



Review article

Interface-derived solid-state viscoelasticity exhibited by nanostructured and microstructured materials containing carbons or ceramics

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ABSTRACT

With 206 references, this paper reviews the field of interface-derived solid-state viscoelasticity that emerged in 1995 and grew with nanotechnology. Viscoelasticity is relevant to vibration and acoustic damping, as needed for cement-based composites, continuous fiber polymer-matrix composites, etc. This viscoelasticity mechanism involves interfacial friction and requires adequate interface area and feasibility of slight interfacial sliding. This mechanism is in contrast to the conventional viscoelasticity that involves bulk viscoelastic deformation, as in rubber. Compared to the conventional mechanism, this mechanism is attractive for the relative independence on temperature, the ability to withstand elevated temperatures, and the feasibility of simultaneously enhancing the loss tangent and stiffness. In monolithic materials, the interfaces are primarily the filler-matrix interface. Concerning multi-walled carbon nanotubes, the wall-wall interface also contributes. For providing interfaces, nanostructured/microstructured carbon or ceramic fillers (nanotubes/nanofibers, exfoliated graphite, ceramic particles, smectite clay, etc.) are effective, provided that their dispersion and interface design are adequate. For continuous fiber polymer-matrix composites, the filler volume fraction should be minimized. For non-monolithic materials (assemblies without a matrix, including fibers, nanotubes, exfoliated graphite and carbon black), which are potentially useful as the constrained layer in constrained-layer damping, the interfaces are primarily those between the structural units of the assembly.

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1. Introduction

Viscoelasticity is mechanical behavior that is a combination of

elastic behavior and viscous behavior. The elastic character is typical for a stiff material, like a spring [1]. The viscous character is typical for a fluid or a material that can flow, as in a dashpot [1], and results in mechanical energy loss, which is attractive for vibration damping (mechanical energy dissipation) and acoustic damping (sound absorption). Vibration damping is valuable for normal

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structural operation, as needed for bridges, high-rise buildings, pavement, aircraft, automobile, railroad sleepers, tennis rackets, skiboards, etc. However, it is also relevant to extreme situations such as earthquakes and hurricanes. Sound absorption is important for environmental noise pollution reduction, as needed for aircraft, rail, highways, factories, etc. Because research on acoustic damping has received much less attention than that on vibration damping, the damping addressed in this paper is vibration damping, unless noted otherwise. Moreover, the damping addressed here is passive damping (which is based on the material behavior and is also known as structural damping) rather than active damping (which involves sensors and actuators working together). In general, strong viscous behavior is not always attractive. For example, creep and stress relaxation [2] are undesirable phenomena that result from the viscous character.

A viscoelastic solid exhibits degrees of both elastic and viscous characters. A classical example of a viscoelastic material is rubber [3]. Polymers tend to be viscoelastic [4–6], particularly above the glass transition temperature [7], with the viscous character stemming from the ability of the molecules or molecular segments in the polymer to move in response to stress application. Elastomers such as rubber are particularly strong in the viscous character, because of the long length and kinked conformation of the molecules [8,9]. In fact, all commercially available viscoelastic materials are polymeric. The associated viscoelastic behavior is a bulk effect, with the bulk (volume) of the material responding to the dynamic applied stress through molecular movement in the volume. Flow behavior is enabled by and mechanical energy is dissipated through the movement.

Due to the viscoelastic character of the polymer matrix, polymer-matrix composites tend to exhibit a degree of viscoelastic behavior. However, in the presence of continuous fibers (particularly stiff fibers such as carbon fibers) as the reinforcement in the composite [10], the elastic behavior is dominant in the fiber direction of the composite, while viscoelastic behavior can occur to a degree in the off-axis directions of the composite [11]. In case of a unidirectional composite, the behavior in the transverse direction is dominated by the polymer matrix [12].

A thermoset matrix such as epoxy tends to be more brittle than a thermoplastic matrix. In case of discontinuous reinforcement (short fibers or particles) and a thermoplastic matrix, the viscous character can be substantial [13,14]. An example of a thermoplastic matrix is pitch, which is used in asphalt for pavements [15]. The viscoelastic behavior of the pavement material is important for both vibration damping and noise reduction [16].

Metals are elastic materials, unless they are at a temperature that is high enough for the atoms to move upon stress application. The minimum temperature for this to occur is typically approximately equal to about 1/3 of the melting temperature in K. Along with the viscous behavior in this temperature range is low stiffness, which is not suitable for structural applications.

A less well-known mechanism of viscoelastic behavior involves interfacial friction rather than the abovementioned bulk viscoelastic deformation. The relevant interfaces are within the material. The interfacial mechanism gives appreciable mechanical energy dissipation only if the interfaces are abundant and capable of undergoing frictional sliding. The attraction of interface-derived viscoelasticity is that the interface may enhance the stiffness, in addition to enhancing the viscous character, depending on the interface design. As a consequence, it is possible that the interface enhances both the elastic and viscous behavior. On the other hand, in the classical bulk viscoelastic deformation mechanism, the easier is the molecular movement, the greater is the viscous character and the lower is the elastic character (the stiffness). In other words, in case of the bulk viscoelastic deformation mechanism, enhancement

of the viscous character occurs at the expense of the stiffness.

Stiffness is not only important for structural performance, it helps the mechanical energy dissipation. This is because a soft material cannot sustain much force and energy relates to the product of force and distance (deformation), as further explained below.

Both vibrations and sound are associated with dynamic mechanical waves. The dynamic mechanical properties are described by the storage modulus (elastic modulus under dynamic loading), which relates to the elastic character, and the loss tangent ($\tan \delta$), which relates to the viscous character. The angle δ is zero for a perfectly elastic material (with no viscous character), whereas δ is equal to 90° for a perfectly viscous material (with no elastic character). The product of the storage modulus and the loss tangent is the loss modulus, which is the viscous modulus that relates to the mechanical energy dissipation. In other words, the loss modulus should be high. For this purpose, both the storage modulus and loss tangent are preferably high.

Because a viscoelastic material based on the bulk viscoelastic deformation mechanism is not stiff enough, it is commonly sandwiched by two stiff sheets, such as two continuous fiber polymer-matrix composite sheets. Upon shear deformation of the sandwich, the viscoelastic material deforms, thereby dissipating mechanical energy. On the other hand, the stiff sandwiching sheets provide adequate structural performance to the sandwich. This sandwich approach is known as constrained-layer damping, in which the viscoelastic layer is constrained by the stiff sandwiching sheets. A related but less effective method involves constraining the viscoelastic layer on one side only, as provided by using a single stiff sheet.

It is desirable for a structural material to have vibration damping ability. This is because a structure contains a large volume of structural material and, for a given material, the mechanical energy dissipation is proportional to the volume. In fact, the hysteresis area in the stress-strain curve corresponds to the mechanical energy loss per unit volume. Thus, even if the loss modulus of a structural material is not high, the structure can dissipate much energy due to its large volume of structural material. In contrast, the viscoelastic material in constrained-layer damping is not a structural material; though it may have strong viscous character, it occupies a small volume compared to the volume of the structure.

Another problem with the bulk viscoelastic deformation mechanism is that the viscous character is sensitive to the temperature [17]. In other words, the ease and manner of movement of the molecules are strongly dependent on the temperature. For example, a viscoelastic material that works well at room temperature may not work well just at 50°C [18]. Furthermore, a polymer cannot resist high temperatures. In contrast, the interfacial mechanism of viscoelasticity does not involve molecular movement and the associated viscous character is relatively independent of the temperature.

The rendering of the vibration damping function to a structural material makes the material multifunctional. This is to be distinguished from the incorporation of damping devices (such as devices involving constrained-layer damping) in the structure. The interfacial mechanism of viscoelasticity has been shown to be capable to render damping function to a structural material, as discussed in this review. In some cases, both the storage modulus and loss tangent of the structural material are increased by using this mechanism.

Liquids tend to be dominated by the viscous character, with little elastic character [19–22]. The viscoelastic behavior of liquids is important for inks, biological systems and industrial processes. However, this review addresses solids rather than liquids.

This paper is a review of interface-derived solid-state

viscoelasticity, with consideration of the materials design and behavior. Partly due to the explosive growth of the field of nanotechnology, this subject has been increasingly reported. The first report was in 1995, when Hudnut and Chung reported interface-derived viscoelasticity in a continuous carbon fiber polymer-matrix composite with the incorporation of carbon nanofibers (CNFs) [12]. The term “carbon filaments” instead of “carbon nanofibers” was used by Hudnut and Chung [12], because the former term is the original term for this material and the latter term had not yet been accepted. This 1995 work of Hudnut and Chung is to be distinguished from their 1996 report of the incorporation of tin-zinc alloy particles of size 25–45 μm in a continuous carbon fiber polymer-matrix composite for damping enhancement that involves the conventional bulk viscoelastic deformation mechanism [23]. Separately, in 1996, Fu and Chung reported interface-derived viscoelasticity in a cement-matrix composite containing silica microparticles (silica fume or microsilia) [24]. Since 1995 and 1996, this form of viscoelasticity has been reported for a large variety of materials, partly due to the explosive growth of nanotechnology, thus warranting a review of this subject.

This review covers the small-strain regime of viscoelasticity, as is relevant to normal structural operation, which occurs in the elastic regime of deformation. In contrast, extreme events such as earthquakes and hurricanes involve deformation in the plastic regime of deformation. From a scientific research viewpoint, it is important to address the viscoelastic behavior in the elastic regime prior to addressing the behavior in the plastic regime, which involves the plasticity of the material.

The relevant materials covered in this review include monolithic and non-monolithic materials. They are nanostructured and microstructured materials containing carbons and/or ceramics. The growth of the field of interface-derived solid-state viscoelasticity occurred along with the growth of the field of nanotechnology.

The monolithic materials that exhibit interface-derived viscoelasticity are structural materials and include cement-based materials (important for civil structures), continuous fiber polymer-matrix composites (important for lightweight structures such as airframe) and polymer-matrix composites with discontinuous fillers (relevant to mechanically less demanding structures and to structures requiring electrical conductivity or electromagnetic interference shielding that is enabled by the conductive fillers). An advantage of the absence of continuous fibers is the feasibility of forming the composites by injection molding or related conventional methods of polymer processing.

The non-monolithic materials that exhibit interface-derived viscoelasticity are not structural materials, but they can be used as constituents of composite materials or as nonstructural viscoelastic materials for constrained-layer damping. They include carbon fibers, carbon nanotubes (CNTs), boron nitride nanotubes (BNNTs), exfoliated graphite, carbon black, smectite clay (including organoclay, also known as nanoclay) and silica particles.

A large interfacial area is needed for substantial interface-derived viscoelasticity. In case of a composite material, an important interface is that between a filler and the matrix. Thus, a nanofiller is preferred, provided that it is adequately dispersed. The nanofillers include fibrous fillers (e.g., CNTs) [25], particulate fillers (e.g., clay, silica and carbon black) [24–28] and plate-like fillers (e.g., clay, graphene and graphite) fillers [25,29]. The interaction between the filler and matrix also matters [30], as it affects the ease and extent of interfacial sliding. Another type of interface is the filler-filler interface [28–30], which is present if the filler units are not perfectly dispersed. Yet another type of interface is phase boundaries, which occur in polymer blends [30].

In spite of the presence of discontinuous fillers and filler-matrix interactions, a composite material with a highly viscoelastic matrix

(such as an elastomeric polymer or rubber matrix) tends to have its viscoelastic behavior dominated by the bulk viscoelastic deformation mechanism of the matrix. Therefore, elastomer-matrix composites such as rubber-matrix composites, though mentioned above [25,28–30], are outside the scope of this review. However, it is possible for polymer-matrix composites with matrices that are not elastomeric to exhibit viscoelasticity that is dominated by interface-derived viscoelasticity.

In relation to composites with discontinuous fillers, this review addresses composites with matrices that are not elastomeric, as explained above. In general, such matrices include cement, carbon, ceramics and polymers that are not elastomeric. However, this review only addresses cement and polymer matrices, due to their having received substantial attention in relation to damping.

In relation to polymer-matrix composites with continuous fibers, this review addresses composites in directions that have the viscoelastic behavior dominated by the fibers rather than the matrix. This is because the domination of the matrix in the viscoelastic behavior results in a substantial contribution of the bulk viscoelastic deformation mechanism to the overall viscoelastic behavior of the composite.

In case of a non-monolithic material in the form of fibrous or particulate materials in the absence of a matrix (but optionally with the presence of a minor amount of a binder), the interface can be that between the structural units (e.g., fibers) of this material. For example, for a carbon fiber tow (bundle), the interface that is responsible for the viscoelasticity is that between the fibers in the tow [31]. Because the fibers are wavy to a limited degree, there are contact points between the fibers, thus providing interfaces.

An attraction of composite materials is that hybrid fillers may be used [32] in order to provide properties that no single filler can provide. For example, one filler is chosen to enhance the storage modulus and another filler is chosen to enhance the viscous character [33].

The objectives of this paper are (i) to review the subject of interface-derived solid-state viscoelasticity in the small-strain regime, (ii) to assimilate the numerous seemingly disjoint reports of this phenomenon for a wide variety of materials, and (iii) to enunciate coherently the scientific principles on this subject.

2. Cement-based materials

Cement-based materials such as concrete are important for construction. From the scientific viewpoint, the basic form of cement-based material is cement paste, which contains no aggregate. In contrast, mortar contains fine aggregate, and concrete contains fine and coarse aggregates. The aggregates serve as reinforcement and also serve to reduce the drying shrinkage during curing (hydration).

Unmodified cement paste, as formed by the curing of a mixture of cement particles and water, is a brittle material with very weak viscoelasticity. Admixtures are commonly added to a cement mix in order to enhance certain properties of the slurry or those of the resulting cement-based material. Admixtures can be in particulate, short fiber or liquid forms.

Silica fume is one of the most common particulate forms of admixture. It is a waste material, with particle size much smaller than that of sand (fine aggregate). Due to its stiff and small particle size (mean particle size between 0.1 and 0.2 μm), silica fume incorporation increases the tensile strength, compressive strength, compressive modulus, flexural modulus and tensile ductility, abrasion resistance, chemical attack resistance, corrosion resistance of the embedded steel, and bond strength to steel rebars [34]. Moreover, it decreases the liquid permeability, drying shrinkage, alkali-silica reactivity, creep rate and coefficient of thermal

expansion [34]. However, it decreases the compressive ductility and workability [34].

The silica fume serves not only as a reinforcement, but also provides interfaces, thereby enabling interface-derived viscoelasticity [24,26,34–37]. Due to the small size of the silica fume particles, the amount of interface is adequate to provide interface-derived viscoelasticity. Because of the pozzolanic activity of the silica fume, the bond between the silica particles and the cement in the cement-based material is substantial. Nevertheless, slight interfacial sliding occurs under dynamic loading, thereby providing mechanical energy dissipation. Hence, the silica fume incorporation enhances the storage modulus, loss tangent and loss modulus. In other words, both the stiffness and damping ability are enhanced. The stiffness enhancement is due to the silica fume acting as a reinforcement. The damping enhancement is due to the silica-cement interface provided by the silica fume. The simultaneous enhancement of stiffness and damping ability is highly attractive for structural damping.

With the incorporation of acid-treated silica fume in the cement paste and with the dynamic flexural testing conducted at 0.2 Hz and 30 °C, the loss tangent is increased from 0.035 to 0.172 (i.e., a 390% increase), the storage modulus is increased from 1.91 to 11.29 GPa (i.e., a 490% increase), and the loss modulus is increased from 0.067 to 1.94 GPa (i.e., a 2800% increase), with all the increases being relative to the plain cement paste (without silica fume) [26]. Both stiffness and viscous character are enhanced by the silica fume incorporation.

On the other hand, the silane treatment of silica fume has little effect on the loss tangent, but increases the storage modulus by up to 38% [36] – less than that for acid-treated silica fume [26], all relative to plain cement paste. The surface treatment affects not only the interfacial bonding, but also the dispersion of the silica fume in the cement. Thus, interface design is important for obtaining substantial interface-derived viscoelasticity.

The use of latex (styrene-butadiene particles dispersed in water) in place silica fume also results in increased loss tangent and storage modulus [26]. The increased loss tangent is due to the bulk viscoelastic deformation mechanism provided by the latex, although the latex-cement interface may contribute to the viscoelasticity. The higher is the latex proportion, the higher is both the loss tangent and storage modulus [26]. The increased storage modulus is due to the reduced porosity provided by the latex addition, which also increases the flexural strength [38]. However, latex even in the highest amount of 30% by mass of cement is not as effective for increasing the loss tangent and storage modulus as acid-treated silica fume in the amount of 15% by mass of cement [26]. At 0.2 Hz and 30 °C, the loss tangent is 0.142 and 0.172 for the latex (30%) and silica fume (15%) cases, respectively; the storage modulus is 3.12 and 11.29 GPa, respectively; the loss modulus is 0.443 and 1.94 GPa, respectively [26]. Furthermore, latex is expensive compared to silica fume.

The addition of sand instead of silica fume to cement paste to form mortar decreases the loss modulus at 30 °C and 0.2 Hz from 0.22 to <0.001 GPa and decreases the storage modulus from 13.7 to 9.43 GPa [35]. This means that both stiffness and damping ability are reduced by the use of sand in place of silica fume. The decrease in loss modulus is attributed to the poor damping capacity of sand compared to cement paste (less homogeneous than a sand particle) and the small contribution of the interface between sand (much larger than the silica fume particles) and cement paste to the loss tangent. However, further addition of silica fume to the mortar (so that both sand and silica fume are present) increases the loss modulus back to 0.28 GPa and increases the storage modulus back to 13.1 GPa [35].

The use of nanosilica particles (size 30 nm) instead of silica fume

in mortar (with sand) is also effective for enhancing the damping ability [39]. The loss tangent is increased from 0.019 to 0.067, but the storage modulus is decreased due to the reduction in workability (associated with the excessive need for water in the mix) resulting from the small size of the nanosilica particles. Nevertheless, the loss modulus is increased. Thus, nanosilica is not as effective as silica fume (microsilica) for enhancing the damping ability of cement-based materials. This means that nanofillers are not necessarily more effective for damping enhancement than microfillers.

Surface treatment of silica fume using silane helps the dispersion of the silica fume in the cement matrix, due to the enhanced hydrophilicity [36,40,41]. As a consequence, the silane treatment enhances the damping ability, in addition to increasing the mechanical properties [34,36,40–44]. The silane treatment of short carbon fibers also enhances the properties of carbon fiber cement-based materials [36,45,46], in which silica fume is used to help the fiber dispersion [47].

Silica fume is also effective for promoting the dispersion of multi-walled carbon nanotubes (CNTs) in cement [48]. In addition, nanosilica particles (14 nm average diameter) as an admixture improve the interaction between CNTs and the cement hydrates, thus resulting in an increase in friction between CNTs and the cement matrix [49].

The silica fume incorporation also enhances the sound absorption ability of cement paste [27]. The sound absorption coefficient of a material is a quantity that describes how much of the incident sound intensity is not reflected by the material. At a sound frequency of 125 Hz, the silica fume incorporation increases the sound absorption coefficient by 40%. Study of cement paste, mortar and concrete shows that the contribution to the sound absorption coefficient decreases in the order: silica fume, cement, sand and stones. Silica fume, sand and stones contribute because of the reflection of the sound wave at the interface between these particles and cement, and the consequent multiple reflections at the cement-particle interfaces [27]. The fact that silica fume is more effective than sand and stones is due to its small particle size.

Both silica fume (15% by mass of cement) and methylcellulose (0.4% by mass of cement) promote the dispersion of short carbon fibers in cement-based materials. The incorporation of short carbon fibers in cement paste containing silica fume and methylcellulose causes the loss tangent under flexure (≤ 1 Hz) to decrease by up to 25% and the storage modulus (≤ 2 Hz) to increase by up to 67%, such that both effects increase in the following order: as-received fibers, ozone-treated fibers, dichromate-treated fibers, and silane-treated fibers [36,50]. Carbon fibers (7 μ m diameter) are more effective than steel fibers (0.55 mm diameter, with hooked ends for mechanical interlocking with the matrix) for enhancing the damping ability [51].

Compared to particulate and microfiber fillers, nanoscale filamentous fillers are attractive for damping improvement due to the large area of the interface between the filler and the cement matrix. These fillers are primarily carbon nanofibers (CNFs, originally known as carbon filaments) and carbon nanotubes (CNTs). The CNFs have larger diameter than CNTs, but they are less expensive. Low cost is critical to the viability of construction materials. Due to the small diameter and intertwined morphology of CNFs and CNTs, as shown in Fig. 1 for CNFs, the dispersion of these fillers in cement is more challenging than that of carbon fibers (diameter typically around 10 μ m). The incorporation of a nanoscale filamentous filler (namely CNFs) in cement was first reported by Fu and Chung in 1996 [52]. The incorporation of CNTs, in particular, in cement was first reported by Li et al. in 2005 [53].

The CNT incorporation enhances the loss tangent, which relates to the reported damping ratio [48,54–56]. The damping ratio is

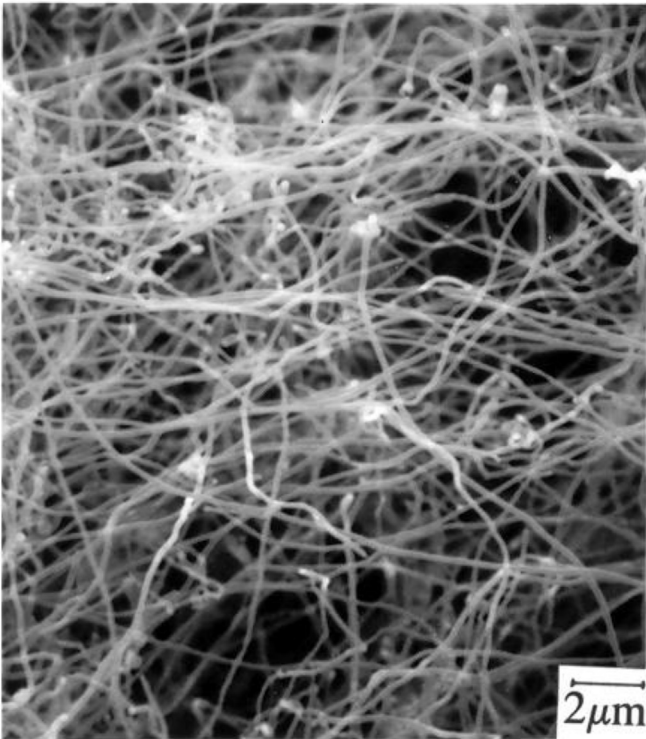


Fig. 1. A scanning electron microscope (SEM) image of carbon nanofibers (CNFs) prepared from methane and an iron-containing catalyst. The diameter is 0.15 μm . The channel diameter is 20–75 nm [204].

increased by only up to 21% through the incorporation of multi-walled CNTs of outer diameter 20–40 nm and length 5–15 μm [48]. This fractional increase is much lower than that provided by acid-treated silica fume [26]. The interfaces involved in the interface-derived viscoelasticity are the CNT-cement interface and the interface between the tubes in a CNT. The acid treatment of the CNTs helps the mechanical properties of the resulting cement-based material [53].

The CNFs commonly have diameter ranging from 0.1 to 0.2 μm , such that the carbon layers are oriented at an angle to the nanofiber axis (i.e., the fishbone morphology) (Fig. 2). In contrast, the carbon layers in CNTs are concentric cylinders. Since the chemical bonding between the carbon layers is weak (as in graphite) compared to the bonding within a carbon layer, the fishbone morphology causes the CNFs to be mechanically relatively weak along the nanofiber axis.

Both the fishbone morphology and the intertwined morphology

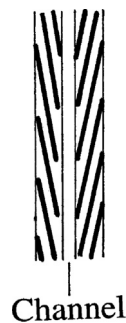


Fig. 2. A common type of carbon nanofiber that has a fishbone morphology. It has a hollow channel at its center. The carbon layers are stacked like a stack of ice-cream cones.

make the CNFs not as effective as the short carbon fibers at the same volume fraction as a reinforcement in cement [52,57–59]. Surface treatment of the CNFs with ozone helps the bond between CNFs and cement [60].

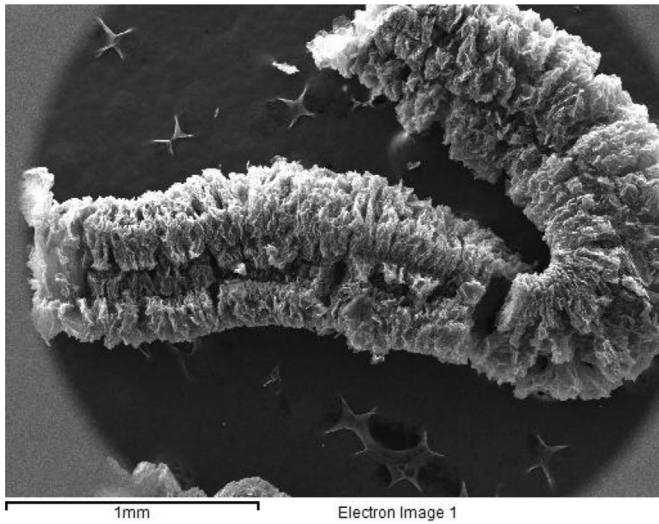
The dispersion of CNFs or CNTs in cement is challenging [59,60], but a number of methods have been devised for this purpose. The presence of silica fume helps the dispersion [61–65]. Sonication also helps [66–69]. In addition, the use of a surfactant, superplasticizer or high-range water reducing agent helps [69–74]. The surface treatment of CNFs by using nitric acid also helps [75]. A less common method of CNF dispersion involves the formation of a hybrid of CNFs and cement clinker particles [76,77]. An expensive method involves the catalytic growth of CNFs on the surface of silica fume or cement particles [78–80]. A problematic method involves using fine-grain cement particles [81]. This method is problematic because of the difficulty of reducing the size of the cement particles, the reactivity of fine-grain cement, and the consumption of a large amount of water at a high rate by fine-grain cement [81]. The dispersion methods are similar for CNFs and CNTs [82]. On the other hand, the CNFs (average diameter 0.1 μm , and the fiber length ranging from 50 to 200 μm) can reinforce the interfacial transition zone between the cement and aggregate [83]. Furthermore, the addition of CNFs increases the proportion of high-density calcium-silicate-hydrate (CSH) gel compared to low-density CSH gel [84].

A non-fibrous filler that has been exceptionally effective for enhancing the damping effectiveness of cement is exfoliated graphite (Fig. 3) [85,86]. The interface between the cell walls in the cellular structure of exfoliated graphite is responsible for the interface-derived viscoelasticity (Fig. 4) [87–90]. The tight inclusion of the exfoliated graphite particles in the cement matrix is important for the exfoliated graphite to be effective in the cement-matrix composite [91,92]. The simple addition of exfoliated graphite to a wet cement mix is not adequate for attaining the tight inclusion, which can be obtained by mixing exfoliated graphite with dry cement particles, followed by compressing the mixture and subsequent exposure to water for curing the cement in the mixture. This is constrained-layer damping in a microscale, using the exfoliated graphite as the distributed constrained layer. The inclusion of 8 vol.% graphite in cement paste results in loss modulus 7.5 GPa and loss tangent 0.81 [91]. These values are much higher than those (1.94 GPa and 0.17, respectively) provided by the incorporation of acid-treated silica fume in cement [26]. However, the combined use of exfoliated graphite and silica fume is effective for enhancing the stiffness while providing high damping [86].

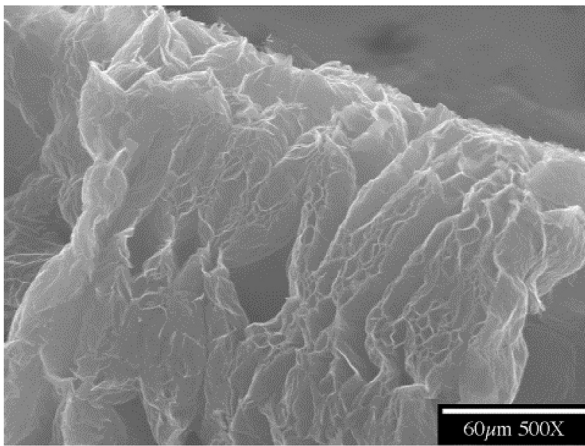
The use of ground waste rubber tire as a partial replacement for the fine aggregate in concrete for enhancing the damping ability [93] involves the bulk viscoelastic deformation mechanism rendered by the rubber. Nevertheless, the treatment of the rubber particles with silica fume helps the damping by improving the friction at the interface between the rubber particles and cement [93].

3. Continuous fiber polymer-matrix composites

Unless stated otherwise, the continuous fibers are carbon fibers. Continuous carbon fiber polymer-matrix composites are the dominant structural materials for aircraft, due to their combination of low density, high strength and high modulus. However, they are poor in the vibration damping ability, particularly in directions that have their mechanical properties dominated by the fibers rather than the matrix. Since a practical structural laminate is multidirectional rather than unidirectional, the situation of dominance by the matrix does not occur, except for the through-thickness direction of a laminate. The damping effectiveness depends on the fiber



(a)



(b)

Fig. 3. SEM images of exfoliated graphite (made by heating sulfuric-acid-intercalated graphite flake) that has been washed with water after exfoliation. A worm is the exfoliated material derived from a single graphite flake. (a) Exfoliated graphite (essentially an entire worm) at a low magnification [205]. (b) Exfoliated graphite (a part of a worm) at a high magnification [206].

orientation, as shown by varying the filament winding angle for carbon fiber composites fabricated by filament winding [94].

The incorporation of suitable nanofillers in the matrix of the composites enhances the damping ability, due to the interface-derived viscoelasticity, with the interface being that between the nanofiller and the polymer matrix. In the case that the nanofiller has interfaces inside each unit of the nanofiller, these interfaces also contribute to the viscoelasticity. An example of such a filler is multi-walled CNT, in which there is an interface between the adjacent concentric walls of the CNT.

Nanofillers rather than fillers of larger dimensions are suitable for several reasons. Firstly, the high volume fraction of continuous fibers in the composite needs to be maintained as far as possible. Since the continuous fibers are the primary reinforcement, their high volume fraction is critical for the strength and modulus of the composite. Secondly, the distortion of the continuous fibers is to be avoided. Because of the strong anisotropy in the mechanical

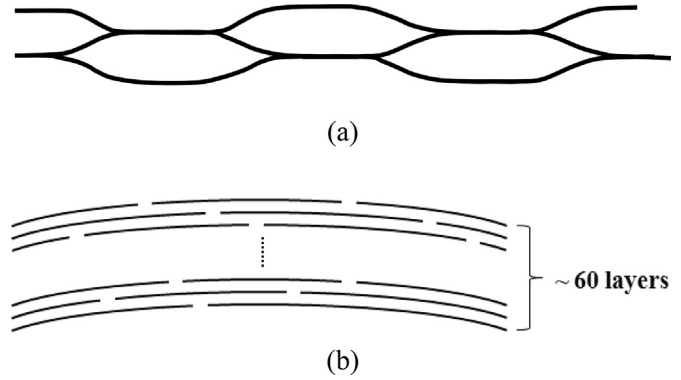


Fig. 4. (a) The cellular structure of exfoliated graphite, with each line representing a cell wall and each cell resulting from an intercalate island. (b) The structure of a cell wall, with each line representing a carbon layer (rather than a cell wall). In the case of exfoliated graphite made from sulfuric-acid-intercalated graphite flakes, the cell wall typically consists of about 60 carbon layers [87].

properties of a fiber, the bending of the fibers would degrade the strength and modulus of the composite. The incorporation of a nanofiller in a microfiber composite results in a multi-scale or hierarchical composite.

The method of nanofiller incorporation in a continuous fiber polymer-matrix composite can involve either (i) adding the nanofiller to the matrix precursor prior to introducing the matrix precursor to the continuous fibers, or (ii) applying the nanofiller to the interface between adjacent continuous fiber layers prior to consolidating the layer stack and fabricating the laminate composite. The nanofiller application in method (ii) can involve coating the continuous fiber layer with the nanofiller, or positioning a nanofiller mat (also known as paper) between the continuous fiber layers in the stack. Method (i) tends to be limited to low filler contents, since the viscosity increases with increasing filler content.

The first report of nanofiller incorporation for vibration damping enhancement of continuous carbon fiber polymer-matrix composites, as provided by Hudnut and Chung (1995), involves a nanofiller in the form of CNFs of diameter 0.1–0.2 μm , in the amount of 0.6 vol.% [12]. The continuous carbon fiber of diameter 7 μm is in the amount of 56.5 vol.%. The matrix is epoxy. The nanofiber is positioned at the interlaminar interface. Its incorporation greatly increases the loss tangent in both longitudinal and transverse directions of the unidirectional composite (Fig. 5), but decreases slightly the storage modulus in the longitudinal direction. Moreover, the nanofiber incorporation increases the loss modulus in both longitudinal and transverse directions [12]. The effectiveness of CNF incorporation in a continuous carbon fiber composite for damping enhancement was later confirmed by Segiet and Chung in 2000 [18], by Gou et al. in 2006 [95], by Liang et al. in 2010 [96] and by Jang et al. in 2012 [97].

The incorporation of CNTs in a continuous fiber polymer-matrix composite for damping enhancement was first reported by Johnson et al. in 2011 for a continuous glass fiber composite [98] and by Khan et al. in 2011 for a continuous carbon fiber composite [99]. In particular, in relation to continuous carbon fiber composites, Khan et al. showed that the damping ratio increases with the CNT content [99] and Kim et al. reported that the damping enhancement is accompanied by a decrease in the tensile strength [100]. On the other hand, concerning continuous carbon fiber polymer-matrix composites, Tehrani et al. reported that the damping enhancement is accompanied by an increase in the tensile ductility, while the tensile modulus and strength are essentially unaffected [101]. After the first report in 2011 [98], the work on glass fiber composite

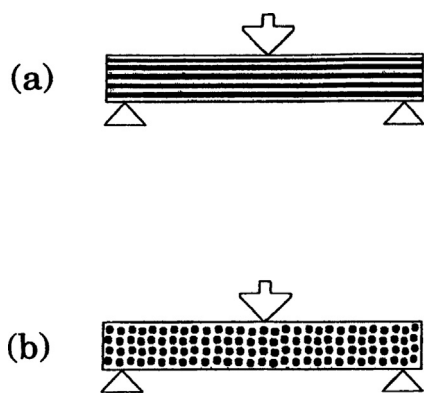


Fig. 5. Geometry for dynamic mechanical testing of a unidirectional continuous fiber polymer-matrix composite under three-point bending. (a) Longitudinal configuration, in which the fibers (in the plane of the figure) are bent during flexure. (b) Transverse configuration, in which the fibers (perpendicular to the plane of the figure) are not bent during flexure [18].

was confirmed by Alnefaie et al. in 2013 [102] and by Khashaba in 2015 [103]. After the first report in 2011 [99], the work on carbon fiber composite was confirmed by Kim et al. in 2013 [100], by DeValve and Pitchumani in 2013 [104], and by Tehrani et al. in 2013 [101].

The hybrid use of multiple fillers can be valuable for obtaining properties that neither single filler can provide. In order for the composite to exhibit both high loss tangent and high stiffness, as needed to obtain a high value of the loss modulus, a filler that functions to increase the loss tangent and another filler that functions to increase the stiffness can be used together. An example is the incorporation of either exfoliated graphite or nanoclay (for increasing the loss tangent) and either CNTs or halloysite nanotubes (HNTs, Fig. 6, low-cost natural clay mineral nanotubes, with aspect ratio lower than those of CNTs, for increasing the storage modulus). In case of the combined use of exfoliated graphite and HNTs as fillers, the consequence is that the loss modulus is increased by 110%, while the flexural strength is decreased by 14% and the flexural modulus is not affected [105]. Another example is the incorporation of multi-walled CNTs in the rubber-particle-toughened (with the rubber being butadiene acrylonitrile) epoxy matrix of a continuous carbon fiber composite [106]. Yet another

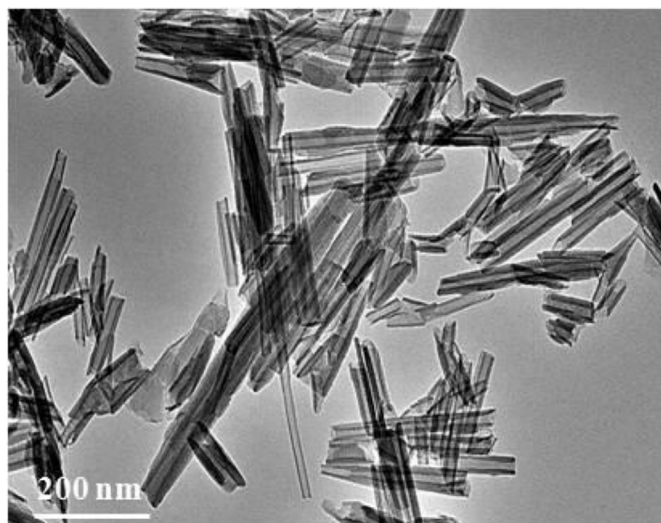


Fig. 6. Transmission electron microscope image of halloysite nanotubes (HNTs).

example is the incorporation of carboxyl-functionalized multi-walled CNTs in flax-fiber-filled continuous carbon fiber epoxy-matrix composite [107]. Still another example is the incorporation of CNF paper that includes short glass fiber in a continuous glass fiber polyester-matrix composite; the entanglement of the CNFs and short glass fibers promotes the damping [108].

A different approach involves the growth of ZnO nanorods on the surface of continuous carbon fibers. The resulting epoxy-matrix composite exhibits good damping ability, due to the interfacial-derived viscoelasticity, with the interfaces being the ZnO/epoxy interface and the ZnO/ZnO interface. The piezoelectric effect of ZnO also contributes to the damping, due to the conversion of mechanical energy to electrical energy through this effect. The electrical conductivity of the continuous carbon fibers facilitates the electrical energy output [109,110].

Boron nitride nanotube (BNNT) is a polymorph of boron nitride. Its structure is similar to that of CNT, though the carbon atoms in CNT are alternately substituted by nitrogen and boron atoms in BNNT. Fig. 7 shows the intertwined morphology of BNNTs. The morphology is similar to that of CNFs (Fig. 1). BNNT is an electrical insulator, whereas CNT is either metallic or semiconducting.

The CNT is more effective than BNNT for enhancing the damping of the polymer. This is attributed to CNT's lower interfacial shear strength with the polymer matrix and its effective sword-in-sheath mechanism compared to BNNT, which has bamboo-like nodes. On the other hand, BNNTs disperse better than CNTs, thus enabling the BNNT concentration to be relatively high. A high nanotube concentration helps the damping of the composite [111].

Graphene oxide is a particulate form of nanocarbon. The incorporation of graphene oxide in a continuous glass fiber epoxy-matrix composite also enhances the damping [112].

A conventional method of enhancing the damping ability of continuous fiber polymer-matrix composites involves the incorporation of viscoelastic polymer interlayers (commonly perforated to enhance the interlayer-matrix bonding by mechanical interlocking), which functions using the bulk viscoelastic deformation mechanism of viscoelasticity [113–120]. The sandwiching of the interlayer by the stiff continuous fiber layers results in constrained-layer damping.

Segiet and Chung in 2000 [18] provided a comparison of the CNF incorporation and viscoelastic polymer interlayer incorporation on the vibration damping performance of continuous carbon fiber polymer-matrix composite, with the polymer being a

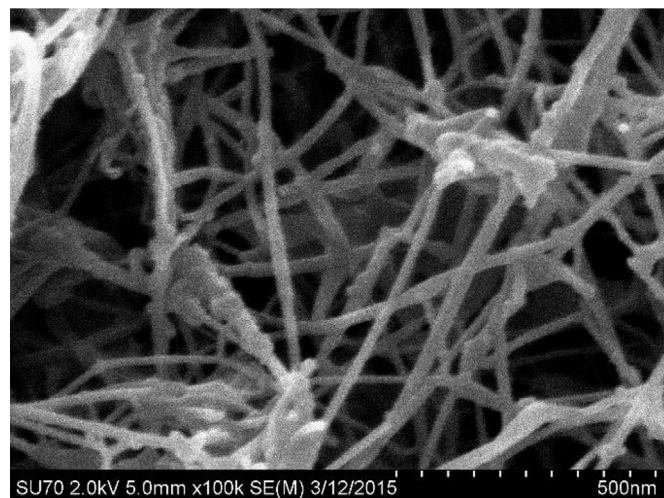


Fig. 7. SEM image of boron nitride nanotubes [197].

thermoplastic polymer, namely Nylon-6, which is more ductile than the epoxy matrix used by Hudnut and Chung [12]. Due to the higher damping ability of the thermoplastic polymer matrix compared to the epoxy matrix, the enhancement of the damping ability by filler incorporation is more challenging for the thermoplastic matrix than the epoxy matrix.

Segiet and Chung [18] used discontinuous ozone-treated carbon nanofibers as a filler in the amount of 0.64 vol.% of the continuous unidirectional carbon fiber Nylon-matrix composite and positioned at the interlaminar interface. The CNFs are effective for increasing the loss tangent under flexure in a direction that involves the bending of the fibers, such that the flexural storage modulus or tensile modulus or strength in the fiber direction is decreased only slightly.

As reported by Segiet and Chung [18], the loss tangent for the longitudinal configuration is increased by the presence of the CNF interlayer or viscoelastic polymer interlayer (Fig. 8(B)), while the storage modulus for the longitudinal configuration is decreased (Fig. 8(C)). For the transverse configuration, the loss tangent is increased by the presence of the viscoelastic polymer interlayer, but not much affected by the CNF interlayer (whether the CNFs are ozone-treated or not); the storage modulus is decreased by the viscoelastic interlayer and the as-received CNF interlayer, but is increased slightly by the ozone-treated CNF interlayer. The loss tangent for both longitudinal and transverse configurations is increased most significantly by the presence of the viscoelastic polymer interlayer, while the storage modulus for both configurations is decreased most significantly by the viscoelastic polymer interlayer. For both configurations, the loss modulus is highest for the case of the viscoelastic interlayer (Fig. 8(A)). All effects are much larger for the longitudinal configuration than the transverse configuration.

The viscoelastic polymer interlayer is even more effective than the CNF interlayer for enhancing the loss tangent, but the accompanying decrease in storage modulus is much more. Moreover, viscoelastic behavior of the viscoelastic polymer is temperature sensitive. The loss tangent and storage modulus of the composite with the viscoelastic polymer interlayer decrease upon heating, so

the loss modulus for this composite is less than that of the composite with the CNF interlayer (with ozone-treated CNFs) at temperatures exceeding 50 °C (Fig. 8(A), (B) and (C)) [18]. The ozone treatment makes the CNFs much more effective.

Fig. 8(D) shows that the tensile strength and modulus are decreased and the ductility is increased by the presence of the CNF interlayer or viscoelastic polymer interlayer, such that the changes are greatest for the composite with the viscoelastic polymer interlayer and least for the composite with the ozone-treated CNFs. Although the continuous fiber volume fraction is lower for the composite with the CNF interlayer than for that with the viscoelastic polymer interlayer, the strength and modulus are higher for the former. The ozone treated CNFs give higher strength, higher modulus and lower ductility than the as-received CNFs [18].

The use of a polymer nanofiber mat at the interlaminar interface is effective for damping enhancement, in addition to improving the interlaminar fracture toughness and impact damage resistance. The penetration of the matrix through the mat contributes to providing the attractive effects [121].

Similarly, a Nylon-6 fiber mat at the interlaminar interface of a carbon fiber epoxy-matrix composite enhances the damping [122]. A related but different approach involving commingling carbon fiber and Nylon-6 fibers [122].

In a one-sided form of constrained-layer damping (with the constrained layer being constrained on only one of its two opposite surfaces), the addition of a CNF paper (the constrained layer) with inclusion of short glass fibers in the paper on the surface of a composite laminate enhances the damping [123]. Although the one-sided form of constrained-layer damping is not as effective as the more ideal two-sided form of constrained-layer damping, it is advantageous in the simplicity of incorporation of the constrained layer.

4. Polymer-matrix composites with discontinuous fillers

Polymer-matrix composites with discontinuous fillers (e.g., short fibers and particles) are not adequate in their mechanical properties for serving as advanced structural materials. However,

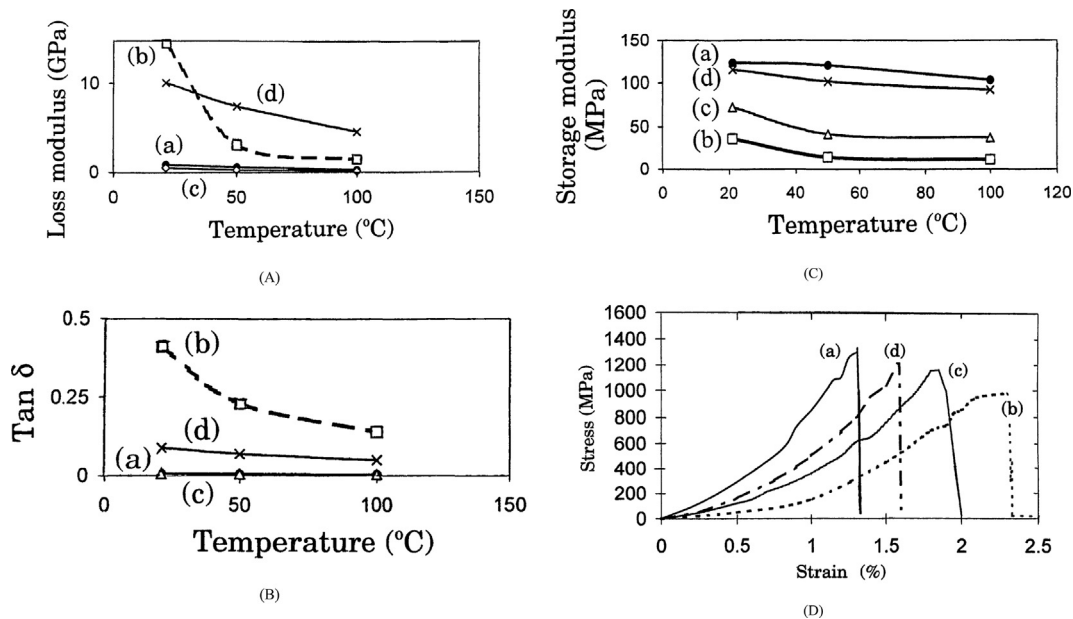


Fig. 8. Effect of temperature on the loss modulus (A), $\tan \delta$ (B), and storage modulus (C) of continuous carbon fiber Nylon-matrix composite for the longitudinal configuration at 0.2 Hz. (D) The corresponding tensile stress–strain curves. (a) Composite without interlayer. (b) Composite with viscoelastic polymer interlayer. (c) Composite with as-received carbon nanofiber interlayer. (d) Composite with ozone-treated carbon nanofiber interlayer [18].

they may serve as structural materials in less demanding structures and as multifunctional materials. Nanoscale fillers are particularly effective for damping enhancement, due to the large filler-matrix interface area.

Single-walled CNTs in polycarbonate increases the loss modulus by 250% due to interface-derived viscoelasticity. However, fullerene (C_{60}) with a similar surface area to volume ratio as the CNTs and at the same loading (1 wt.%) does not increase the loss modulus [124,125]. This is because the CNTs are more effective than fullerene for stiffening and the stiffness partly governs the loss modulus.

A polymer is less ductile below the glass transition temperature. CNTs in epoxy provide interfaces that improve damping below the glass transition temperature at low strains [126–129].

Polyisobutylene-based polyurethane filled with multi-walled CNTs are effective for damping. The CNTs serve as H-bonds acceptor chain extenders [130].

The addition of CNFs to polyester increases the damping [131]. However, the addition of CNFs to polyvinylidene fluoride (PVDF, which is piezoelectric) decreases the damping, while increasing the storage modulus [132]. This is because of the piezoelectric damping provided by the PVDF.

The dense packing of well-aligned multi-walled CNTs in an epoxy film that is sandwiched by steel plates gives up to 1400% increase in loss factor (damping ratio) relative to the unfilled epoxy. The damping enhancement is not accompanied by the decrease of the strength or stiffness [133].

The interfacial stick-slip mechanism, which is associated with interfacial friction, is responsible for the damping enhancement resulting from the nanofiller incorporation. In this mechanism, a nanofiller first sticks and then slips suddenly when the force applied to it is large enough. However, the interfacial slippage tends to be detrimental to the strength and stiffness. Functionalization of the CNTs can enhance the interaction between CNT and matrix, thereby reducing the interfacial slippage, thereby improving the strength and stiffness at the expense of the damping effectiveness [134–144]. By tailoring the nanostructure and interfacial interaction, the stiffness can be maintained while the damping is enhanced.

A low CNT-matrix interfacial shear strength promotes damping. CNT waviness degrades the damping [145]. The free volume in a polymer promotes molecular motion and hence damping; it also promotes damping in a CNT-filled polymer [146,147]. Modeling shows that the damping is insensitive to the CNT segment length, while the CNT diameter is influential to the damping [148]. Suitable functionalization of single-walled or multi-walled CNTs can be used to tailor the filler-matrix interface, thereby extending the temperature range for damping effectiveness [149,150]. The functionalization of multi-walled CNTs has been shown to enhance the damping of epoxy beyond that provided by the pristine CNTs [151–154]. The functionalization of multi-walled CNTs by carboxylation is quite common [155]. Silane treatment is a type of surface treatment that helps the dispersion of multi-walled CNTs in epoxy [154]. Mechanical dispersion using a high shear force is effective and may be feasible for industrial scale-up [155]. In situ emulsion polymerization and blending are two of the methods that are used for combining CNTs with the polymer to form a composite [153].

Among multi-walled CNTs, those with a high aspect ratio and specific surface area are preferred for improving damping [156]. CNTs that are aligned are more effective than those that are randomly oriented [157]. CNTs that are coiled in shape improve the vibration damping, stiffness, fracture toughness of polymers [158]. The coiled shape induces mechanical interlocking between the CNTs and the matrix. For enhancing the damping of epoxy, it has been reported that single-walled CNTs are more effective than multi-walled CNTs [150], and it has also been reported that multi-

walled CNTs are more effective than single-walled CNTs [159].

Although more work has been performed using CNTs than CNFs, CNFs are also effective for damping enhancement of polymers, as shown for epoxy and polypropylene matrices [160–165]. The orientation and waviness of the CNFs in epoxy affect the damping [166,167]. CNF-filled polymers are also effective for acoustic damping [166]. Single-walled CNTs in the form of bundles (known as ropes) are also effective for damping [167].

Polymers in the form of interpenetrating polymer networks are attractive for damping, due to the interface between the network components. The incorporation of multi-walled CNTs in a polyurethane/epoxy interpenetrating polymer network composite further enhances the damping [168,169]. An additional advantage of an interpenetrating polymer network is the difference in affinity for a nanofiller between the two network components and the consequent positioning of the nanofiller in one of the two network components.

Polymer foams are widely used for acoustic damping, due to the viscoelasticity of the polymer and the ability of the sound wave to hit the substantial accessible surface of the open-cell foam. The incorporation of multi-walled CNTs in an open-cell polyurethane foam enhances the acoustic damping [170], due to the interface between CNTs and polyurethane causing additional reflection of the sound wave.

Through the interface-derived viscoelasticity, the addition of CNTs to a constrained polymer enhances the constrained-layer damping. A sandwich composite in the form of two aluminum plates sandwiching epoxy for constrained-layer damping has the damping enhanced by the incorporation of multi-walled CNTs in the sandwiched epoxy [171]. Instead of constraining by sandwiching, one-sided constraining can be obtained by bonding the polymer to a substrate. Thus, the covering of an aluminum beam with a composite film of multi-walled CNTs in polyaniline provides effective constrained-layer damping [172]. The incorporation of multi-walled CNTs in polymer also enhances the damping when the polymer is sandwiched by metal and ceramic for constrained-layer damping [173].

As shown by modeling, the functionalization of single-walled CNTs in epoxy increases the interfacial shear strength. The increased shear strength may enhance or reduce the damping ability of the composite, depending on the stress range and CNT aspect ratio [174]. This points to the importance of designing the filler-matrix interface with consideration of the stress range and filler structure.

Another route to enhancing the damping uses a CNT network assembly. This network can be formed by the re-agglomeration of single-walled CNTs decorated with poly(methylmethacrylate). The network has been shown to be effective for enhancing the damping of epoxy [175,176].

A three-dimensional CNT assembly in the form of microtrusses is similarly effective for enhancing the damping of a polymer. With the crevices of the trusses filled with soft polyurethane, the composite exhibits high stiffness and high damping. The damping mechanism involves mainly the bulk viscoelastic deformation mechanism provided by the polyurethane, though the CNTs supplement the damping with interface-derived viscoelasticity [177].

The hybrid use of CNTs and ceramic particles as fillers in a polymer is attractive, as the CNTs enhance the damping, while the ceramic particles enhance the stiffness and decrease the coefficient of thermal expansion (CTE). Below are examples of the use of hybrid fillers. The hybrid use of multi-walled CNTs and alumina nanoparticles in epoxy is attractive, with the CNTs serving to enhance the loss modulus and the alumina nanoparticles serving to enhance the storage modulus [178]. The hybrid use of CNFs (200–500 nm diameter) and silica particles (~12 μm size) in epoxy

enhances the damping, in addition to decreasing the CTE [179,180]. The hybrid use of CNFs and hollow glass spheres (microballoons) in epoxy results in syntactic foams that are lightweight and effective for damping [181]. The hybrid use of CNFs and clay in epoxy results in enhanced damping [182], with CNFs and clay both providing a large filler-matrix interface area. Similarly, the hybrid use of multi-walled CNTs and clay in polymer-impregnated wood is effective for damping enhancement [183]. The hybrid use of CNTs, clay and silica nanoparticles in epoxy is potentially attractive for damping [184]. The hybrid use of CNFs and 0–3 piezoelectric ceramic PMN (lead magnesium niobate-lead zirconate-lead titanate) in epoxy enhances the damping, due to the piezoelectric damping provided by PMN and the conductivity provided by the CNFs [185]. The further incorporation of carbon black further enhances the damping, due to the increased conductivity that is needed for capturing the electrical energy provided by the piezoelectric energy conversion [185]. The hybrid use of lead zirconate titanate (PZT) and multi-walled CNTs is also effective [186]. Similarly, the hybrid use of multi-walled CNTs and a piezoelectric complex of lead magnesium niobate and lead zirconate titanate (PMN-PZT) is also effective [187]. The similar hybrid use of CNTs (mostly double-walled) and lead zirconate titanate piezoelectric particles in a semi-crystalline thermoplastic polymer is effective for damping, whether the composite is sandwiched by metal plates for constrained-layer damping or not [188]. The sandwiching of a multi-walled CNT film (grown on silicon) by a piezoelectric PZT (lead zirconate titanate) sheet and a silica sheet through adhesive bonding is effective for damping, which involves a combination of constrained-layer damping (using the CNT mat as the constrained layer) and piezoelectric damping [189].

Vertically aligned CNT arrays in a polymer matrix provide a large CNT/matrix interface area. In addition, the aligned configuration facilitates the coordinated bending of the CNTs. These arrays may be attractive for damping, as suggested by calculation [190,191].

Dynamic mechanical testing ideally involves a sinusoidal stress or strain input at a chosen frequency, as illustrated in Fig. 9. This is known as forced resonance. In such testing, a static pre-strain superimposed on the dynamic strain facilitates the activation of interfacial slip at the CNT-polymer interfaces at relatively low dynamic strain amplitudes. As a consequence, the interfacial sliding becomes easier, thereby enhancing the damping [192].

5. Materials in the absence of a matrix

Fibrous and particulate materials in the absence (or near absence) of a matrix can exhibit interface-derived viscoelasticity,

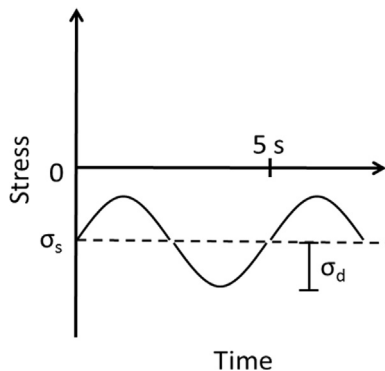


Fig. 9. Schematic illustrations of the variation of the stress with time during dynamic mechanical compression at 0.2 Hz. The static and dynamic stresses are σ_s and σ_d respectively [201].

due to the interfaces (e.g., nanotube-nanotube interfaces) in these materials. These materials may be used as the constrained layer in constrained-layer damping. Due to the absence of a polymer matrix, the viscoelastic behavior is relatively temperature independent and can occur at elevated temperatures, in contrast to the temperature dependence and limited elevated temperature resistance of polymers that exhibit the bulk viscoelastic deformation mechanism.

Viscoelastic behavior is exhibited by a tow of continuous carbon fibers in the absence of a matrix material [193]. The sliding at the interface between the fibers in the tow allows interface-derived viscoelasticity. The interface is in the form of contact points between nominally parallel adjacent fibers, as enabled by the fact that the fibers exhibit a degree of waviness. Similarly, viscoelastic behavior is exhibited by an assembly of CNTs [194–196] and an assembly of boron nitride nanotubes [197]. Due to the intertwined morphology of the nanotube assemblies, the nanotubes contact one another at points (Fig. 7). Such assemblies can be in the form of mats (also known as paper), which can be prepared using the paper-making process.

Viscoelastic behavior based on the interface mechanism is also exhibited by particulate compacts. A compact of the silica fume in the absence of a binder [198,199] exhibits this behavior.

Strong viscous behavior occurs in exfoliated graphite (Fig. 3) [87,88]. The degree of viscous character decreases with increasing degree of compaction of the exfoliated graphite (Fig. 10). However, even at a high degree of compaction, as in the case of commercially available compacted exfoliated graphite known as flexible graphite (or known as graphite foil), the viscoelastic character is still substantial [200]. The sliding at the interface between the cell walls in the cellular structure of exfoliated graphite (Fig. 4) accounts for the viscous character. This mechanism is supported by an analytical model [90].

Viscoelastic behavior also occurs in smectite clay [201], which is a layered silicate. The nanoscale layered structure provides interfaces. Viscoelastic behavior also occurs in carbon black [202,203], which is in the form of aggregates of nanoparticles (Fig. 11). The aggregate structure provides interfaces between nanoparticles in an aggregate, as well as aggregate-aggregate interfaces. The viscous character increases with the aggregate size (Fig. 12).

6. Conclusions

This paper reviews of the subject of interface-derived solid-state

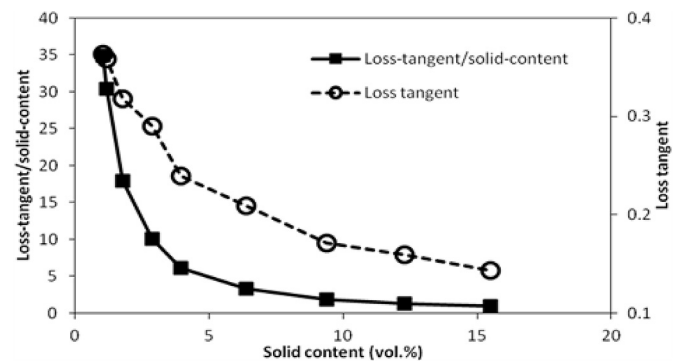


Fig. 10. Dynamic flexural properties of exfoliated graphite compacts at various solid contents (vol. % solid, as obtained by controlling the compaction pressure) under dynamic flexure, with the static strain at 2% and the frequency at 0.2 Hz. The loss tangent divided by the solid content relates to the degree of viscous character of the cell wall; its highest value of 35 occurs at the lowest solid content of 1 vol.% [87].

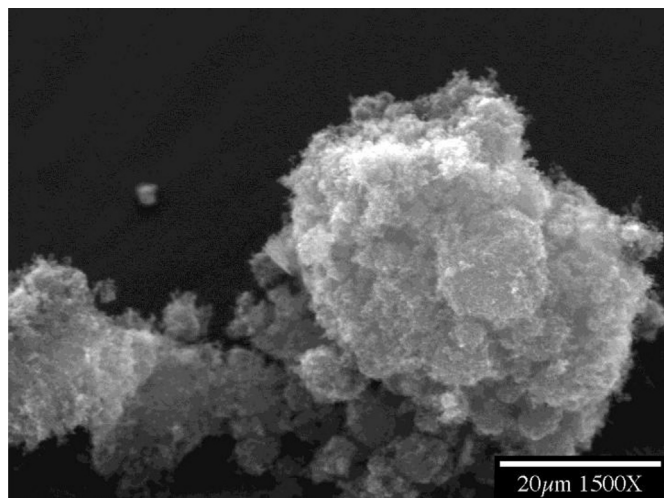


Fig. 11. SEM image showing the structure of carbon black, which is in the form of aggregates of nanoparticles. The carbon black is Product Vulcan XC72R from Cabot Corp., Billerica, MA, U.S.A., with DBP 188 ml/100 g, average pH 7.5, specific surface area 254 m²/g, primary particle size 30 nm, bulk density 0.09 g/cm³, and aggregate size 36–1122 nm.

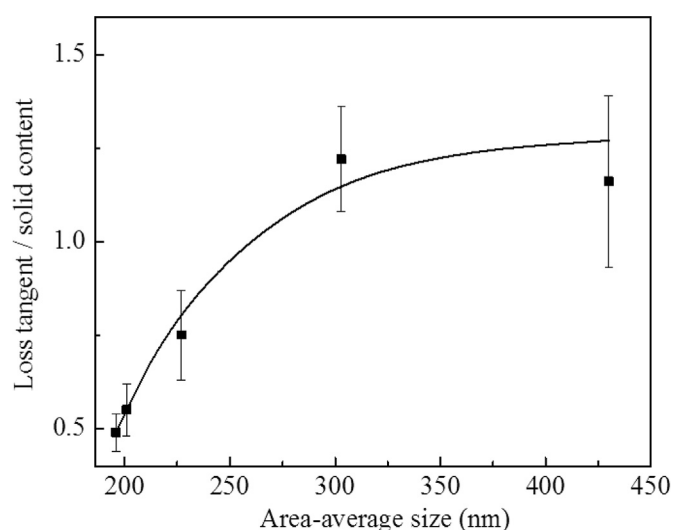


Fig. 12. Correlation of the average aggregate size with the loss-tangent/solid-content. The frequency is 0.2 Hz. The data are for five types of carbon black [203].

viscoelasticity, which is relevant to vibration and acoustic damping. It is the first review since 2003 [36]. The explosive growth of the field of nanotechnology since 2003 has fueled the growth of the field of this review. This review emphasizes vibration damping rather than acoustic damping, because prior work is concentrated in this area. The low-strain elastic regime of dynamic deformation is addressed, as it is relevant to normal structural operation and basic scientific understanding.

Compared to the conventional bulk viscoelastic deformation mechanism of viscoelasticity, the interface-derived viscoelasticity is attractive for the relative independence on temperature, the elevated temperature resistance, and the feasibility of enhancing the loss tangent and stiffness at the same time. This review includes comparison of these two mechanisms in terms of the resulting viscoelastic behavior. For example, the addition of silica fume to cement results in the enhancement of both loss tangent and stiffness. In contrast, the conventional bulk viscoelastic deformation

mechanism, as commonly provided by polymers, is temperature sensitive, with limited ability to withstand elevated temperature, and with the loss tangent enhancement accompanied by a stiffness decrease. For structural damping, both high loss tangent and high stiffness are needed.

The subject of interface-derived solid-state viscoelasticity emerged in 1995 [12] in relation to continuous fiber polymer-matrix composites, and in 1996 [24] in relation to cement-matrix composites. Fillers are incorporated in the composites in order to provide filler-matrix interfaces that can undergo slight frictional sliding, thereby dissipating mechanical energy and providing passive damping. Excessive ease of sliding is not desirable, since it would cause the strength and stiffness to decrease. Ideally, a filler is itself stiff, serving as a reinforcement, while the filler-matrix interface provides interface-derived viscoelasticity. This is the case for silica fume in cement. However, in case that the filler is not stiff, as in the case of exfoliated graphite, the inclusion of the filler in a stiff host (e.g., continuous fiber polymer-matrix composite and cement) provides stiffness and distributed constrained-layer damping in a microscale.

Nanostructured fillers such as CNTs, CNFs, exfoliated graphite and smectite clay are attractive for the large interface area that they provide. Microfillers such as silica fume can also be effective, as shown for the case of cement as the host. Microfillers can be dispersed more easily than nanofillers. In case of CNTs, the interface between the adjacent walls of the multi-walled nanotube adds to the interface for providing the interface-derived viscoelasticity. In case of exfoliated graphite, the interface between the cell walls in the cellular structure plays a major role in the viscoelasticity. In order to obtain the combination of high loss tangent and high stiffness, hybrid fillers can be used. For example, one filler in the hybrid is effective for enhancing the loss tangent (e.g., exfoliated graphite), while the other filler in the hybrid is effective for enhancing the stiffness (e.g., halloysite nanotubes).

The filler geometry, filler content, filler-matrix bonding (as controlled, for example, by the functionalization of the filler surface), and filler assembly configuration affect the energy dissipation (as described in terms of the loss modulus) and the stiffness. In case that the filler is incorporated in a continuous fiber polymer-matrix composite, the filler volume fraction needs to be minimized so that the continuous fiber volume fraction remains high, as needed to provide high strength and high modulus to the composite.

The filler may be incorporated by adding it to the matrix precursor (e.g., the resin) prior to introducing the matrix precursor to the assembly of continuous fibers. However, the filler addition increases the viscosity of the matrix precursor. This viscosity should remain low for effective introduction of the precursor to the continuous fiber assembly. Alternatively, the filler may be positioned at the interface between the adjacent continuous fiber laminae prior to forming the composite laminate.

Composites in the absence of continuous fibers can also be rendered viscoelastic with the incorporation of fillers. Without continuous fibers, the composites are not adequate for advanced structural applications such as airframe, but they are adequate for applications that are less demanding mechanically, such as computer and cell phone enclosures. In the absence of continuous fibers, the composites can be formed into complex shapes by injection molding. The feasibility of such molding is an important advantage of using discontinuous fillers.

The composites with or without continuous fibers, as mentioned above, are monolithic materials. Non-monolithic materials without a matrix can be rendered viscoelastic through the utilization of the interfaces in the material (e.g., the nanotube-nanotube interfaces in a nanotube assembly). Such materials can be used as the viscoelastic constrained layer in constrained-layer

damping. Examples of such materials include carbon fibers, exfoliated graphite, carbon black, CNT/BNNT mats and smectite clay.

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