



Research paper

The phase stability of terephthalic acid under high pressure



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ABSTRACT

Terephthalic acid has been investigated by Raman spectroscopy up to 15 GPa. According to *ab initio* calculations, it can be speculated that both of π - π stacking interactions between molecules and the symmetry of hydrogen bonds are enhanced with gradually increasing pressure. Furthermore, we use the Hirshfeld surface to map the π - π stacking interaction in the TPA molecule at high pressure. The Raman spectra and *ab initio* calculation results indicate that the phase stability of TPA is related to the one-dimensional hydrogen bond network and inter-chain aromatic π - π stacking interaction.

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1. Introduction

Non-covalent interactions are much weaker than covalent interaction in strength (typically on the order of 1–40 kcal/mol [1]) but play a vital role in the design of new functional materials with desirable physical and chemical properties [2–4]. And it can be divided into different categories, such as hydrogen bonds, π - π stacking interaction, halogen bonds, hydrophobic effects and van der Waals forces. Their role has been very well recognized in different fields related to biological [5,6], pharmacological [7], chemical [8,9], physical [10,11] and materials sciences [12]. Among those interactions, it has been found that hydrogen bonds with high orientation properties are very effective in supermolecular components. Besides hydrogen bonds, the π - π stacking interactions occupy prominent position amongst those non-covalent interactions and their importance in the area of chemistry and biology is very well recognized. These two types of interactions determine the structure and dynamics of nucleic acids [13–15]. Besides the significance of the two non-covalent interactions played in biomolecules, the cooperative and competitive interactions between hydrogen bonds and π - π stacking interactions are still a subject of debate.

High pressure investigations on the molecular crystals with non-covalent interactions have been conducted to study the response of physical and chemical properties to the external compression [16,17]. Pressure is an effective thermodynamic parameter can significantly alter the inter-atomic distances and thus the

nature of intermolecular interactions, chemical bonding, molecular conformation, crystal structures, and stabilities of solids as a result of decreasing in volume. Pressure-induced phase transitions are a common phenomenon in high pressure science. Several pressure-induced phase transitions have been discovered in benzene and water, and liquid-solid phase transition has been measured at 0.3 GPa [18] and 0.2 GPa [19] for benzene and water, respectively. The intermolecular interactions in benzene are mainly π - π stacking and are hydrogen bonds in water. When the π - π stacking and hydrogen bonding intermolecular interactions are both present in the molecular crystals, what are the effects of these intermolecular interactions on the phase stability under external compression? Aromatic carboxylic acid molecules are typically molecules that have π - π stacking and hydrogen bonding interactions. Such as benzoic acid, terephthalic acid, trimellitic acid are aromatic carboxylic acids. Benzoic acid [20] is a dimer structure, and does not occur the phase transition under high pressure. Trimellitic acid [21] is two-dimensional structure, and may occur the phase change under high pressure. Terephthalic acid is one-dimensional infinite chain structure [22], so far its high pressure properties have not been studied. Exploring the physical properties of TPA under high pressure, not only can understand its structural changes, but also can supplement the aromatic carboxylic acid system research.

Terephthalic acid (1,4-benzenedicarboxylic acid) (TPA) has been used mainly as a raw material of polyester fiber, and it also has been applied for various uses such as non-fiber field, PET-bottle, PET-film and engineering plastics and as poultry feed additives [23,24]. Due to its extensive applications, large-scale production and its chemical characteristics, this refractory organic material and its associated class of organics have become ubiquitous

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environmental pollutants, so far some studies on the destruction of TPA have been by biological means, using microorganisms [25–28]. Detailed studies have been carried out on TPA using a number of experimental methods such as neutron scattering [29], infrared spectroscopic [30] and Raman scattering methods. Raman spectroscopy is efficient and accurate tool for studying the changes of terephthalic acid under high pressure [31].

In this study, we report comprehensive investigations of TPA using Raman spectroscopy up to ~15 GPa. Meanwhile, the lattice parameters of terephthalic acid were analyzed by theoretical calculation under high pressure. This work shed light on the effect of hydrogen bonding and π - π interactions on the stability of the crystal.

2. Experiment

Samples of 1,4-benzenecarboxylic acid were obtained from Sigma Aldrich A.R and were well-preserved before utilization. Apparatus for producing ultrahigh-pressure were Mao-Bell diamond anvil cell (DAC) with 500 μm diameter of diamond anvil. A sheet of T301 steel, constituting a sealing device for generating super high pressure, with thickness of 250 μm were used as gasket material. Having compressed the gasket to 40 μm thickness, a super high pressure cell with a diameter of 200 μm was created by drilling compressed gasket with a shaped tetragonous Tungsten carbide needle. Owing to its Linear Correlation of the pressure-induced shifts of R_1 lines of Ruby, Ruby powder whose diameters are roughly 10 μm were used as calibration standard to precisely monitor pressure in the cavity in real-time.

High pressure Raman spectra were recorded by confocal laser microscopy Raman spectrometer of Reni Shaw in via. Incentive wavelength light of Spectra Physics 160 M laser was 514.5 nm and the output power of the laser was 8 mW connected to LEICA DMLM microscope system with 50 times amplification objective lens. Laser beam can accurately focus on the samples in the high pressure chamber and the integration time of spectra was 20 s. All the experiments were carried out at room temperature.

3. First-principals DFT calculations

Ab initio plan-wave pseudopotential density functional (DFT) method performed in the CASTEP code [32] has been used for the investigation. The initial structure model was constructed based on reported experimental data [22]. The optimization is not finished until the forces on the atoms are less than 0.01 eV/Å and all the stress components are less than 0.02 GPa. The tolerance in the self-consistent field (SCF) calculation is 5×10^{-7} eV/atom. The norm-conserving pseudopotential is implemented to describe interactions

between nuclear core and valent electrons. The scheme of Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation (GGA) displays exchange and correlation effects in details. Convergence tests give a $2\pi \times 0.03 \text{ \AA}^{-1}$ grid spacing and 900 eV energy cutoff. With such a choice the error bars of total energies are about 0.2 meV per atom. The theoretical calculation is not extended to the pressure higher than 15 GPa because of not knowing the detailed crystal structures of the high pressure phases.

4. Results and discussion

4.1. Raman spectrum of terephthalic acid

At ambient conditions, the TPA crystallizes in the triclinic system with group P-1. The crystal structure is characterized by the existence of the infinite chains formed through double hydrogen bonding. Crystal structure and hydrogen bond networks of TPA are shown in Fig. 1. The TPA molecule belongs to C_i point group symmetry, consists of 18 atoms, and it has 48 normal vibrational modes. The Raman spectrum of TPA at ambient condition is shown in Fig. 2. The Raman spectrum of TPA is consistent with that reported in the previous article [33]. The strong Raman scattering lines are assigned to C–H bending, C–O bending and C=O stretching vibrations at 824, 1124, and 1624 cm^{-1} respectively. And more detailed Raman peak vibrational assignments [34] are listed in Table 1.

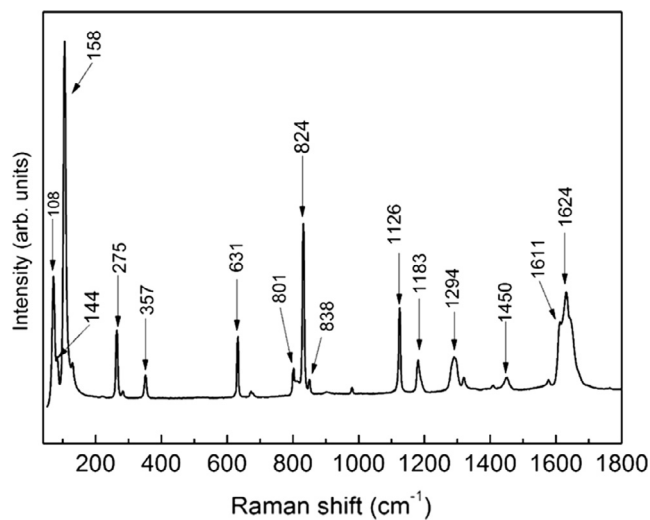


Fig. 2. Raman spectra of TPA at ambient conditions.

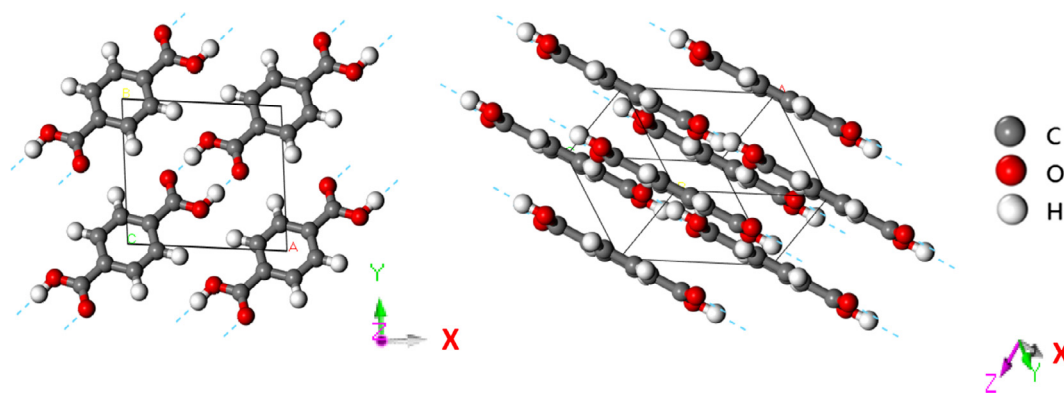


Fig. 1. Crystal structure and hydrogen bonded networks of TPA in cell [22]. (a) The a-b plane; (b) the b-c plane (orientation standard, C along Z, B in YZ plane).

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