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Electronic transport properties in donor–acceptor single-walled carbon nanotubes filled with caffeine molecules.

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Abstract

In light of the successful use of Caffeine in perovskite solar cells, a novel nano-hybrid system comprised of caffeine packed in a metallic single-walled carbon nanotube is proposed as a viable candidate for organic solar cells. Investigations have been made on the stability of this hybrid system that has a single Cf molecule inside of NT8. In particular, the ideal diameter is found. The investigation's findings show evidence of a charge transfer between the Cf molecules and the NT8 nanotube, which also attests to the stability of the structure. Electronic and optical computations are used to determine this Charge Transfer's location in the Cf @NT8 hybrid system as well as its direction. The electronic computations and transmission spectra led us to the conclusion that a CT link from caffeine to carbon nanotube is implied by the increased Fermi level position of carbon nanotube after encapsulation.

Keywords: SWCNT, Caffeine, organic solar

1 Introduction

In the last five years, a significant challenge in nanotechnology has been constructing durable and high-quality devices at the atomic level. Organic solar cells (OSCs) that directly convert solar energy into electricity offer a practical and sustainable solution to address the increasing global energy demand. These OSCs have gained considerable attention recently due to their straightforward fabrication methods [1, 2]. Their development allows for the expansion of renewable energy sources, contributing to approximately 16% of the world’s total primary energy consumption. To address market demands, these devices need to prioritize affordability, extended longevity, lightweight design, semi-transparency, flexibility, and achieving high power conversion efficiency (PCE) [3–7]. Consequently, the primary concerns for OSCs [1, 8, 9] are achieving both high power conversion efficiency (PCE) and long-term stability. Despite their promise, organic solar cells currently encounter multiple hurdles that restrict their effectiveness and feasibility for commercial use. These challenges encompass issues such as low efficiency, vulnerability to degradation, and a limited operational lifespan, particularly when subjected to oxygen, moisture, and light exposure [10, 11]. Currently, numerous global organizations are dedicated to improving the optical and electrical characteristics of thin films in anticipation of the next generation of organic solar cells [12, 13]. In this context, existing literature highlights the practice of encapsulating various guest substances, which may include organic molecules, within single-walled carbon nanotubes (SWNTs) [14–16]. This approach is recognized as one of the most effective techniques for merging the physical attributes of two systems without causing mechanical or chemical damage to either of them [17, 18]. In this paper, we introduce a particular organic compound, 1,3,7-trimethylxanthine, commonly recognized as caffeine (C_f), into the cavity of a metallic single-walled carbon nanotubes of the chirality (8,8) ($C_f@NT8$). This configuration holds promise as a photoactive substance for OSCs. From this standpoint, this method represents one of the optimal approaches to merge the physical properties of these two systems, all without causing any mechanical or chemical damage. It has the potential to support OSCs in achieving heightened efficiency and long-term stability in the future.

our research was focused on single caffeine molecule enclosed within NT8, we delve into the optoelectronic properties of our systems, including pure NT8, $C_f@NT8$, and C_f2NT8 , employing density functional theory (DFT). Subsequently, the final section offers a concise summary of the outcomes.

2 Methods and model

We conducted an estimation of the density of states (DOS) and optical properties for pristine caffeine and caffeine enclosed within an NT8 tube through first-principles calculations based on density functional theory [19, 20]. These calculations employed a plane-wave basis set and wave projector, utilizing the Vienna Ab initio Simulation (VASP) software [21]. The DFT calculations were carried out at the generalized approximation GGA level, employing HSE06 and B3LYP level of theory [4], along

with the 6-31G(d) basis set to determine the geometries and optoelectronic characteristics of caffeine (C_f). A kinetic energy cutoff of 520 eV was set for the wave function expansion. The GGA effectively describes chemical bonding and electron transport in hybrid systems. However, for regions with low electron density, the GGA functional inaccurately represents dispersion or van der Waals (vdW) forces. To address this issue, we applied Grimme’s method [22] to make the necessary corrections for the vdW interface between C_f and SWNTs (i.e., GGA plus vdW).

For our calculations, we considered two distinct hybrid SWCNT models as illustrated in Figure 1:

The $C_f@NT8$ hybrid model, where a single C_f molecule is positioned in front of the tube. The $C_f2@NT8$ hybrid model, where a single caffeine molecule is horizontally inserted into the tube.

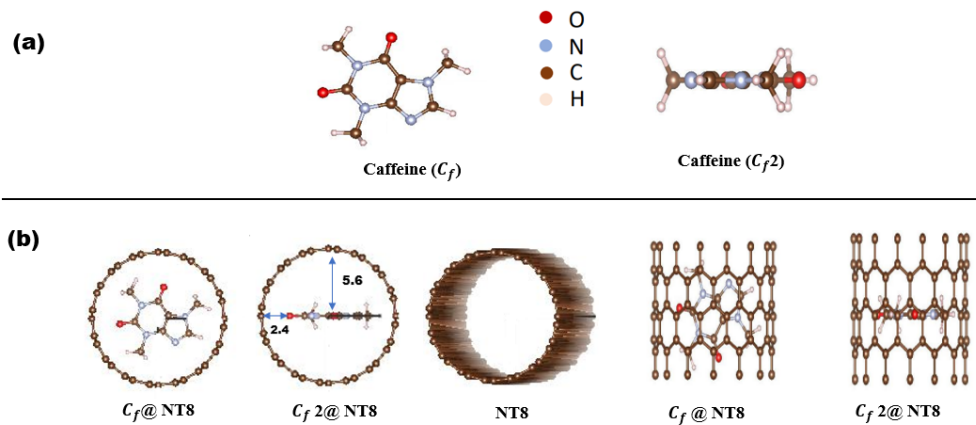


Fig. 1: Unveiling the Architecture of, (a) C_f Molecule. (b) hybrid systems ($C_f@NT8$, $C_f2@NT8$)

3 Results and discussion

Our primary focus was on the analysis of characteristics such as the density of states, absorption coefficient, and dielectric functions. These computations were carried out through the utilization of the GGA xc functional within the framework of DFT, employing the VASP software as previously noted. The objective of this study was to enhance the performance and thermal resilience of organic solar cells (OSCs) by incorporating C_f and NT8 into their active layer.

In Figure 2, the total density of states for the hybrid systems ($C_f@NT8$ and $C_f2@NT8$) exhibit significant distinctions when compared to the pristine NT8 nanotube. It is evident that the bandgap of NT8 is diminished upon encapsulation, resulting in the complete disappearance of the bandgap in the $C_f@NT8$ hybrid,

forming a new localized energy level. This observation indicates that the encapsulation effect enhances the conductivity of NT8, leading to NT8's acquisition of metallic properties when encapsulated.

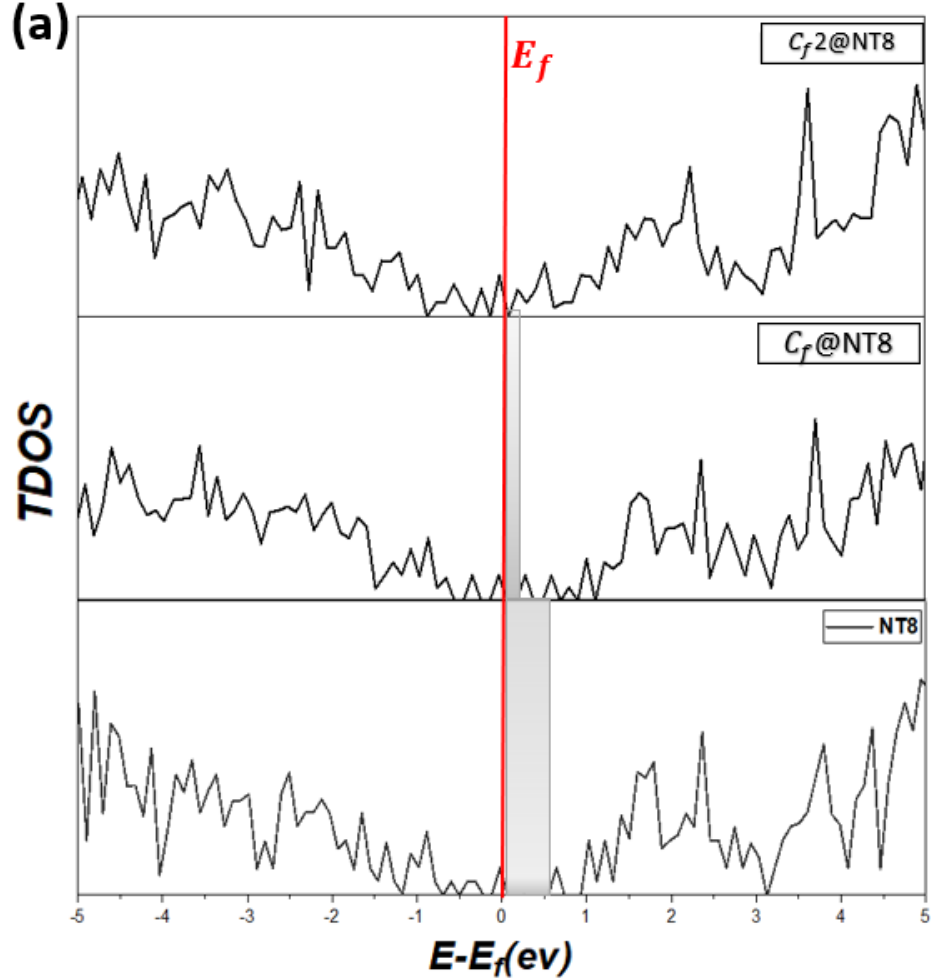


Fig. 2: The overall density of states (TDOS) of the pristine NT8 and the hybrid systems ($C_f@NT8$, $C_f2@NT8$)

Furthermore, absorption coefficient, as shown in Figure 3-(a), is a crucial factor that profoundly affects the optical properties of devices, such as solar cell efficiency. The relatively short bandgap of NT8 results in these hybrid nanosystems displaying strong optical absorption characteristics, not only in the ultraviolet range but also in the near-infrared part of the spectrum. This enhanced absorption is attributed to the

smaller bandgap of the hybrid systems when compared to that of the pristine NT8, as indicated in Figure 3-(a). Subsequently, we utilized the electron energy levels and their corresponding wave functions obtained from the electronic structure simulations to calculate the dielectric functions for both the real and imaginary components of the pristine NT8 tube and the C_f encapsulated systems ($C_f@NT8$ and $C_f2@NT8$). In terms of electronic polarizability, it's important to note that the pure NT8 tube and the C_f encapsulated systems exhibit consistent behavior across all energy regions. In the infrared spectrum, specifically, the real component of the dielectric function shows a maximum peak intensity of approximately 15, as illustrated in Figure 3-(b). This indicates the potential use of the C_f encapsulated systems as effective photonic sensors in this particular spectral range. These systems exhibit prominent peaks in the imaginary part of the dielectric function at photon energies of around 1.2 eV, as depicted in Figure 3-(c).

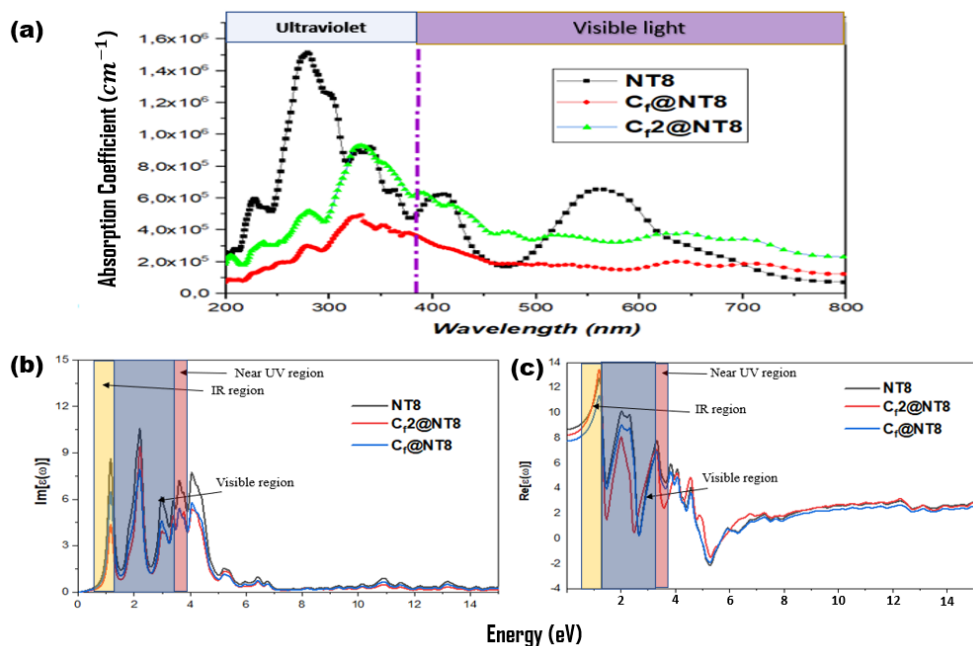


Fig. 3: (a) Absorption coefficient of the pure NT8 tube, $C_f@NT8$ and $C_f2@NT8$ hybrid nano-systems, (b) imaginary and real dielectric functions for the pure NT8 tube, $C_f@NT8$ and $C_f2@NT8$ hybrid systems

4 Conclusion

In this investigation, we introduced three computational approaches to scrutinize the encapsulation of C_f molecules within the NT8 nanotube cavity. These methods were

applied to examine both the structural features and optoelectronic characteristics of the system. Molecular mechanics and density functional theory were employed to elucidate the dynamic models and transmission of the hybrid systems. The primary goal was to investigate the optoelectronic behavior Initially of the pristine NT8 then for the hybrid systems ($C_f@NT8$ and $C_f2@NT17$) using the VASP code. The outcomes yielded promising results, suggesting that C_f molecules and NT8 nanotubes exhibit favorable attributes, rendering them viable candidates for incorporation in oSC devices. The analyses produced notable results, revealing a narrowed band gap of approximately 0.15 eV following the encapsulation process.

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