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Title of Doctoral Thesis: Influence of halogen elements in organic salts on n-type doping of CNT yarn for thermoelectric applications

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Thermoelectricity is a direct energy conversion process that involves converting heat into electricity. The performance of thermoelectric materials is typically determined by three key factors: high Seebeck coefficient (α), high electrical conductivity (σ), and low thermal conductivity (κ). Recently, significant efforts have been focused on enhancing thermoelectric performance using promising materials. Among of the promising material that has gained considerable attention is carbon nanotubes (CNT) due to their exceptional properties, including high electrical conductivity, mechanical robustness, and flexibility. However, there is existing challenges when utilizing CNTs as thermoelectric materials. For instance, it is difficult to produce n-type CNTs due the unintentional p-type doping to semiconducting CNTs under ambient atmosphere tends to override the n-type doping. One method to produce n-type CNT with long term stability is by using the charge stabilization doping process. Recently, organic salts have gained popularity as charge stabilization dopants.

The ability of organic salts as dopants makes them a good candidate for enhancing TE performance, especially in improving n-type CNTs stability. Furthermore, doping n-type CNTs with organic salts, particularly with different halogen atoms, provides an opportunity to explore the interaction mechanism between CNTs and dopants. Moreover, exploring the impact of organic salts with different alkyl chain lengths as dopants for weak n-type CNTs opens up new possibilities for TE material engineering.

By probing these interactions, we aim to uncover the doping mechanisms that optimize the TE properties of CNTs. Overall, this research seeks to advance understanding of doping against CNTs and its impact on TE performance. By leveraging weak n-type CNTs and exploring the effects of organic salt dopants, this work aims to develop novel strategies to enhance the efficiency and stability of CNT-based TE devices, ultimately contributing to the advancement of sustainable energy technologies and energy-efficient electronic devices.

By using weakly n-type CNTs is a foundational approach to initiate the n-type doping process. Through deliberate annealing treatments, CNTs produced with Emulgen as the surfactant exhibit distinct characteristics by applying the variations of the annealing temperature. In the presence of the surfactant, subjecting CNT yarns to annealing at 200 °C and 300 °C induced an n-type behavior. Conversely, annealing at different temperature ranges generated p-type behavior. This shift in behavior is attributed to the annealing process occurring at 400-500 °C, which effectively eliminated the surfactant and escalated defects within the CNTs, consequently leading to a p-type behavior. Notably, the most favorable TE properties were observed in the n-type CNT yarns annealed at 300 °C and the p-type CNT yarns annealed at 500 °C.

To obtain the stability of n-type CNTs, doping using organic salts with different halogen types was applied. The results show that incorporating organic salts effectively increases σ and reduces κ of CNT threads. Doping with tetrabutylammonium iodide (TBAI) resulted in a much higher figure of merit (ZT) than pure CNT yarn, with about three times the increase. The study also provided insights into the doping mechanism, showing that the redox potential of the halogen element influenced the amount of excess large organic cations, which stabilized negative charges (free electrons) in the CNTs. This

further supported the effectiveness of charge stabilization using large organic cations and iodine for efficient and stable n-type doping of CNTs.

Continuing the exploration, additional investigations involved doping with organic salts featuring varying alkyl chain lengths. Contrary to expectations, the experimental findings indicate that doping efficiency is not dependent on alkyl chain lengths; instead, it is governed by the redox potential of the halogen element. However, the PF's stability exhibits sensitivity to the alkyl chain length.