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Viscoelastic behavior of the cell wall of exfoliated graphite

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ABSTRACT

The cell wall (~60-graphite-layer thick, average) in the cellular structure of exfoliated graphite compacts is viscoelastic, due to the shear between graphite layers. The viscous character decreases with increasing solid content (volume fraction), due to the increasing difficulty of shear, which becomes limited at solid contents above 4 vol.%. The elastic character is essentially independent of the solid content, weakening slightly with increasing solid content. The viscous character is stronger under in-plane flexure than compression in the compaction direction, due to the preferred orientation of the graphite layers. At the lowest solid content of 1.0 vol.%, the loss-tangent/solid-content is 35 and 25 under flexure and compression respectively, the storage-modulus/solid-content is 125 MPa and 46 kPa under flexure and compression respectively, and the loss-modulus/solid-content is 45 MPa and 13 kPa under flexure and compression respectively. The viscous character is strong under both flexure and compression, whereas the elastic character is much stronger under flexure than compression. The loss-tangent/solid-content decreases with increasing solid content, leveling off at 0.9 at 15 vol.% solid. The loss-modulus/solid-content also decreases with increasing solid content. The highest values of the loss-tangent/solid-content, storage-modulus/solid-content and loss-modulus/solid-content are greater for exfoliated graphite compacts than rubber or carbon black compacts.

1. Introduction

Mechanical vibrations can affect the performance, durability and safety of structures, whether they occur during normal structural operation or extreme events (such as earthquakes). Relevant structures include the civil infrastructure, railroad, aircraft, satellites, automobiles, wind turbines, skis, washing machines, microelectromechanical systems, etc. Viscoelastic solids are valuable for vibration damping and mechanical isolation. Damping refers to the reduction of the vibration amplitude by the conversion of the mechanical energy to another form of energy, typically thermal energy. This energy conversion is known as mechanical energy dissipation. Mechanical isolation refers to the avoiding of the propagation of mechanical energy from one object to another, typically achieved through the spreading of the mechanical energy along an isolation material positioned between the two objects.

Rubber and other elastomers are highly effective for mechanical isolation, due to their cushioning effect. However, they are not effective for damping, due to their softness and the consequent low ability for mechanical energy dissipation. Furthermore, rubber suffers from their tendency to degrade in the environment, particularly upon exposure to ultra-violet radiation. In addition, rubber is poor in the ability to withstand elevated temperatures and in the chemical resistance. The viscoelastic behavior of exfoliated graphite [1–4] compacts (also known as flexible graphite [5–10]) under dynamic flexure and compression has been reported [10,11]. Moreover, the viscoelastic behavior of cement containing exfoliated

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graphite [12,13] and of continuous carbon fiber epoxy-matrix composites containing exfoliated graphite has been reported [14]. In particular, the addition of exfoliated graphite to cement or a carbon fiber epoxy-matrix composite enhances the viscous character.

Exfoliated graphite has a cellular structure (an accordion-like structure) [11], due to the vaporization of the intercalate or the emission of gases resulting from the decomposition of the intercalate during heating and the consequent ballooning of each intercalate island during exfoliation [15]. The stretching of the balloon wall during the ballooning is enabled by the sliding of the graphite layers in the balloon wall relative to one another. Thus, exfoliated graphite consists of cell walls (balloon walls) and voids. The cell wall is a nanoscale multilayer that consists of graphite layers. The feasibility of exfoliation relates to the easy sliding of the graphite layers relative to one another. The ease of sliding and the large area of the interfaces between the layers in the multilayer suggest that the cell wall may exhibit a high degree of viscous character. This understanding motivates us to study the viscoelastic behavior of the cell wall in this work.

In spite of the prior work mentioned above, the viscoelastic behavior of exfoliated graphite has not been previously studied with focus on the cell wall of the exfoliated graphite. By focusing on the cell wall, this work has found that the cell wall exhibits an usually high degree of viscous character.

Upon compaction of a collection of pieces of exfoliated graphite, the pieces are consolidated (as enabled by the large amount of voids in each piece) and are mechanically interconnected (mechanically interlocked, as enabled by the cellular structure), thus resulting in a sheet (known as flexible graphite) with the plane of the sheet being perpendicular to the direction of compaction. The higher is the compacting pressure, the higher is the solid content in the compact, the higher are the storage and loss moduli, and the lower is the loss tangent [11]. It is possible for the compaction to affect the ease of sliding of the graphite layers in the cell wall, but this possible effect has not been previously investigated.

This paper is aimed at (i) studying the viscoelastic behavior of the cell wall in the cellular structure of exfoliated graphite, (ii) studying the effect of compaction on the viscoelastic behavior of the cell wall, such that the range of the degree of compaction is wider than that in prior work on the viscoelastic behavior of exfoliated graphite [11], (iii) achieving cell walls that exhibit an unusually high degree of viscous behavior by proper choice of the degree of compaction, and (iv) comparison of the cell wall of exfoliated graphite with carbon black and rubber in terms of the viscoelastic behavior.

The approach used in this work involves dynamic mechanical testing at a controlled frequency. In this method (forced resonance method), a sinusoidal stress wave with specified values of the static stress and the dynamic stress is imposed on the specimen by using a load cell and the resulting strain wave (along with the associated static strain and deformation amplitude) is measured by using a displacement transducer. The phase difference between the input stress wave and the output strain wave is measured, thereby giving the loss tangent (tan δ), which describes the viscous character. The ratio of the dynamic stress to the dynamic strain gives the storage modulus, which describes the elastic character and corresponds to the real part of the complex modulus. The imaginary part of the complex modulus is the loss modulus, i.e., the viscous modulus, which corresponds to the product of the storage modulus and the loss tangent.

The specimen is an exfoliated graphite compact (flexible graphite). For testing under flexure (three-point bending), the specimen is in the shape of a beam with the neutral plane of the beam in the plane perpendicular to the compaction direction. For testing under uniaxial compression in the compaction direction, the specimen is in a rectangular cuboid shape, with the compression axis during testing parallel to the compaction direction. Flexure allows shear between the graphite layers, whereas uniaxial compression does not.

Based on the measured density of the specimen, the solid volume fraction is determined. The solid volume fraction is systematically varied by varying the pressure used in compacting the exfoliated graphite. The viscoelastic properties of the solid part of the compact (i.e., the cell walls) are obtained from the measured viscoelastic properties of the compact by assuming linearity between a compact property and the solid volume fraction (i.e., the rule of mixtures). The validity of this assumption is well established for the storage modulus, but it has also been shown for the loss tangent [14].

2. Experimental methods

As in prior work [11], exfoliated graphite (worms) is obtained by rapid heating of expandable graphite flake from Asbury (No. 3772) at 900 °C for 2 min with flowing nitrogen. The worms of length 2–4 mm and specific surface area 41 m^2/g (measured in this work by nitrogen adsorption using a Micromeritics ASAP 2010 instrument and corresponding to about 60 graphite layers stacked in a cell wall on the average) are compressed in a steel mold at pressures ranging from 0.25 to 7.71 MPa (compared to 0.3–1.7 MPa in prior work [11]) for 5 min to form flexible graphite plates of density ranging from 0.0236 to 0.3503 g/cm^3 (compared to 0.0267–0.0891 g/cm^3 in prior work [11]) and carbon volume fraction (i.e., 1 – porosity) ranging from 1.04 to 15.51% (compared to 1.2–3.9% in prior work [11]). The density is determined by measurement of the mass and volume; at least four specimens are measured for each value of the fabrication pressure. The fabrication pressures used are low compared to prior work [16] and are chosen in order to accentuate the viscous behavior. As a result, the density of the resulting graphite sheets is quite low compared to those that are commercially available (1.1 g/cm^3) and that were used in the prior work [16].

The method of testing is the same as in prior work [11]. The specimen configurations for dynamic mechanical testing are illustrated in Fig. 1. For flexural testing, the thickness ranges from 3 to 4 mm and the specimens are in the form of beams of size 25×8 mm in the plane of the flexible graphite sheet (Fig. 1(a)). For compressive testing, the out-of-plane thickness of the fabricated sheet ranges from 3–4 mm for compressive testing (Fig. 1(b)). Specimens for compressive testing are subsequently obtained by cutting the sheets. The cutting is such that the specimen after cutting is 3 to 4 mm in the direction of compression, and the area perpendicular to the compression direction is 10×10 mm.

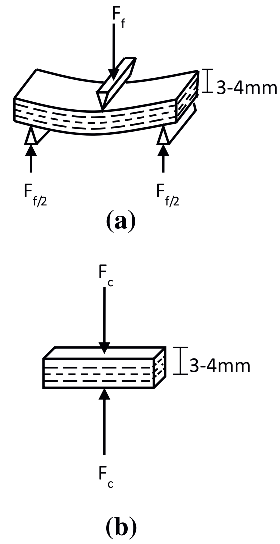

Fig. 1 – Specimen configurations for dynamic mechanical testing. (a) Flexure. (b) Uniaxial compression (out-of-plane). The dashed parallel lines show the preferred orientation of the graphite layers.

Dynamic testing (ASTM D 4065-94) using a sinusoidal stress wave at a controlled low frequency of 0.2 Hz is conducted at room temperature using a dynamic mechanical analyzer (DMA7, Perkin Elmer Corp., Shelton, CT). The variation of stress with time is illustrated in Fig. 2. Since the loss tangent decreases with increasing frequency, the low frequency is chosen in order to make the loss tangent measurement more accurate. The chosen frequency is far away from any vibration resonance frequency of the specimens.

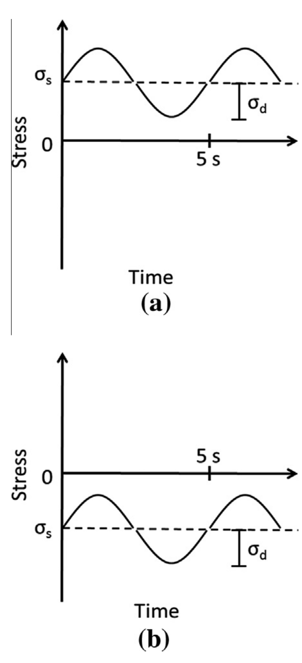

Fig. 2 – Schematic illustrations of the variation of the stress with time during dynamic mechanical testing. (a) Flexure. (b) Compression. The static and dynamic stresses are σ_s and σ_d respectively.

As in prior work [11], the dynamic stress σ_d used ranges from 1/3 to 2/3 of the corresponding static stress σ_s (Fig. 2). The static and dynamic stresses are much higher under flexure than compression, as chosen so that the static strain is comparable under flexure and compression. The flexural strain is calculated from the midspan deflection, span and specimen dimensions; the flexural stress is calculated from the load, span and specimen dimensions.

The static and dynamic stresses are progressively increased in the testing of a specimen. The stresses are chosen so that different specimens are tested at comparable values of the static strain (2%) and deformation amplitude (below 10 μm). Similar values of the static strain are obtained by adjusting the static stress and dynamic stress, such that the deformation amplitude is kept below 10 μm in order to avoid the presence of multiple vibration modes.

For flexural testing under three-point bending, the span is 20 mm, the static flexural stress ranges from 2.5 to 3.8 kPa (compared to 1.6–3.5 kPa in prior work [11]), the dynamic flexural stress ranges from 1.3 to 1.9 kPa (compared to 0.8–1.7 kPa in prior work [11]), the static flexural strain ranges from 1.9% to 2.1% (compared to 0.21–7.9% in prior work [11]), and the dynamic flexural strain ranges from 0.048 to 0.091% (compared to 0.022–0.085% in prior work [11]). For compressive testing, the static stress ranges from 236 to 1320 Pa (compared to 80–800 Pa in prior work [11]), the dynamic stress ranges from 118 to 900 Pa (compared to 40–400 Pa in prior work [11]), and the static strain ranges from 1.8–2.2% (compared to 0.34–13.8% in prior work [11]). Four specimens are tested for each combination of specimen type and test configuration.

3. Results and discussion

Table 1 and Fig. 3 show the dynamic flexural properties of exfoliated graphite compacts at various solid contents, with the static strain at 2%. As shown in Fig. 3(a), both the loss tangent and the loss-tangent/solid-content decrease with increasing solid content, but the curve is smoother for the latter, supporting the notion that the latter quantity is a better description of the material property than the former quantity and that the latter quantity reflects the inherent behavior of the cell wall. The latter quantity decreases monotonically from 35 with increasing solid content, such that it levels off at 0.9 at 15 vol.% solid. This means that the viscous character of the cell wall decreases with increasing solid content.

Preferred orientation of the graphite layers in the plane perpendicular to the direction of compaction occurs in an exfoliated graphite compact [4], as supported by X-ray diffraction [4,16] and by the low in-plane electrical resistivity compared to the out-of-plane resistivity of flexible graphite [17]. The degree of preferred orientation of the graphite layers increases with increasing solid content, as supported by the report that the in-plane resistivity decreases with increasing solid content [16]. A higher degree of preferred orientation is expected to facilitate the shear between the graphite layers during flexure. Nevertheless, the viscous character decreases with increasing solid content. Thus, this trend of the viscous character is attributed to the increasing difficulty of shear as the solid content increases.

Table 1 – Dynamic flexural properties of exfoliated graphite compacts at various solid contents, with the static strain at 2%.

Fabrication pressure (MPa)	0.25	0.32	0.60	1.15	1.70	2.40	3.56	5.51	7.71
Density (g/cm^3)	0.0236 ± 0.0019	0.0267 ± 0.0010	0.0401 ± 0.0012	0.0650 ± 0.0015	0.0891 ± 0.0018	0.1442 ± 0.0025	0.2124 ± 0.0066	0.2780 ± 0.0089	0.3503 ± 0.0102
Solid content (vol.%)[a]	1.04 ± 0.07	1.18 ± 0.04	1.77 ± 0.05	2.88 ± 0.07	3.94 ± 0.08	6.38 ± 0.11	9.40 ± 0.29	12.30 ± 0.39	15.51 ± 0.46
Loss tangent	0.363 ± 0.007	0.358 ± 0.008	0.318 ± 0.005	0.290 ± 0.005	0.239 ± 0.006	0.209 ± 0.003	0.171 ± 0.004	0.159 ± 0.004	0.143 ± 0.003
Storage modulus (MPa)	1.30 ± 0.10	1.33 ± 0.08	2.09 ± 0.08	3.10 ± 0.11	4.08 ± 0.18	5.66 ± 0.29	10.40 ± 0.64	11.90 ± 0.55	13.34 ± 0.52
Loss modulus (MPa)	0.47 ± 0.05	0.48 ± 0.06	0.66 ± 0.04	0.89 ± 0.05	0.96 ± 0.06	1.18 ± 0.11	1.77 ± 0.30	1.89 ± 0.22	1.91 ± 0.19
Loss-tangent/solid-content	34.9 ± 2.4	30.3 ± 1.2	18.0 ± 0.6	10.1 ± 0.3	6.1 ± 0.2	3.27 ± 0.07	1.81 ± 0.07	1.29 ± 0.05	0.92 ± 0.03
Storage-modulus/solid-content (MPa)	125 ± 13	113 ± 8	106 ± 6	108 ± 5	104 ± 5	89 ± 5	111 ± 7	97 ± 5	86 ± 4
Loss-modulus/solid-content (MPa)	45.2 ± 5.7	40.7 ± 5.3	37.3 ± 2.5	30.9 ± 1.9	24.4 ± 1.6	18.5 ± 1.8	18.9 ± 3.2	15.4 ± 1.9	12.3 ± 1.3

[a] Density divided by the graphite density (2.26 g/cm^3).

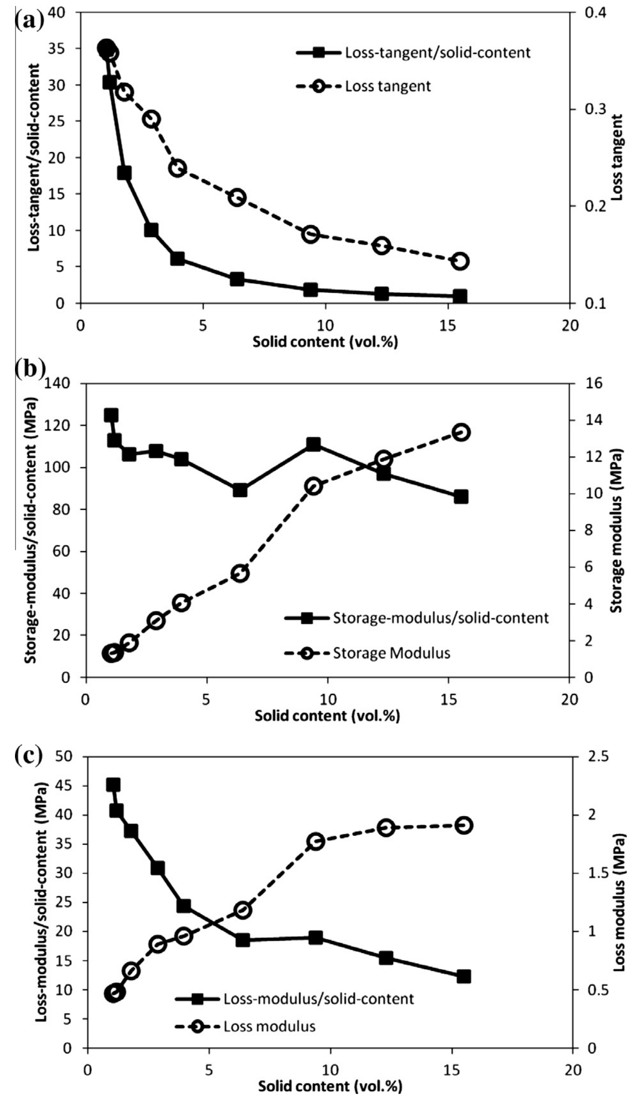

Fig. 3 – Dynamic flexural properties of exfoliated graphite compacts at various solid contents, with the static strain at 2%.

As shown in Fig. 3(b), the storage modulus of the compact under flexure increases with increasing solid content, as expected, since the stiffness is derived from the solid part of the compact. However, the storage-modulus/solid-content is relatively independent of the solid content, indicating that the stiffness of the cell wall is essentially independent of the solid content and that the Rule of Mixtures is essentially obeyed for the storage modulus. In spite of some irregularity in the variation of the storage-modulus/solid-content with the solid content, this quantity tends to decrease slightly with increasing solid content. This means that the stiffness of the cell wall tends to decrease slightly with increasing solid content, presumably due to the defects generated in the graphite layers during compaction and the increase in the amount of defects as the compaction pressure increases.

As shown in Fig. 3(c), the loss modulus of the compact increases with increasing solid content; this trend corroborates with the increase of the storage modulus of the compact with increasing solid content (Fig. 3(b)). However, the

Table 2 – Dynamic compressive properties of exfoliated graphite compacts at various solid contents, with the static strain at 2%. Note that this table addresses the compressive properties, whereas Table 1 addresses the flexural properties.

Fabrication pressure (MPa)	0.25	0.32	0.60	1.15	1.70	2.40	3.56	5.51	7.71
Density (g/cm^3)	0.0236 ± 0.0019	0.0267 ± 0.0010	0.0401 ± 0.0012	0.0650 ± 0.0015	0.0891 ± 0.0018	0.1442 ± 0.0025	0.2124 ± 0.0066	0.2780 ± 0.0089	0.3503 ± 0.0102
Solid content (vol.%)[a]	1.04 ± 0.07	1.18 ± 0.04	1.77 ± 0.05	2.88 ± 0.07	3.94 ± 0.08	6.38 ± 0.11	9.40 ± 0.29	12.30 ± 0.39	15.51 ± 0.46
Loss tangent	0.263 ± 0.005	0.258 ± 0.005	0.248 ± 0.003	0.238 ± 0.003	0.234 ± 0.003	0.219 ± 0.002	0.185 ± 0.003	0.160 ± 0.002	0.134 ± 0.003
Storage modulus (kPa)	0.52 ± 0.05	0.54 ± 0.04	0.83 ± 0.04	1.12 ± 0.07	1.34 ± 0.07	1.78 ± 0.06	2.90 ± 0.11	4.18 ± 0.19	5.09 ± 0.24
Loss modulus (kPa)	0.13 ± 0.02	0.13 ± 0.02	0.20 ± 0.02	0.26 ± 0.03	0.29 ± 0.03	0.39 ± 0.02	0.53 ± 0.04	0.67 ± 0.05	0.68 ± 0.05
Loss-tangent/solid-content	25.3 ± 3.1	21.9 ± 2.3	14.0 ± 1.7	8.6 ± 1.1	5.9 ± 0.7	3.4 ± 0.3	1.9 ± 0.3	1.3 ± 0.2	0.9 ± 0.2
Storage-modulus/solid-content (kPa)	46 ± 5	42 ± 4	45 ± 3	38 ± 3	33 ± 2	28 ± 1	31 ± 2	34 ± 2	33 ± 2
Loss-modulus/solid-content (kPa)	12.5 ± 2.1	11.0 ± 1.7	11.3 ± 1.2	9.0 ± 1.1	7.4 ± 0.8	6.1 ± 0.4	5.6 ± 0.5	5.4 ± 0.4	4.3 ± 0.3

[a] Density divided by the graphite density (2.26 g/cm^3).

loss-modulus/solid-content decreases with increasing solid content; this trend corroborates with the decrease of the loss-tangent/solid-content with increasing solid content (Fig. 3(a)). In other words, the loss modulus of the cell wall is mainly governed by the degree of viscous character, whereas the loss modulus of the compact is mainly governed by the stiffness of the compact.

The loss modulus of the compact increases with increasing solid content, such that it starts to level off at around 9 vol.%, with the leveled-off value at 1.9 MPa. The loss-modulus/solid-content decreases with increasing solid content, such that it starts to level off at around 6 vol.%, with the leveled-off value at 12–18 MPa. This means that, for mechanical energy dissipation using the compact, the solid content should be at least 9 vol.%. However, for mechanical energy dissipation using a cell wall (assuming that a cell wall can be isolated), the lowest possible solid content is most effective.

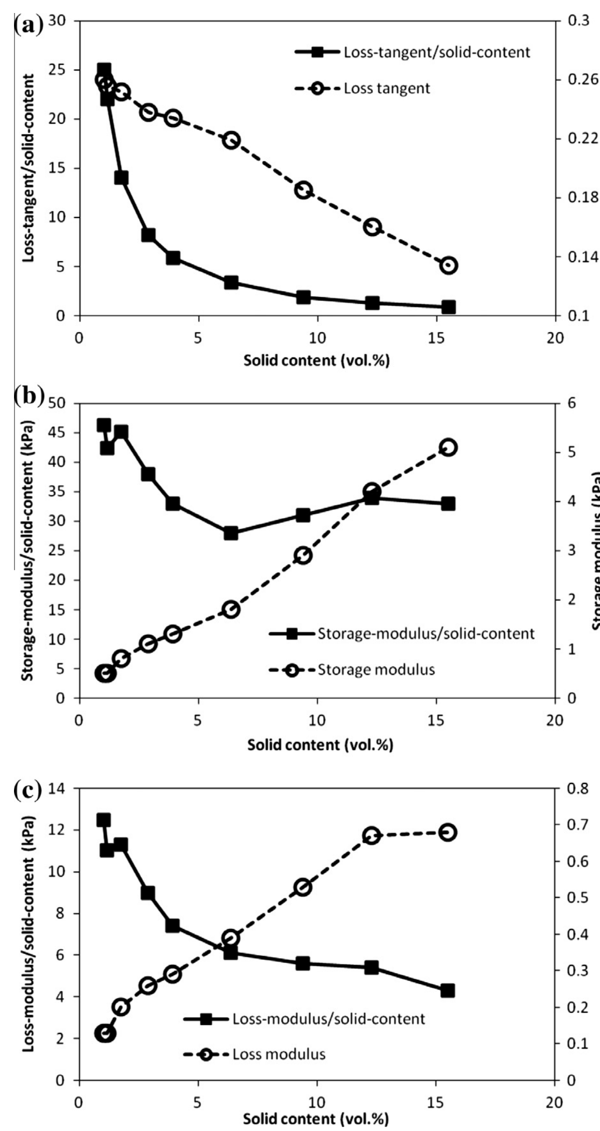

Fig. 4 – Dynamic compressive properties of exfoliated graphite compacts at various solid contents, with the static strain at 2%.

In prior work [11], it was reported that, under flexure, the storage modulus is essentially unaffected by slightly increasing the static strain, while the loss tangent and loss modulus decrease. A small increase in the static strain relates to a small increase in the solid content. These effects of the static strain (over a small range) are not consistent with the effects observed in this work for the effect of the solid content (over a large range) on these quantities, but are consistent with the effects of the solid content on the storage-modulus/solid-content (Fig. 3(b)), the loss-tangent/solid content (Fig. 3(a)) and the loss-modulus/solid-content (Fig. 3(c)). The consistency with the normalized quantities relates to the notion that the deformation involves shear of the graphite layers relative to one another and that this deformation occurs in the solid part of the compact.

Table 2 and Fig. 4 show the dynamic compressive properties of exfoliated graphite compacts at various solid contents. The loss tangent of the compact decreases with the increasing solid content, such that it does not level off. However, the loss-tangent/solid-content decrease with increasing solid content, such that it levels off at around 15 vol.% (Fig. 4(a)), as in the case under flexure (Fig. 3(a)). The loss-modulus/solid-content decreases with increasing solid content, such that it starts to level off at around 6 vol.% (Fig. 4(c)), as in the case of flexure (Fig. 3(c)). The consistency in the leveling-off behavior between the loss-tangent/solid-content under compression and flexure and the consistency in the leveling-off behavior between the loss-modulus/solid-content under compression and flexure support the notion that these quantities reflect the viscous behavior of the cell wall.

In prior work [11], it was reported that, under out-of-plane compression, both storage and loss moduli increase with increasing static strain over a small range, while the loss tangent slightly decreases. These trends are consistent with the effect of the solid content (over a wide range) on these quantities, as observed in this work, but are not consistent with the effect of the solid content on the storage-modulus/solid-content (Fig. 4(b)), the loss-modulus/solid-content (Fig. 4(c)) and the loss-tangent/solid-content (Fig. 4(a)). The inconsistency with the normalized quantities relates to the fact that the out-of-plane compression involves relatively little shear of the graphite layers relative to one another and hence a less clear-cut role of the solid part.

The trends for the variation of the dynamic compressive properties with the solid content (Fig. 4) are essentially the same as those for the variation of the dynamic flexural properties (Fig. 3). However, the magnitudes of the property values are very different between compression and flexure. The loss tangent is smaller for compression (out-of-plane) than flexure, as expected from the notion that the viscous character is mainly derived from the shear of the graphite layers relative to one another. This difference is consistent with the prior report [11] that the loss tangent is higher for in-plane compression than out-of-plane compression.

Under out-of-plane compression, shear between the graphite layers occurs to a limited degree, due to the preferred orientation of the graphite layers. Due to the in-plane stiffness of the graphite layers, the bending of the graphite layers is not expected to contribute significantly to the viscous character. Due to the limited specific surface area (Section 2), which corresponds to an average of about 60 graphite layers stacked up in a cell wall, the friction between air and the graphite surface during mechanical deformation is expected to contribute insignificantly to the viscous character. Therefore, we believe that the limited shear between the graphite layers is the main mechanism of the limited viscous character under out-of-plane compression.

The storage modulus is smaller for compression than flexure by four orders of magnitude, as expected from the preferred orientation of the graphite layers in the compact and the much greater stiffness of graphite in the in-plane direction than the out-of-plane direction. The loss modulus is smaller for compression than flexure by three orders of magnitude, also due to the preferred orientation and graphite anisotropy.

Table 3 shows the ratio of the flexural loss-tangent/solid-content to the out-of-plane compressive loss-tangent/solid-content. This ratio decreases with increasing solid content, such that it is 1.4 at the lowest solid content of 1.04 vol.% and is 1.0 at solid content $\geqslant$3.94 vol.%. Since shear of the graphite layers relative to one another is much less significant for out-of-plane compression than flexure, a ratio of 1.0 means that the shear mechanism contribution to the viscous character is limited to the relatively low level that occurs under out-of-plane compression. Hence, the shear-driven viscous character occurs extensively under flexure only when the solid content is below 4 vol.%.

Under uniaxial compression at the same frequency of 0.2 Hz, a carbon black (Vulcan XC72R, Cabot Corp., Billerica, MA, USA., with specific surface area 254 m^2/g) compact exhibits loss-tangent/solid-content 1.2 [18]. This is the highest value among quite a variety of carbon blacks [18], but it is

Table 3 – Ratio of the flexural loss-tangent/solid-content to the compressive loss-tangent/solid-content.

Solid content (vol.%)[a]	1.04 ± 0.07	1.18 ± 0.04	1.77 ± 0.05	2.88 ± 0.07	3.94 ± 0.08	6.38 ± 0.11	9.40 ± 0.29	12.30 ± 0.39	15.51 ± 0.46
Flexural loss-tangent/solid-content	34.9 ± 2.4	30.3 ± 1.2	18.0 ± 0.6	10.1 ± 0.3	6.1 ± 0.2	3.27 ± 0.07	1.81 ± 0.07	1.29 ± 0.05	0.92 ± 0.03
Compressive loss-tangent/solid-content	25.3 ± 3.1	21.9 ± 2.3	14.0 ± 1.7	8.6 ± 1.1	5.9 ± 0.7	3.4 ± 0.3	1.9 ± 0.3	1.3 ± 0.2	0.9 ± 0.2
Ratio	1.4 ± 0.2	1.4 ± 0.2	1.3 ± 0.1	1.2 ± 0.1	1.0 ± 0.1	1.0 ± 0.1	1.0 ± 0.1	1.0 ± 0.1	1.0 ± 0.2

[a] Density divided by the graphite density (2.26 g/cm^3).

much smaller than the highest value of 25 obtained in this work for an exfoliated graphite compact under dynamic compression. The viscous character of the carbon black compact stems from the movement among the particles in a carbon black aggregate [18]. If the carbon black were graphitic, the specific surface area of 254 m^2/g corresponds to a structure in which each graphite region consists of 10 graphite layers stacked up. Such a structure, though too idealistic, is consistent with the nanoparticulate structure of the carbon black (particle size 30 nm). Based on geometric considerations, the interface area per unit volume is greater between the graphite layers in the cell wall of exfoliated graphite than that between the particles in a carbon black aggregate. Therefore, the highest value of the loss-tangent/solid-content is much greater for an exfoliated graphite compact than a carbon black compact. This contrast between an exfoliated graphite compact and a carbon black compact further supports the notion that the sliding of the graphite layers relative to one another in the cell wall of exfoliated graphite contributes largely to the viscous behavior of exfoliated graphite.

The compressive storage-modulus/solid-content of the carbon black compact that exhibits the abovementioned value of 1.2 for the loss-tangent/solid-content is 27 kPa [18], which is lower than the compressive storage-modulus/solid-content value of 46 kPa for the exfoliated graphite compact that exhibits the highest value of 25 for the loss-tangent/solid-content. This means that the cell wall of exfoliated graphite is stiffer than the solid part of a carbon black compact.

The compressive loss-modulus/solid-content of the carbon black compact that exhibits the abovementioned value of 1.2 for the loss-tangent/solid-content is 1 kPa [18], which is lower than the compressive storage-modulus/solid-content value of 13 kPa for the exfoliated graphite compact that exhibits the highest value of 25 for the loss-tangent/solid-content. This means that the cell wall of exfoliated graphite is superior to the solid part of a carbon black compact in terms of the viscous character, the stiffness and the mechanical energy dissipation ability.

Rubber is a well-known viscoelastic material [19]. Under flexure at the same frequency of 0.2 Hz, Neoprene rubber exhibits loss tangent 0.7 [10]. Since the solid content is 100% for the rubber, the loss-tangent/solid-content is also 0.7, which is lower than those of all the exfoliated graphite compacts of this work. The corresponding storage modulus of this rubber is 7.5 MPa [10], so that the storage-modulus/solid-content is also 7.5 MPa, which is much lower than the value of 125 MPa for an exfoliated graphite compact. The corresponding loss modulus of this rubber is 6.7 MPa [10], so that the loss-modulus/solid-content is also 6.7 MPa, which is smaller than the value of 45 MPa for an exfoliated graphite compact. Hence, the cell wall of exfoliated graphite is superior to rubber in terms of the viscous character, the stiffness and the mechanical energy dissipation ability.

4. Conclusion

The cell wall (with about 60 graphite layers on the average) in the cellular structure of an exfoliated graphite compact is viscoelastic, due to the shear between graphite layers. The viscous character of the cell wall decreases with increasing solid content (corresponding to increasing compaction pressure), due to the increasing difficulty of shear between the graphite layers. The elastic character of the cell wall is essentially independent of the solid content, essentially in accordance with the Rule of Mixtures, though it tends to decrease slightly with increasing solid content.

The viscous character is greater under in-plane flexure than uniaxial compression in the compaction direction, due to the preferred orientation of the graphite layers in the plane perpendicular to the compaction direction and the consequent stiffness anisotropy. At the lowest solid content of 1.0 vol.% (99.0 vol.% voids), the loss-tangent/solid-content is 35 and 25 under flexure and compression respectively, the storage-modulus/solid-content is 125 MPa and 46 kPa under flexure and compression respectively, and the loss-modulus/solid-content is 45 MPa and 13 kPa under flexure and compression respectively. The loss-tangent/solid-content decreases with increasing solid content, leveling off at 0.9 at 15 vol.% solid, whether under flexure or compression. The loss-modulus/solid-content decreases with increasing solid content, leveling off at 6 vol.% solid at 19 MPa and 6.1 kPa for flexure and compression respectively. The shear-driven viscous character occurs extensively under flexure only when the solid content is below 4 vol.%.

The cell wall of exfoliated graphite is superior to rubber and the solid part of a carbon black compact in the viscous character, the stiffness and the mechanical energy dissipation ability. The highest values of the loss-tangent/solid-content, the storage-modulus/solid-content and the loss-modulus/solid-content are much greater for an exfoliated graphite compact than rubber or a carbon black compact.

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