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M4Or1B-07: Study on cryogenic mechanical and gas-barrier properties of graphene oxide modified epoxy resin

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As a renewable and clean energy, hydrogen energy plays a crucial role in constructing the low-carbon and efficient modern energy infrastructure. Hydrogen storage is a key link in the hydrogen energy application chain. The common vessels with metal or plastic inner liners are prone to defects such as hydrogen embrittlement, collapse, and delamination. All-composite vessel (Type V vessel) is a new type of cryogenic liquid hydrogen vessel. Its composite layers serve both as a pressure-bearing and gas-barrier function. However, the resin matrix of composite materials, which tends to become brittle in cryogenic environments, experiences a decline in mechanical properties. In addition, since the polymer contains a large amount of free volume, gas molecules are easy to escape in the free volume. Therefore, the resin matrix must be modified to improve its cryogenic mechanical strength and gas barrier properties simultaneously.

In this study, KH550 silane coupling agent was used to modify graphene oxide (GO) through a one-step hydrothermal method. The surface modification of GO by KH550 silane coupling agent enhanced the interfacial bonding between GO and the resin matrix. In this case, agglomeration of particles occurred less often at cryogenic temperatures. The product was named k-GO (functionalized graphene oxide). The modification effect was analyzed using infrared spectroscopy, X-ray diffraction, and scanning electron microscopy. The results of structural analysis revealed changes in the functional groups, interlayer spacing and surface morphology of the modified GO. The outcome confirms the successful grafting of KH550. Then, the k-GO/epoxy resin composite was prepared by blending method. The mechanical and gas barrier properties of composites with different k-GO contents were investigated. The results demonstrated that when k-GO content was 0.1 wt.%, the k-GO/epoxy resin composite achieved its maximum mechanical strength. Compared to pure epoxy resin, the tensile strength increased by 7.7% and 16.3% at room and cryogenic temperature, respectively. When k-GO content was 0.2 wt.%, the k-GO/epoxy resin composite achieved its maximum gas-barrier properties. The gas permeability coefficient decreased by 15.8%, compared to pure epoxy resin. Although this method enhanced both mechanical strength and gas barrier properties, the peak values occurred at different k-GO concentrations. This prevented the simultaneous improvement of the two attributes required for the Type V vessels. Therefore, to meet the requirements of all-composite vessels under cryogenic conditions, k-GO and PEG (polyethylene glycol) were used to modify the resin, which enhanced the cryogenic mechanical strength while maintaining its gas barrier properties. The results gave that when the incorporation of k-GO was 0.2 wt.% and PEG was 5 wt.%, the cryogenic tensile strength of the composites was increased from 79.73 MPa to 98.55 MPa, which was increased by 28.8%, compared with pure epoxy resin. At the same time, the gas permeability coefficient only increased by 0.3% compared to the case without PEG.

In this paper, the epoxy resins with high cryogenic strength and high gas barrier performance are prepared by the cooperative modification of k-GO and PEG. It is expected to be used in the manufacturing of Type V all-composite vessels, and promote the application of liquid hydrogen energy supply technology.

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