Abstract

In the pursuit of economic development and the establishment of a sustainable future, there exists a pressing need to transition from conventional energy sources to renewable alternatives. This imperative underscores the critical role of materials science, particularly in the realm of sustainable energy solutions. This present dissertation is centred on the utilization of multiscale modeling coupled with experimental validation to investigate composite materials designed for energy applications. Through the exploration of these advanced materials, we aim to address critical energy challenges and contribute to economic progress. Initially, a multiscale model is developed to ascertain the thermophysical properties of nanofiller-enhanced thermoset and thermoplastic polymer composites. These materials hold significant relevance in diverse application, including the use of epoxy composites for elongated wind turbine blades, polypropylene composites in separator films for lithium-ion batteries and polylactic acid composites for supercapacitor electrodes. Specifically, the thermoset polymer under examination, comprises an epoxy resin system containing bisphenol-A diglyceryl ether (DGEBA) as an epoxy monomer and dicyandiamide (DICY) and diethylene triamine (DETA) as cross-linking agents. To establish the interatomic coarse-grained (CG) potential for the mesoscopic model and achieve a density match with material studied via atomic simulations, the iterative Boltzmann inversion method is employed. The derived CG potentials effectively reproduces various thermophysical properties. Furthermore, nanocomposites consisting of the aforementioned epoxy reinforced with graphene nanofillers are simulated at the mesoscopic level and corroborated against continuum approaches. Our findings indicate that a moderate inclusion of nanofillers (up to 2 wt.%) enhances the elastic modulus and thermal conductivity of the epoxy resin concurrently reducing the Poisson's ratio.

For thermoplastic polymers, this dissertation introduces a multiscale modeling framework that exploits mesoscopic models to predict the thermal and mechanical properties of nanocomposites based on their molecular structure. Mesoscopic models consisting of polypropylene (PP), polylactic acid (PLA), and graphene-based nanofillers (graphene (Gr), graphene oxide (GO), and reduced graphene oxide (rGO)) are examined. The newly developed mesoscopic models for the PP/Gr and PLA/Gr nanocomposites furnish mechanistic insights into the mechanical and thermal properties at the matrix–filler interface, which can subsequently be utilized to enhance the prediction accuracy of traditional continuum simulations by calibrating the mechanical and thermal properties of said interface. Upon validated through a dedicated experimental campaign, this multiscale model underscores that the addition of nanofillers (up to 2 wt.%) augments the elastic modulus and thermal conductivity of the neat PP and PLA, while slightly decreasing the Poisson's ratio. Among the

various combinations investigated, the PP/Gr and PLA/Gr nanocomposite exhibit superior mechanical properties, whereas PP/rGO and PLA/Gr demonstrates enhanced thermal conductivity. Notably, we present the first CG model for DGEBA-DICY-DETA/graphene, PP/Gr, PP/GO, PP/rGO, PLA/Gr, PLA/GO, and PLA/rGO, thereby enabling the design of polymer composites with tailored properties for a broad range of applications extending beyond the energy sector.

In addition to the work on polymer composites, this dissertation also investigates solid-solid phase-change materials (SS-PCMs) for thermal energy storage, which offer several advantages over traditional solid-liquid phase-change materials (SL-PCMs), including high thermal stability, reduced risk of leakage, ease of handling, and enhanced durability. Employing a combination of computational modeling and experimental characterization, we investigate the thermal properties and phase change behavior of SS-PCMs. In the start, we employed classical molecular dynamics (MD) simulations to investigate the thermophysical properties of four main polyalcohols (Pentaerythritol (PE), Pentaglycerine (PG), Neopentyl glycol (NPG), and (tris(hydroxylmethlyl)aminomethane (TAM)) using the GROMOS force field. Results reveal a linear decrease in the densities of polyalcohols across both solid phases with increasing temperature, accompanied by a corresponding increase in their specific volumes. Notably, a sharp increase in specific volume is observed at the solid-solid phase transition. The evaluation of latent heat of storage of polyalcohols in MD simulation is based on the change in potential energy between the two phases at the phase transition temperature. However, polyalcohols exhibit low thermal conductivity, posing limitation to their application. To elucidate the thermal conductivity variations associated with changes in the crystalline structure, non-equilibrium molecular dynamics (NEMD) simulations were conducted. The results indicate that the thermal conductivity decreases with increasing temperature across both phases of polyalcohols. Notably, a pronounced drop in thermal conductivity is observed at the phase transition temperature, underscoring the influence of structural factors, particularly the hydrogen bonds (HBs) network, on thermal conductivity. Further analysis of the quantity and energy of HBs provides deeper insights into the micromechanics of polyalcohols. Upon solid-solid phase transition, the strong intermolecular HB in the α phase breaks and forms weaker HBs in the γ phase.

In the realm of experimental characterization, Differential scanning calorimetry (DSC) measurements were conducted to investigate the thermal behavior of PE and PG materials. This entailed determining their solid-solid phase transition temperature alongside their energy storage density. Subsequently, multiscans (three heating and two cooling scans) were executed with the solid-solid phase change region for the both the materials in powder form. Post-testing, mass loss measurements were conducted to assess sample integrity, revealing mass loss in PE while PG remained stable. To address the mass loss in PE powder, a dedicated closed system was designed, minimizing the impact on the solid-solid phase transition temperature while increasing its latent heat of storage. Tablets of PE and PG were fabricated from their respective powder using cold pressing techniques and subjected to DSC analysis. However, a notable reduction in the latent heat was observed in the tablets compared to their powder counterparts. Similarly, PE tablet exhibited mass loss. Furthermore, the subcooling degree, increases from powder to tablets in both materials. For the SS-PCM composites, PG was selected as base PCM, and Multiwalled carbon nanotubes (MWCNTs) were incorporated in various weight

percentages (1, 5, and 10 wt.%) through manual mixing in powder form followed by cold pressing to fabricate tablets. Analysis of DSC curves indicated a decrease in latent heat with increasing weight percentage of MWCNT nanofillers. Additionally, the subcooling effect exhibited a reduction of approximately 2.2 °C for the 10 wt.% MWCNT composite compared to the neat PG tablet. Preliminary thermal conductivity measurements of neat PG tablets revealed a slight decrease from 0.318 W/mK to 0.310 W/mK as the temperature increases from 25 °C to 75 °C. However, a significant decrease in thermal conductivity was observed upon exceeding the solid-solid phase transition temperature, reaching 0.20 W/mK at 118 °C, indicating a change in the material's heat transport properties. Further exploration of the thermal conductivity of the composites is warranted to ascertain the potential impact of MWCNT loading on this key property for practical applications.

The findings of this study hold significant progress in both polymer nanocomposite and solid-solid phase change materials, providing more reliable and efficient solutions for various energy-related applications. These include epoxy composites for wind turbine blades, , polypropylene composites in separator films for lithium-ion batteries, polylactic acid composites for supercapacitor electrodes, and solid-solid phase change materials for building heating and cooling systems, solar energy storage, and waste heat recovery. Moreover, this research establish a foundation for further studies on the multiscale modeling and experimental characterization of polymeric nanocomposites and solid-solid phase change materials reinforced with a broader range of carbon based nanofillers, such as carbon nanotubes in polymer nanocomposites and graphene-based nanofillers into solid-solid phase change materials.