Supporting Information

Enhancing thermal transport of epoxy composite with vertically aligned graphene in-situ grown on the thermal interface

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Note S1: Experiment details of VG preparation

The quartz boat with a copper sheet was placed in the center of the heating furnace (to avoid uneven heating of the quartz boat), then the inlet valve and baffle valves at both ends were closed, leaving only the outlet valve open. Turned on the vacuum unit to make the vacuum in the quartz tube cavity reach 5×10^{-2} mbar, then turned off the vacuum unit and the outlet valve. Opened the inlet valve and passed the argon gas to make the air pressure in the quartz tube cavity reach 7×10^{-2} mbar, opened the outlet valve again and closed the inlet valve, turned on the vacuum unit to pump out the argon gas until the tube vacuum degree reaches 3×10^{-2} mbar, the process was repeated twice. Closed the vacuum unit and opened the inlet valve, through the 10 sccm (standard cubic centimeter per minute), turned on the heating furnace, set the heating program for 40 min from room temperature (25 ℃) to 700 ℃, and let the sample in the hydrogen atmosphere be fully exposed for some time. After the temperature was stabilized, 52 sccm of acetylene was introduced to maintain a 5.2 : 1 flow ratio of acetylene to hydrogen in the tube, at this time, the air pressure in the tube was stabilized at approximately 10 mbar. Turned on the PE radio frequency power supply, and gradually increased the power up to 200 W, the surface of the copper substrate began to slowly generate vertical graphene, the process lasted 120 min. After the reaction was completed, the heating furnace, acetylene gas valve, and PE radio frequency power supply were turned off simultaneously, and the continuous flow of hydrogen was maintained, so that it could be used as an etchant to modify the defects of the sample during cooling to room temperature. Finally, the hydrogen gas valve was turned off, and the baffle valves at both ends of the quartz growth furnace were opened to take out the quartz boat with the copper sheet on which had grown vertical graphene.

Note S2: Simulation details

1. Simulation of the heat transfer of VG-EP, 3DG-EP, and EP

Fig. S1 shows that the three materials are all set up as squares with a side length of 5 μm. The three squares from left to right are the VG-EP, the 3DG-EP, and the EP, the graphene thermal conducting layer is distributed in the epoxy resin layer. The thermal conductivity of the graphene heat-conducting layer is set to 500 $W \cdot m^{-1} \cdot K^{-1}$, the thermal conductivity of the epoxy resin layer is set to 0.22 $W \cdot m^{-1} \cdot K^{-1}$, the bottom power is a constant heat source of 0.01 W, and the convective heat transfer coefficient of the boundaries is set to 10 $W \cdot m^{-1} \cdot K^{-1}$.

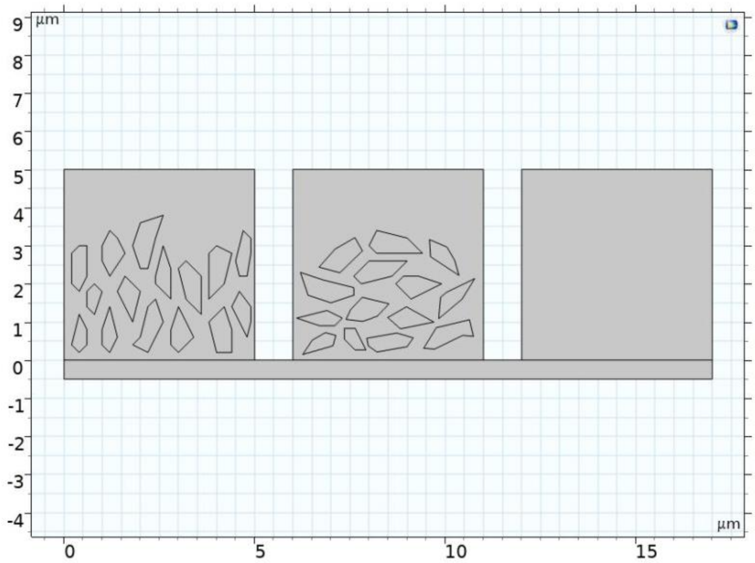

Fig. S1. Heat transfer simulation model diagram for three materials.

2. Simulation of the evaporation process of water droplet

To ensure the reliability of the model, the contact angle of the epoxy resin surface is used as a reference, considering that all three materials are mainly composed of epoxy resin. From **Fig. S2**, it can be seen that the contact angle on the left side is 70.7° and the contact angle on the right side is 69.3°, so the average value of 70° (θ_w = 1.22) is taken as the contact angle of the three films in the modeling. The dimensions of all three films are set to 5 μm in height and 40 μm in width, similar to the actual thickness as shown in **Fig. S3**. The three rectangles from left to right are the VG-EP, the 3DG-EP, and the EP. The geometric parameters of the water droplets are then set to a height of 5 μm and a radius of 7.4 μm and placed in the center of the films. The largest rectangular area is the air domain with 40 μm height and 150 μm width. The bottom edge of the films is the heat source, which is set to a constant 80 °C for the first type of boundary conditions. The initial temperatures of air, water, and all three films are 25 °C. A multiphysics field coupling heat transfer in solid and fluid, laminar flow, and phase field are used to simulate the change in volume fraction of water droplets during heating from 0 to 10 ms.

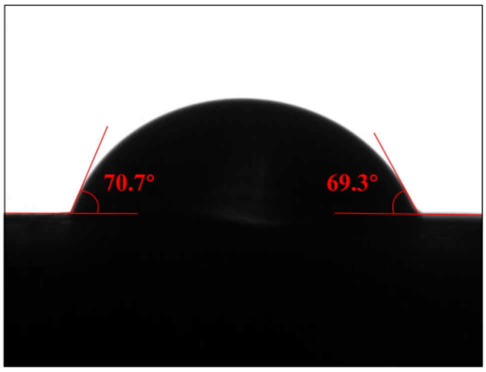

Fig. S2. Contact angle of a water droplet on an epoxy material.

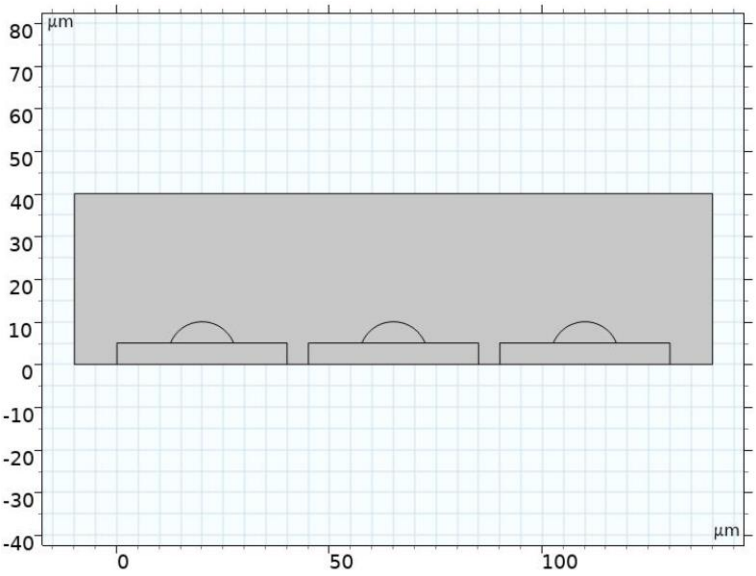

Fig. S3. Model diagram for water droplet evaporation simulation.