Heat capacity features of epoxy-based composites with different graphene oxide contributions at low temperatures

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The study of the properties of materials with impurities is of current interest and has important scientific and technical implications. The introduction of impurities makes it possible to obtain a new material with completely different unique properties, which depend on both the type of impurity and its amount. In recent years, a considerable amount of work has been devoted to the study and preparation of epoxy composite nanomaterials. [1]

In this work, samples of epoxy with graphene oxide (GO) at concentrations of 0.25%, 0.5%, 3%, 6% and pristine epoxy have been studied. The heat capacity of the epoxy-based samples was measured using Quantum Design Physical Property Measurement System (PPMS) in the temperature range from 2 K to 50 K.

[1] D. E. Hurova, S. V. Cherednichenko, N. A. Aksenova, N. A. Vinnikov, A. V. Dolbin, and N. N. Galtsov, Low Temperature Physics, 50, 2, 177 (2024). https://doi.org/10.1063/10.0024329

Samples:

pristine epoxy

epoxy with graphene oxide (GO)

Sample preparation procedure:

The composite was prepared according to the following procedure. The required amount of TRGO was added to the epoxy resin and mechanically mixed for 5 min. The mixture was then ultrasonically treated for 30 min using the USDN-1 ultrasonic treatment unit. After the ultrasonic treatment was completed, PEPA hardener was added to the resulting mixture. After mechanical stirring for 5 minutes, the resulting mixture was placed in polymerization molds and incubated at a temperature of about 50 °C for at least 24 hours.

Measurement setup:

The specific heat was determined using the method of thermal relaxation on a commercially available Physical Properties Measurement System (PPMS, Quantum Design Inc.)

Experimental results:

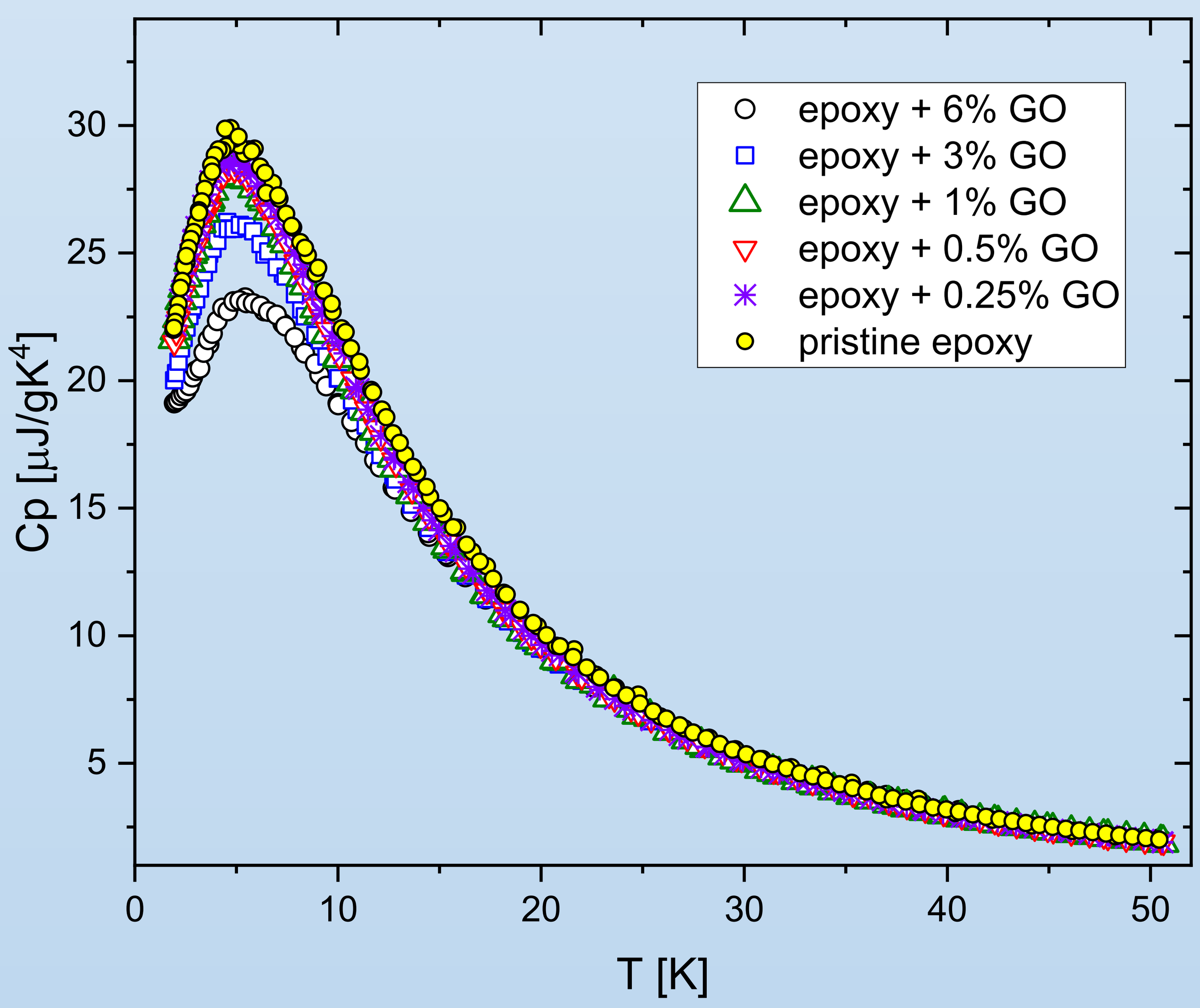

Fig. 1 Temperature dependence of the heat capacity for pristine epoxy resin and epoxy with different graphene oxide concentration.

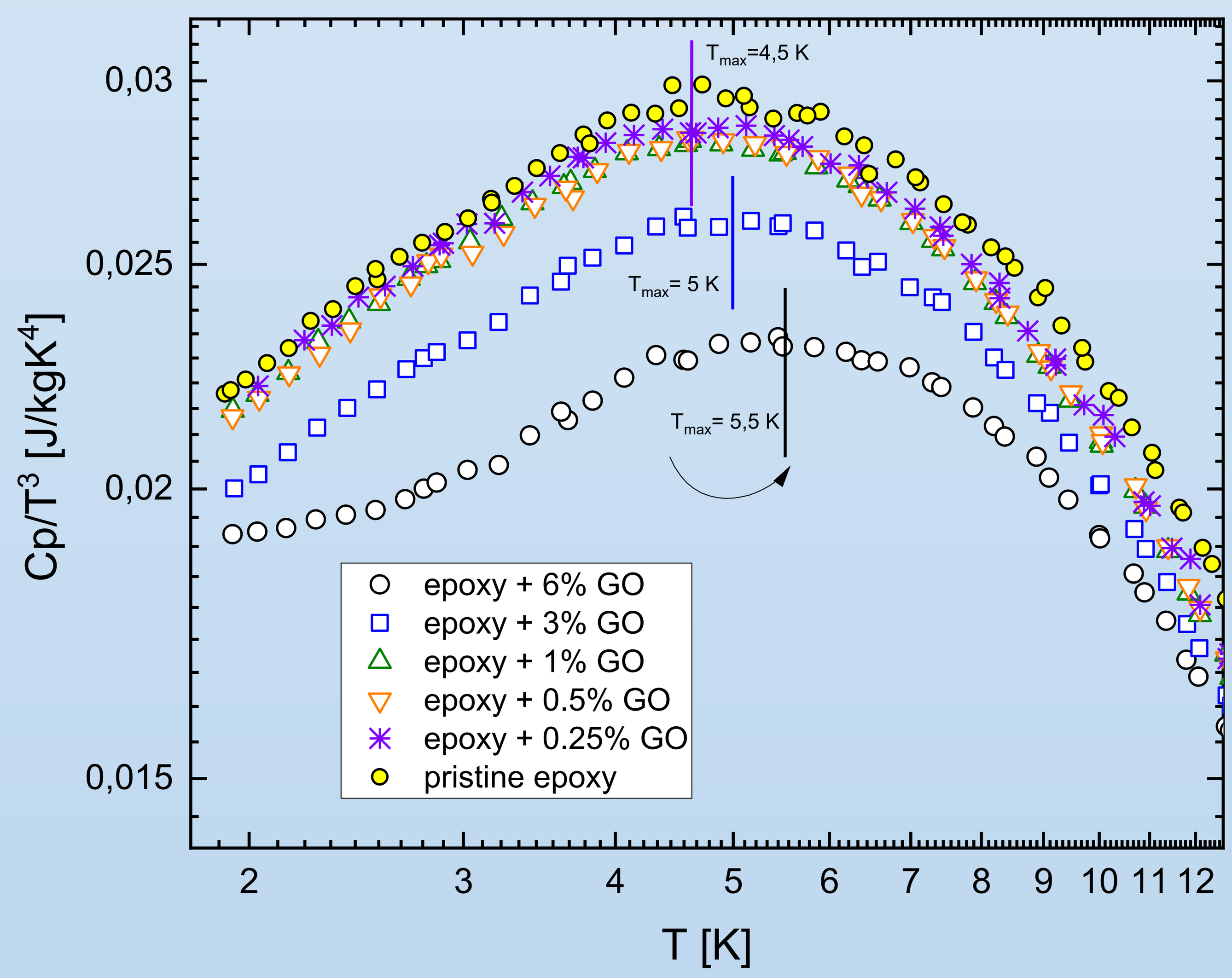

Fig. 2. Position of boson peak at low-temperature part of the heat capacity for epoxy resin with different graphene oxide concentration.

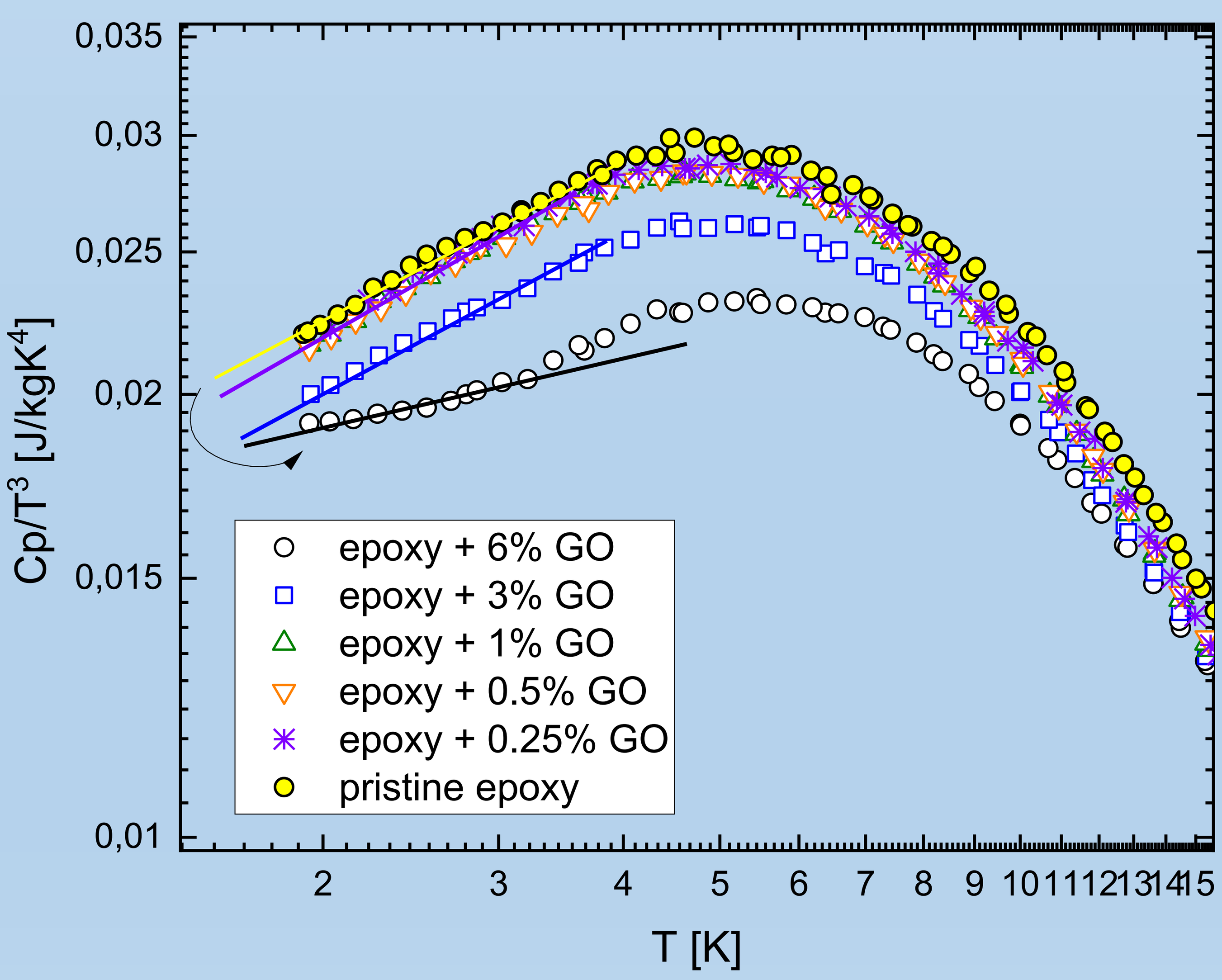

Fig. 3 Low-temperature dependence of the heat capacity for epoxy resin with different graphene oxide concentration in C/T³ coordinates.

The "boson peak" is observed in the heat capacity data for epoxy with GO impurity and pristine epoxy, the position and height of which varies with filler loading. This peak slightly decreases with increasing filler content. In addition, a shift as a function of temperature is observed. The experimental data were analyzed in comparison with literature data, where the eventual decrease of the peak was related to inherited "crystalline" characteristics of the material due to the high graphene content [2].

[2] Z. E. Nataj, Y. Xu, D. Wright, J. O. Brown, J. Garg, X. Chen, F. Kargar & A. A. Balandin, Nature Communications, 14, 3190 (2023). https://doi.org/10.1038/s41467-023-38508-3.

The main result of this research is the observation of the anomalous temperature dependence in heat capacity of composites epoxy-GO at the lowest temperatures. While the motivation for the studies was a paper by Nataj et al. in Nature Comm. *14*, 3190 (2023) claiming that one can obtain both thermal conductor or thermal insulator features in those composites, our results show that a "simple" modification in introducing graphene oxide into the epoxy matrix instead of simple graphene flakes changes the resulting behaviour into an unified one.

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