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Art and Science of Reinforcing Ceramics with Graphene via Ultrasonication Mixing

Jiqiang Wu, Maxim Trubyanov, Delia Prvački, Karen Lim, Daria V. Andreeva,* and Kostya S. Novoselov

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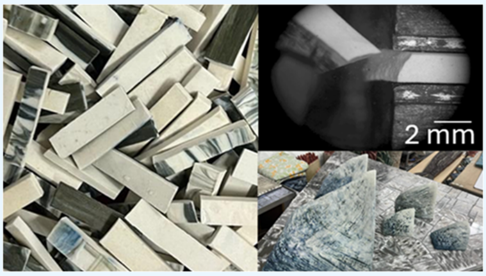

ABSTRACT: This work presents an interdisciplinary approach combining materials science, ultrasonication, artistic expression, and curatorial practice to develop and investigate novel composites. The focus of the approach is incorporating graphene oxide (GO) into kaolin and exploring its effects on material properties. The composites were prepared with varying GO concentrations and sonication times, and their mechanical, thermal, and morphological characteristics were evaluated. The results reveal that the addition of 0.5 wt % GO, combined with a sonication time of 10 min, leads to the highest storage modulus and improved thermal stability. Ultrasonication proved to be an effective method for dispersing and distributing GO particles within the kaolin matrix, resulting in an enhanced material performance. Furthermore, the application of novel composites provided by Prvački adds a unique dimension to the study. Through the artistic interpretation, the tactile qualities and aesthetic potential of the composites are explored, shedding light on the transformative power of materials and cultural significance organized as part of an artist-in-residence commission, introduced in conjunction with the NUS Public Art Initiative. This interdisciplinary collaboration accompanied by an exhibition at the NUS Museum demonstrates the value of merging scientific research, technological advancements, and artistic exploration.

1. INTRODUCTION

The combination of art and science in the field of graphene[1] and, in particular, its reinforcement of ceramics via ultrasonication[2] offers an intersection of materials engineering, advanced manufacturing, and artistic expression. Graphene,[3] a two-dimensional carbon material known for its mechanical, electrical, and thermal properties, can be incorporated into ceramic matrices to enhance their performance.[4] Graphene and its derivatives naturally interact with many areas of technology and art. They not only provide new materials to improve the performance of existing applications but also introduce novel concepts that have the potential to revolutionize entire fields. Custom manufacturing, multifunctionality, responsivity, built-in intelligence are just a few of the possibilities that sustainable material science offers to end user.

In this work, we created novel composite materials using sonicated mixtures of kaolin and an oxidized form of graphene, graphene oxide (GO). GO is an amphiphilic compound[5] compatible with a variety of materials, including inorganic[6] and organic[7] composite systems, to enhance their properties. GO is frequently employed to improve the mechanical properties of ceramics, such as toughness, strength, and fracture resistance.[8−11] When incorporated into ceramic matrices, GO serves as a reinforcing agent by inhibiting crack propagation,[12,13] thereby enhancing the material's overall durability.[14] Additionally, GO can improve the environmental stability of ceramics,[15] making them more resistant to high-temperature environments and thermal shock.[16,17]

Mixing GO with ceramics involves incorporating GO into the ceramic matrix.[18,19] Typically, GO is first dispersed in water to create a stable aqueous solution or suspension. This GO dispersion is then added to the ceramic slurry, which consists of solid particles, water, and sometimes additional admixtures. To achieve a uniform distribution of GO throughout the inorganic matrix, the mixture is thoroughly homogenized by using mechanical mixers or high-shear mixing equipment. In this work, we employed a fast and energy-efficient high-intensity ultrasonication technique that accomplishes three critical tasks simultaneously: (1) dispersion of GO, (2) mixing GO with the ceramic matrix, and (3) ensuring uniform distribution of GO throughout the matrix.

High-intensity ultrasonication,[20] a technique that utilizes low-frequency sound waves (20−100 kHz), is used for dispersing[21,22] and incorporating functional particles into solid matrices.[23] The process involves the application of

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ultrasonic energy to create cavitation, which generates intense localized forces and promotes the exfoliation[24] and dispersion of graphene sheets[25] within the ceramic matrix.[26] This results in improved mechanical strength, toughness,[27] radical scavenging,[28] and thermal properties[29] of the architectural composite material.[30]

In this work, we demonstrated that the combination of ultrasonication and the addition of GO leads to enhanced mechanical properties of kaolin while also improving the processability and the appearance of the materials. As an application, we explored artistic potential by experimenting with novel ceramic forms and textures. We illustrated how harnessing the properties of graphene pushes the boundaries of traditional ceramic art, enabling the creation of delicate and intricate ceramic structures that were previously unattainable. This approach not only enhances the artistic possibilities but also can potentially reduce the need for structural elements, making the overall process more sustainable.

2. RESULTS AND DISCUSSION

2.1. Enhanced Stiffness and Mechanical Properties of Graphene-Reinforced Ceramics Prepared via Ultrasonication. Figure 1 presents the storage moduli measured for the samples prepared using different concentrations of GO and varying sonication times. It is important to note that we sintered all samples at 1100 °C for 1.5 h in a kiln with a heating rate of 18 °C/min.

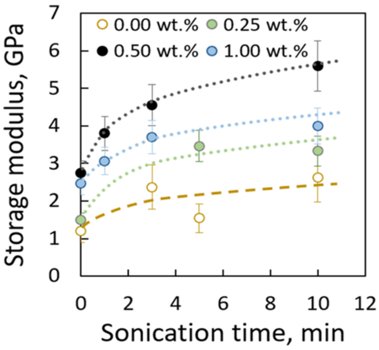

Figure 1. Enhanced stiffness and mechanical properties of graphene-reinforced ceramics prepared via ultrasonication. The effect of sonication time and GO concentration (indicated in wt %) on the storage modulus.

Using dynamic mechanical analysis (DMA), we observe that the samples sonicated with 0.5 wt % of GO exhibit the highest storage modulus, specifically measuring 5.5 GPa. In comparison, samples prepared with GO concentrations below 0.5 wt % and above 0.5 wt % demonstrate lower storage moduli. Notably, the storage modulus values obtained for the sonicated pure kaolin samples, which are approximately 2 GPa, are surpassed by those of all of the samples containing GO.

These findings indicate that the addition of GO, specifically at a concentration of 0.5 wt %, enhances the stiffness and mechanical properties of the ceramic matrix. The storage modulus serves as a measure of a material's ability to store and release energy elastically, reflecting its stiffness and resistance to deformation. The higher storage modulus exhibited by the 0.5 wt % GO samples after sonication suggests an improved reinforcement effect and enhanced structural integrity compared to both lower and higher GO concentration samples, as well as the sonicated kaolin samples without GO.

It is noted that the samples mixed without sonication display smaller storage moduli compared with the sonicated samples. The storage modulus of kaolin without GO is approximately 1 GPa. The samples containing 0.5 wt % of GO exhibit a significantly higher storage modulus, measuring around 2.7 GPa. This indicates an improvement in stiffness and mechanical properties compared with the kaolin-only samples. The addition of GO at this specific concentration appears to enhance the reinforcement effect, resulting in an increase in the storage modulus of the composite material. However, the largest storage modulus is achieved by a combination of both sonication and addition of GO.

Thus, the results presented in Figure 1 demonstrate the impact of the GO concentration and sonication time on the mechanical behavior of the composite materials. The optimal combination of GO concentration and sonication time can lead to significant improvements in the storage modulus, indicating enhanced mechanical performance and the potential for various applications in the field of materials science.

2.2. Effect of the GO Content on the Thermal Stability of Ultrasonicated Ceramic Composites. The thermal decomposition behavior of samples with different GO contents and a fixed sonication time was investigated using thermogravimetric analysis (TGA) in a nitrogen (N_2) atmosphere. The TGA results, presented in Figure 2, provide insights into the effect of the GO content on the thermal stability of the ceramic composites.

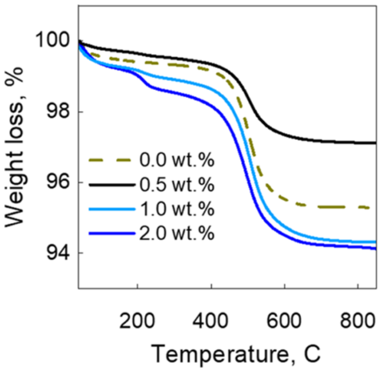

Figure 2. Thermal decomposition behavior of samples with various GO content (indicated in wt %). TGA analysis.

The analysis of the TGA curves identified three distinct regions: below 200 °C, 200−600 °C, and above 600 °C. At lower temperatures (below 200 °C), the physically adsorbed water in kaolin is removed.[31] This is the initial dehydration stage, where water molecules that are loosely bound to the kaolin surface evaporate. As we see, the presence of 1 and 2 wt % of GO leads to a more efficient removal of water due to increased surface interactions and possibly faster heat transfer within the composite.

In the temperature range (200−600 °C), kaolin undergoes dehydroxylation,[32,33] where chemically bound water (from the hydroxyl groups in the kaolin structure) is released, leading to the formation of metakaolin.[34] The TGA curves show that the addition of GO enhances the dehydroxylation process by providing additional sites for heat absorption and facilitating the removal of hydroxyl groups. At this stage, kaolin loses its crystalline structure and becomes an amorphous phase.

The samples prepared with higher GO content, specifically 1 and 2 wt %, show an increase in the weight loss during thermal decomposition. These samples lose around 4 and 6 wt % of their weight, respectively, when heated up to 900 °C. That might be caused by the structural damage[6] that occurs through the extrusion of carbon dioxide generated by heating GO to 1050 °C.

In contrast, we observe that the samples with 0.5 wt % GO exhibit a weight loss of approximately 2 wt % when heated to 900 °C. Thus, the TGA analysis highlights the significance of the GO content on the thermal stability of the ultrasonicated ceramic composites. The optimum GO content of 0.5 wt % demonstrates the highest thermal stability, while excessive GO loading can lead to reduced thermal stability.

2.3. Characterization of Morphological Changes and GO Distribution in Kaolin-GO Composites via SEM and EDX Mapping. As shown in the digital photos in Figure 3, the samples containing GO before firing have a dark gray color (Figure 3b), which is attributed to the GO powder. After firing, both the samples with GO (Figure 3c) and those without GO (Figure 3a) exhibit a white-yellowish color, typical of kaolin.

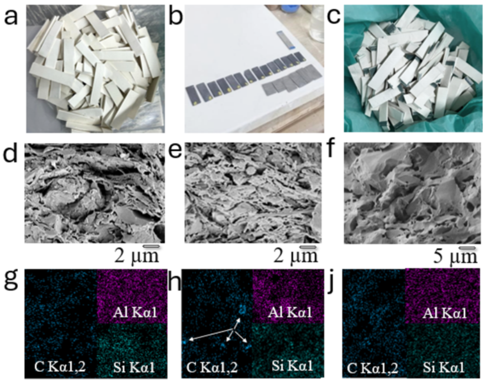

Figure 3. (a−c) Photos of the samples: fired kaolin (a); unfired kaolin with different concentrations of GO (b); fired kaolin with 0.5 wt % of GO (c). (d−f) Cross-sectional SEM images: fired kaolin (d); unfired kaolin with 0.5 wt % of GO (e); fired kaolin with 0.5 wt % of GO (f). (g−j) EDX mappings: fired kaolin (g); unfired kaolin with 0.5 wt % of GO (h). GO particles are highlighted; fired kaolin with 0.5 wt % of GO (j).

Scanning electron microscopy (SEM) images in Figure 3d−f reveal the morphological changes of the kaolin samples with the addition of GO particles. The kaolin samples, both with and without GO, exhibit a similar layered morphology with irregular particle shapes and sizes. This indicates that the presence of GO particles does not cause drastic changes in the microscopic morphology of the kaolin material. The fundamental structure of the kaolin matrix remains intact, and the GO particles do not appear to significantly alter the overall shape or arrangement of the kaolin particles. The fired samples containing 0.5 wt % of GO exhibit a slightly denser and smoother morphology.

The distribution of GO particles was investigated by using complementary analysis, such as energy-dispersive X-ray spectroscopy (EDX) mapping (Figure 3g−j). By performing EDX mapping, it is possible to identify and visualize the distribution of carbon atoms, including graphene, within the kaolin matrix. The presence of GO particles can be detected in the unfired samples (Figure 3h) by analyzing the elemental mapping corresponding to carbon, which is a characteristic element of GO. The mapping shows localized regions with higher carbon signals, indicating the presence of GO particles within the kaolin matrix after sonication. After firing, EDX mapping (Figure 3j) shows the presence of Al and Si in the samples but not carbon, indicating the thermal decomposition of GO particles.

2.4. Impact of GO on the Chemical Transformations of Kaolin. The Fourier-transform infrared (FTIR) spectra in Figure 4 demonstrate the impact of firing on the phase transformations of kaolin in the presence and absence of GO. In its raw, unfired state, kaolin exhibits characteristic O−H and Si−O vibrations, with key peaks at approximately 3695 cm^{-1} (O−H stretching) and 1035 cm^{-1} (Si−O stretching). Upon firing, the material undergoes dehydroxylation, leading to the formation of new crystalline phases such as mullite (Si−O−Si bending vibrations around 765 cm^{-1} and Si−O stretching vibrations between 1050−1150 cm^{-1}), cristobalite (Si−O stretching vibrations in the 800−1100 cm^{-1} range), and metakaolin (broad Si−O stretching vibrations typically appearing in the range of 950−1000 cm^{-1}).

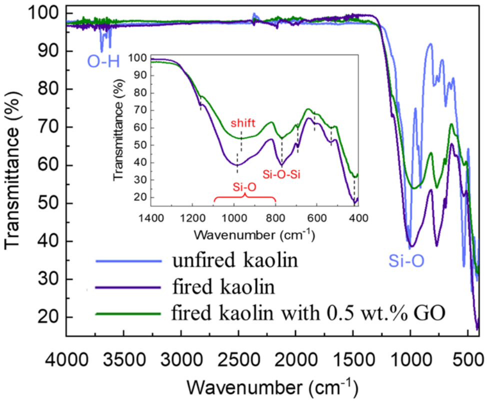

Figure 4. FTIR spectra of unfired kaolin (blue), fired kaolin without GO (lilac), and fired kaolin with 0.5 wt % of GO (green).

As seen in the spectral comparison, the introduction of GO alters Si−O vibrational modes. The presence of GO appears to facilitate the more uniform formation of an amorphous phase like metakaolin, as evidenced by the less intense absorption band in the Si−O stretching region of 800−1100 cm^{-1}, with the peak shifting to a lower wavenumber around 975 cm^{-1}. In contrast, the FTIR spectrum of the GO-free fired sample shows a more intense and narrower Si−O stretching band, with the peak shifting to a higher wavenumber around 990 cm^{-1}.

The uniform amorphization induced by GO enhances the material's ability to absorb stress and deform without fracturing. Additionally, the presence of GO during the early stages of firing may influence the sintering behavior, potentially acting as a flux and promoting densification with improved bonding at the grain boundaries. This densification leads to a denser morphology in the final material, as observed through SEM, thereby enhancing its mechanical strength.

2.5. Visualization of the Breakage Behavior of Samples Prepared under Different Conditions. As part of an exhibition at the NUS Museum, we visualized the breakage behavior of kaolin that was fired (Figure 5a), fired and sonicated (Figure 5b), and kaolin with 0.5 wt % GO that

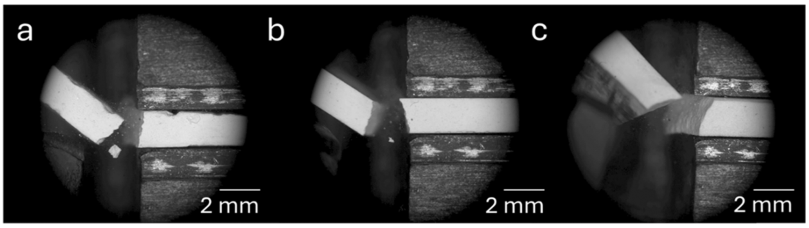

Figure 5. Snapshots from high-speed video recording show different breakage behaviors of samples prepared under varying conditions: a, fired kaolin sample; b, fired kaolin sample after sonication; c, fired kaolin sample with 0.5 wt % of GO after sonication. High-speed video recording was performed at a resolution of 1024 × 1024 pixels and a frame rate of 12,800 fps, with a shutter speed set to 76,100 ns.

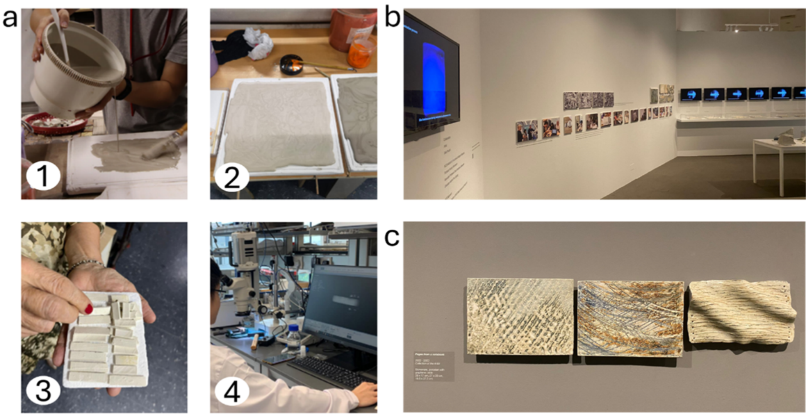

Figure 6. (a) The translation of scientific methodology from the chemical laboratory to the art studio: (1) the preparation of mixture; (2) casting; (3) the preparation of samples, (4) the visualization of breakage process. (b) The photo of the exhibition in the NUS Museum. (c) Artistic ceramics by Delia Prvački.

was fired and sonicated (Figure 5c) using a high-speed camera. The snapshots provide a visual comparison of how different preparation methods—firing, sonication, and the addition of GO—affect the structural integrity of the kaolin samples for broad audience of the museum.

As observed, the fired sample demonstrates the formation of relatively large flake-like debris upon breakage, indicating the lamellar morphology of fired kaolin. Fired kaolin after sonication produces finer debris, suggesting that sonication improves the homogeneity of the kaolin and may reduce the particles size. The combination of GO and sonication, followed by firing, results in breakage behavior without debris formation. This suggests enhanced reinforcement that alters mechanical characteristics due to the synergistic effects of GO and ultrasonic treatment, indicating an overall improvement in the material's structural integrity.

2.6. Exploring the Art-Science Intersection. The enhanced mechanical strength ensures that the ceramics are more robust and durable, making them suitable for practical applications and handling during transportation and installation. Improved thermal stability makes the ceramics more versatile in various environmental conditions, enhancing their longevity and functionality. This is particularly valuable in both functional applications and art installations exposed to varying temperatures. The ability to control surface texture enhances the visual and tactile appeal of ceramic pieces, adding depth and interest to the artwork.

Figure 6a(1−4) is a photographic documentation of the collaborative process, illustrating the translation of scientific methodology from the chemical laboratory to the art studio. It demonstrates that art-making and innovation are not a matter of chance; they are methodical and grounded in technical principles. The use of GO in ceramics pushes the boundaries of traditional ceramic techniques, enabling the creation of contemporary pieces that stand out in an art exhibition.

This artistic exploration prompts broader questions about the creation, perception, and significance of ceramics as cultural artifacts. It emphasizes the varied contributions and histories behind each piece and its role in global ceramic production. By combining art and science, this collaborative project deepens our understanding of the material, its artistic expression, and its wider impact.

Exploring the novel composite materials through an artistic lens encourages further materials studies and invites reflection on the social, philosophical, and economic aspects, such as trade and cultural value. By merging art and science, this exploration promotes interdisciplinary dialog and deepens our understanding of materials, their artistic possibilities, and their cultural significance.

The artistic exploration of a novel composite material, combining kaolin with GO and sonication, offers insights into its material properties and cultural significance. Artist Delia Prvaki evaluated the composite through tactile and visual means. The scientific data and artistic creations were exhibited at the NUS Museum as part of the exhibition "Continuity, Persistence, Line: Thinking Through Clay"—a selection of works by Delia Prvački, which opened in August 2024 (Figure 6b,c).

3. CONCLUSIONS

In conclusion, this collaborative work encompassing materials science, ultrasonication, artistic exploration, and curatorial practice has yielded significant insights into the development and characterization of novel composites. Through the incorporation of graphene oxide (GO) into kaolin, enhanced material properties were achieved, as evidenced by the observed improvements in storage modulus, thermal stability, and morphology.

The systematic investigation of GO concentration and sonication time has provided valuable data on the influence of these parameters on the mechanical and thermal behavior of the composites. The findings demonstrate that an optimal concentration of 0.5 wt % GO, combined with a sonication time of 10 min, leads to the highest storage modulus and improved thermal stability. Combining ultrasonication and GO allows achieving uniform materials processing. This resulted in denser morphologies and improved thermal stability and material performance.

Furthermore, artistic exploration has added a unique dimension to this collaborative work. Prvački's interpretation of the composites has provided valuable insights into the transformative power of materials, the evolution of form, and the serendipitous surface effects achieved in these novel composites. This artistic perspective enriches our understanding of the materials beyond their scientific properties, engaging with the cultural and conceptual aspects associated with them.

4. EXPERIMENTAL SECTION

4.1. Materials.

For composites preparation, we used a 0.4 wt % GO dispersion in DI water (monolayer >95%, Graphenea Inc.). The GO dispersion provided by Graphenea had a carbon content (C) ranging from 49 to 56%, hydrogen content (H) ranging from 0 to 1%, nitrogen content (N) ranging from 0 to 1%, sulfur content (S) ranging from 2 to 3%, and oxygen content (O) ranging from 41 to 50%. GO dispersions were sonicated (40 kHz, 55 $W \cdot L^{-1}$) for 0.5 h before use, where monolayer flakes with average sizes of 2.1 ± 0.7 μm were obtained. The Raman spectroscopy analysis showed peaks at 1348 cm^{-1} (D band) and 1585 cm^{-1} (G band) with an ID/IG ratio of 1.02. The FTIR spectrum displayed characteristic peaks corresponding to the O−H, C═O, C═C, and C−O functional groups of GO.

Kaolin type WSS, White Porcelain Casting Slip (Firing range: orton cone 8 to 9) was sourced from Clayworks Potters Supply Pty Ltd., Australia. It is a specific type of kaolin clay formulation used in the production of white porcelain ceramics. The chemical formula is $Al_2Si_2O_5(OH)_4$, indicating that it consists of aluminum (Al), silicon (Si), oxygen (O), and hydrogen (H) atoms.

4.2. Samples Preparation.

First, we centrifuged kaolin at 1000 rpm for 20 min using an Allegra X-30R centrifuge. This step helps to separate the water from the kaolin particles. Once the centrifugation is complete, the water is carefully removed and the tube containing the kaolin sediment is transferred to a convection oven. The oven temperature is set to 80 °C, and the kaolin sample is left to dry completely overnight. After drying, the dry kaolin is ground into a powder. An appropriate amount of the kaolin powder is weighed, and various concentrations of GO are added ranging from 0 to 1 wt %. Water is then added to the samples, bringing the volume of the kaolin-GO-water suspension to 400 mL. The resulting suspensions were sonicated using a UIP1500hdT probe sonicator equipped with a 40 mm titanium tip. The sonication times were 1, 3, 5, and 10 min. Then we pour the suspension into the homemade gypsum molds to form rectangular samples. The sample size was 33 mm long, 13 mm wide, and 2 mm thick and letting them dry at ambient conditions. After that, we fired the samples at 1100 °C for 1.5 h in a kiln (Tabletop Furnace Company) with a heating rate of 18 °C/min.

4.3. Instrumental Characterizations.

The samples were characterized by a scanning electron microscope (SEM) model Supra 40 from Carl Zeiss, an energy-dispersive X-ray spectroscopy (EDX) system (Oxford Ulti Max 65) from Oxford Instruments, and a Discovery TGA (thermogravimetric analyzer) from TA Instruments. Thermal decomposition was studied under an inert N_2 atmosphere in a 30−900 °C temperature range at a 10 °C/min ramping rate. The storage modulus of the materials was measured using a DMA 850 instrument from TA Instruments, operating in a single cantilever oscillation mode with 5−50 μm amplitude in a 1−200 Hz frequency range at room temperature. This instrument allowed us to characterize the mechanical properties of the materials. Fourier-transform infrared (FTIR) spectra were recorded using an IRTracer-100 instrument (Shimadzu, Japan) equipped with a diamond attenuated total reflectance (ATR) accessory. The spectra were collected over the range of 4000−400 cm^{-1}, with a resolution of 4 cm^{-1}, and 32 scans were coadded to improve the signal-to-noise ratio. Raman spectroscopy (WITEC ALPHA300R, 532 nm laser) was used for the characterization of GO properties. High-speed recording was performed using a FASTCAM Nova S12 (Photron) at a resolution of 1024 × 1024 pixels and a frame rate of 12,800 FPS, with a shutter speed set to 76,100 ns.

■ AUTHOR INFORMATION

Corresponding Author

Daria V. Andreeva − *Department of Materials Science and Engineering, National University of Singapore, 117575, Singapore; Institute for Functional Intelligent Materials, National University of Singapore, 117544, Singapore;* orcid.org/0000-0003-0273-2064; Email: daria@nus.edu.sg

Authors

Jiqiang Wu − *Department of Materials Science and Engineering, National University of Singapore, 117575, Singapore*

Maxim Trubyanov − *Department of Materials Science and Engineering, National University of Singapore, 117575, Singapore; Institute for Functional Intelligent Materials, National University of Singapore, 117544, Singapore;* orcid.org/0000-0001-6290-6201

Delia Prvački − *Deliarts Pte Ltd., 757718, Singapore*

Karen Lim − *NUS Museum & Secretariat, Public Art Committee, National University of Singapore, 119279, Singapore*

Kostya S. Novoselov − *Department of Materials Science and Engineering, National University of Singapore, 117575, Singapore; Institute for Functional Intelligent Materials, National University of Singapore, 117544, Singapore*

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.4c05748

Notes

The authors declare no competing financial interest.

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