and the residuals are shown in Fig. 10. The fitted $a_i$, $b_i$ and $\epsilon_i$ approximately follow normal distribution, which validates the model assumption of $a_i$ and $b_i$. From these histograms, we see the linear mixed effects model can model the attenuation curves well.

Fig. 10. Histograms of the fitted random effects and residuals for the experimental measurements of A206-5wt.% Al₂O₃ (top) and simulated attenuation curves shown in Fig. 8 (c3) (bottom).
To test the similarity between the simulation and the experimental measurement in terms of the non-uniformity of the acoustic attenuation, we need to compare the covariance matrix $\boldsymbol{\Sigma}^{(s)}$ and $\boldsymbol{\Sigma}^{(r)}$ calculated in the model fitting, where $\boldsymbol{\Sigma}^{(s)}$ is for the simulation and $\boldsymbol{\Sigma}^{(r)}$ is for the experimental measurement. Testing if the covariance matrices of different groups of dataset are equal has been well studied, where the likelihood ratio test is the most commonly used methodology [42]. However, these studies focus on the covariance matrices of the observations, whose dimension would be very high for attenuation curves. Instead, we focus on the covariance matrix of model parameters $(a_i, b_i)^T$ with significantly reduced dimension. Therefore the likelihood ratio test cannot be directly applied in this study. We employ the permutation test [40], a non-parametric and computationally efficient method to tackle this issue. Intuitively, if two sets of data (e.g., the acoustic attenuation curves from simulation and experimental measurements) are similar enough to each other, i.e., they come from the same distribution, then by randomly shuffling the data components between the two data sets, we expect to see the similar statistics (e.g., the difference of means or variances between the original two sets and the sets after random shuffling). We may now test how similar the two datasets by comparing the statistics before and after the random permutation. To make the comparison more rigorous, repeat the permutation process many times to get the sample distribution of the test statistic and calculate the $p$ value for the statistic of the original data sets. An example of permutation test on testing means of two data sets is presented in Fig. 11. Suppose we want to test if the means are equal for two datasets $X_1$ and $X_2$, which are generated from the same uniform distribution. The values in each set are randomly generated just for illustration purpose. By randomly shuffling the components in the two sets, we obtain $X_1^*$ and $X_2^*$. The difference of the mean of the new sets is calculated. Repeat the permutation 1000 times to get the sample distribution of $|\bar{X}_1^* - \bar{X}_2^*|$. Calculate the $p$-value of test statistic of the original data sets. Here the $p$-value is the percentage of the generated 1000 samples that satisfy $|\bar{X}_1 - \bar{X}_2| \geq |\bar{X}_1^* - \bar{X}_2^*|$. If the $p$-value is very small, it is likely that $X_1$ and $X_2$ have different means. In Fig. 11 the $p$-value is about 0.9, therefore the two means are likely to be equal.
In our case, the null hypothesis is $H_0: \Sigma^{(s)} = \Sigma^{(r)}$, which is equivalent to $H_0: \sigma_1^{(s)}(s) = \sigma_1^{(r)}(r)$, $\sigma_2^{(s)}(s) = \sigma_2^{(r)}(r)$, and $\rho^{(s)}(s) = \rho^{(r)}(r)$. In this testing there are simultaneously three components to be tested. To avoid the inconvenience of multiple testing problems [44], we reformulate it to an identical hypothesis testing $H_0: T = \max\{ |\sigma_1^{(s)} - \sigma_1^{(r)}|, |\sigma_2^{(s)} - \sigma_2^{(r)}|, |\rho^{(s)} - \rho^{(r)}| \} = 0$. In other words, the hypothesis testing of equality of the two covariance matrices is performed by testing if the maximum difference of the matrix entries is zero. Considering that the sample variances of the three absolute differences in the testing statistic $T$ may be different in the permutation test, it is necessary to standardize these three terms first by dividing their standard deviations (SD). Therefore the hypothesis test can be expressed as

$$H_0: T = \max\left\{ \frac{|\sigma_1^{(s)} - \sigma_1^{(r)}|}{SD|\sigma_1^{(s)} - \sigma_1^{(r)}|}, \frac{|\sigma_2^{(s)} - \sigma_2^{(r)}|}{SD|\sigma_2^{(s)} - \sigma_2^{(r)}|}, \frac{|\rho^{(s)} - \rho^{(r)}|}{SD|\rho^{(s)} - \rho^{(r)}|} \right\} = 0$$

$$H_1: T \neq 0$$

The permutation test can be summarized as: (1) random shuffle curves between the simulation set and the experimental measurement set and fit the linear mixed effects model to each new dataset to get $\Sigma_p^{(s)}$ and $\Sigma_p^{(r)}$; (2) calculate the absolute differences of $\sigma_1$, $\sigma_2$ and $\rho$ between the two
covariance matrices; (3) repeat (1) and (2) \( N_s \) times to obtain the three sets of samples for 
\[
\left| \sigma_1^{(s)} - \sigma_1^{(r)} \right|, \left| \sigma_2^{(s)} - \sigma_2^{(r)} \right| \text{ and } \left| \rho^{(s)} - \rho^{(r)} \right| ;
\]
(4) calculate their standard deviations and standardize these samples; (5) calculate the \( T \) statistic for these samples to obtain a set of samples \( \{T_1, \ldots, T_{N_s}\} \); (6) calculate the standardized \( T \) statistic for the observations (i.e., simulation set and experimental measurement before random shuffle) \( T_o \); (7) calculate the p-value (the percentage of \( T \) samples that are larger than \( T_o \)) for the observations. If the p-value is smaller than a certain threshold, e.g., 0.1, then reject the null hypothesis \( H_0 \), i.e., \( \Sigma^{(s)} \neq \Sigma^{(r)} \).

Fig. 12 shows two examples of the permutation test, where we compare the covariance matrices between Fig. 8 (c) (A206+5wt.% Al\(_2\)O\(_3\)) and Fig. 8 (c3), Fig. 8 (b) (A206+1wt.% Al\(_2\)O\(_3\)) and Fig. 8 (c3). The p-values for these two tests are 0.99 and 0.08 respectively. If we select the 0.1 as the testing threshold, then we can conclude that the variation of attenuation curves in Fig. 8 (b) is statistically different from Fig. 8 (c3), and Fig. 8 (c) is quite similar to Fig. 8 (c3) in terms of the between-curve variation. Also, we can use the p-value to evaluate the similarity of the covariance matrices of two sets of attenuation curves. The higher the p-value, the closer the two sets of attenuation curves. On the other hand, small p-values (\( \leq 0.1 \)) indicate a large difference between simulated attenuation curves and the experimental measurements due to mismatched microstructures. The p-value are 0.24, 0.71 and 0.99 for the comparisons between Fig. 8 (a) and Fig. 8 (a1), Fig. 8 (b) and Fig. 8 (b2), Fig. 8 (c) and Fig. 8 (c3), respectively. These large p-values (>0.1) indicate that the simulated microstructures can well reproduce the attenuation variation.

6. Discussion and Conclusion

In this research, we propose a microstructure modelling and wave propagation simulation method to generate the microstructures and to simulate the ultrasonic attenuation curves for A206-Al\(_2\)O\(_3\) MMNCs. Based on the micrographs and morphology modification mechanism of the nanocomposites, a modified Voronoi diagram is developed to simulate 2D microstructures and capture the microstructural features, where three key parameters are used to control the grain size of the primary phases and the morphology modification of the intermetallic phases. The numeric method EFIT is used to simulate the wave propagation through the generated microstructures. The attenuation curves are calculated by performing the fast Fourier transform (FFT) on the extracted signals from the outputs of EFIT. The simulated acoustic attenuation curves are quite consistent with the experimental measurements.
Fig. 12. Illustration of the permutation test. (a) and (b): Fig. 8 (c) VS. Fig. 8 (c3), p-value=0.99; (c) and (d): Fig. 8 (b) VS. Fig. 8 (c3), p-value=0.06. The vertical dashed lines denote the observed test statistics.

The linear mixed effects model is used to model the attenuation curves. A permutation test based on the maximum difference of each matrix entry is developed to test the equality of the covariance matrices from the simulated and experimental attenuation curves. The hypothesis tests show that by adjusting the microstructural parameters of the simulation, the simulated attenuation curves are able to closely match the experimental measurement in terms of the between-curve variations.

This research directly supports the experimental results and findings in [3] through the simulation approach. It helps us better understand the phenomenon of the non-uniformity of the attenuation curves and how the microstructural features influence this non-uniformity. In the future, the quantitative model will be conducted to infer the microstructural features based on the experimental and simulation database of different microstructures and the corresponding attenuations. Using that model, the statistical process control (SPC) charts will be developed to control the quality of A206-Al$_2$O$_3$ MMNCs based on the attenuation profiles or inferred microstructural features. Although 2D simulation has been commonly used, it may be not as accurate as 3D simulations. Therefore it is desirable to extend the simulation to three dimensions.
to better represent the experimental microstructures and wave propagation processes, which will also be our future work.

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References


The 2D EFIT equations of (1) and (2) are presented as

\[ v^n_{x,i,j} = v^{n-1}_{x,i,j} + \frac{\Delta t}{\Delta x} B_x \left( \tau^{n-\frac{1}{2}}_{xx,i+1,j} - \tau^{n-\frac{1}{2}}_{xx,i,j} + \tau^{n-\frac{1}{2}}_{xy,i,j} - \tau^{n-\frac{1}{2}}_{xy,i,j-1} \right) \]  

(5)

\[ v^n_{y,i,j} = v^{n-1}_{y,i,j} + \frac{\Delta t}{\Delta y} B_y \left( \tau^{n-\frac{1}{2}}_{xy,i,j} - \tau^{n-\frac{1}{2}}_{xy,i,j-1} + \tau^{n-\frac{1}{2}}_{yy,i,j+1} - \tau^{n-\frac{1}{2}}_{yy,i,j} \right) \]  

(6)

\[ \tau^{n+\frac{1}{2}}_{xx,i,j} = \tau^{n-\frac{1}{2}}_{xx,i,j} + \frac{\Delta t}{\Delta x} \left( (\lambda + 2\mu) \left[ v^n_{x,i,j} - v^n_{x,i-1,j} \right] + \lambda \left[ v^n_{y,i,j} - v^n_{y,i,j-1} \right] \right) \]  

(7)

\[ + (\eta + 2\phi) \left[ \hat{v}^n_{x,i,j} - \hat{v}^n_{x,i-1,j} \right] + \eta \left[ \hat{v}^n_{y,i,j} - \hat{v}^n_{y,i,j-1} \right] \]

\[ \tau^{n+\frac{1}{2}}_{yy,i,j} = \tau^{n-\frac{1}{2}}_{yy,i,j} + \frac{\Delta t}{\Delta y} \left( (\lambda + 2\mu) \left[ v^n_{y,i,j} - v^n_{y,i-1,j} \right] + \lambda \left[ v^n_{x,i,j} - v^n_{x,i-1,j} \right] \right) \]  

(8)

\[ + (\eta + 2\phi) \left[ \hat{v}^n_{y,i,j} - \hat{v}^n_{y,i-1,j} \right] + \eta \left[ \hat{v}^n_{x,i,j} - \hat{v}^n_{x,i,j-1} \right] \]

\[ \tau^{n+\frac{1}{2}}_{xy,i,j} = \tau^{n-\frac{1}{2}}_{xy,i,j} + \frac{\Delta t}{\Delta x} \left( \mu_{xy} \left[ v^n_{x,i,j+1} - v^n_{x,i,j} + v^n_{y,i+1,j} - v^n_{y,i,j} \right] \right) \]  

(9)

where the velocities \((v_x, v_y)\) at time \(n\) are calculated using the sum of the velocities at time \(n - 1\) and a linear combination of the stresses \((\tau_{xx}, \tau_{yy}, \tau_{xy})\) in the spatial coordinates \(i, j\) and the half
step time $n - \frac{1}{2}$. Similarly, the stresses ($\tau_{xx}, \tau_{yy}, \tau_{xy}$) at time $n + \frac{1}{2}$ are calculated by adding the stresses at $n - \frac{1}{2}$ and the linear combination of velocities multiplied by the lame constants $\lambda$ and $\mu$ for the elastic factor and the viscous factor $\eta$ and $\phi$ multiplied by the rate of change in velocities (denoted as $(\dot{v}_x, \dot{v}_y)$). $\Delta x$ and $\Delta t$ represent spatial step and time step respectively. $B_x$ and $B_y$ are the effective buoyancies defined as:

$$B_x = \frac{2}{\rho_{i+1,j} + \rho_{i,j}},$$
$$B_y = \frac{2}{\rho_{i,j+1} + \rho_{i,j}},$$

where $\rho$ is the mass density for the spatial coordinates $i,j$. $\mu_{xy}$ and $\nu_{xy}$ are the effective rigidity defined as:

$$\mu_{xy} = \frac{4}{\mu_{i,j} + \mu_{i+1,j} + \mu_{i,j+1} + \mu_{i+1,j+1}},$$
$$\nu_{xy} = \frac{4}{\nu_{i,j} + \nu_{i+1,j} + \nu_{i,j+1} + \nu_{i+1,j+1}}.$$

The spatial resolution $\Delta x$ and the time resolution $\Delta t$ must be chosen small enough to provide sufficiently smooth representations of the computed filed. However, these two resolutions cannot be chosen independently, they must satisfy the Courant’s stability condition, that is:

$$\Delta t \leq \frac{\Delta x}{c_{\text{max}} \sqrt{d}},$$

where $d$ is the space dimension ($d = 2$ is used for our simulation) and $c_{\text{max}}$ is the largest wave speed in the media. $\Delta x$ is recommended ranging from $\frac{\lambda}{8}$ to $\frac{\lambda}{20}$ for the wavelength $\lambda$ [42].
Figure Captions List

Fig. 1  Illustration of the ultrasonic testing using ultrasonic attenuation curves [3].

Fig. 2  Microstructures for pure A206 and A206-Al2O3 MMNCs. Left panel: experimental micrographs. Right panel: simulated microstructures.

Fig. 3  Example of Voronoi diagram with 20 random points.

Fig. 4  The microstructure modeling process: (a) initial Voronoi diagram, (b) after edge dissolving step controlled by $\alpha$ and $\beta$, (c) after assigning random thickness to each edge, (d) the random thickness assigning process.

Fig. 5  Microstructures generated using different parameters $\alpha$, $\beta$ and $N$.

Fig. 6  Examples of input phantom, wave propagation snapshots and transducer output by VEFIT.

Fig. 7  Simulation procedure using VEFIT and attenuation measurement.

Fig. 8  The comparison of experimental attenuation curves and the simulated attenuation curves with different simulation parameters (attenuation units: dB/mm, frequency unit: MHz).

Fig. 9  The influence of $\alpha$ and $\beta$ on the attenuation curves ($N=1200$).

Fig. 10  Histograms of the fitted random effects and residuals for the experimental measurements of A206-5wt.% Al2O3 (top) and simulated attenuation curves shown in Fig. 8 (c3) (bottom).

Fig. 11  Illustration of permutation test on population means of two data sets.

Fig. 12  Illustration of the permutation test. (a) and (b): Fig. 8 (c) VS. Fig. 8 (c3), p-value=0.99; (c) and (d): Fig. 8 (b) VS. Fig. 8 (c3), p-value=0.06. The vertical dashed lines denote the observed test statistics.
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