

## REGULAR ARTICLE

# Theoretical Investigation on Nonlinear Optical Properties of Novel O-carborane Derives

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**Abstract:** A calculation investigation using DFT/TDDFT methods on a series of o-carborane derives 1-6 has been performed on the molecular structures, lower transition energy, along with the nonlinear optical responses. Our theoretical calculations show that among the components, value of  $\beta_y$  contributes the most to  $\beta_{tot}$  for all these studied compounds which indicates that the main CT within the molecular along the y-axis direction. Moreover, the static first hyperpolarizability ( $\beta_{tot}$ ) values as well as the ground state dipole moment ( $\mu_g$ ) gradually increase with increasing donating ability of group. Further, the TDDFT studies suggest that the excited transition energy experiences a decline trend and a redshift occurs on the absorption spectrum. Our studies demonstrate that compound 6 can be used as excellent 2D second-order NLO materials from the standpoint of large  $\beta_{tot}$ , large  $\mu_g$  values and low excited energy.

**AMS subject classifications:** 65D18, 74E40, 78M50, 82D99

**Keywords:** Nonlinear optics properties, O-carborane clusters, DFT/TDDFT, First hyperpolarizability

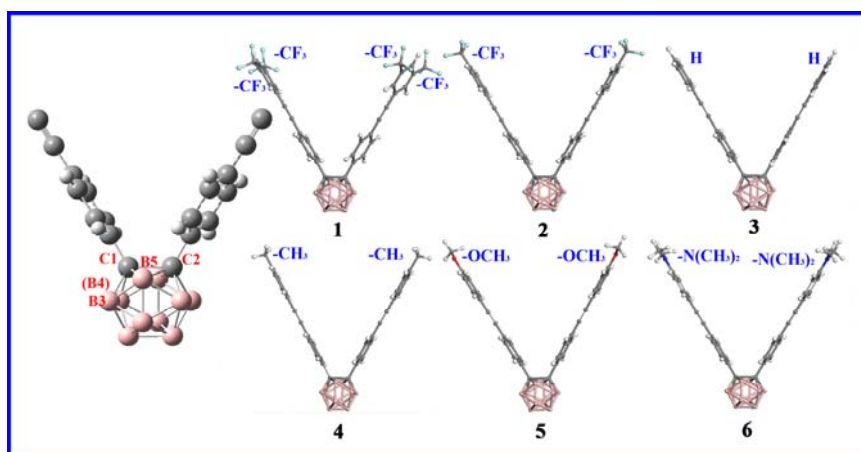
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In the past few decades, the searches for luminescent materials were continuously expanding, and the nonlinear optical (NLO) materials have attracted significant interests of experimental and theoretical chemists due to their large nonlinear optical coefficient, fast nonlinear optical response times and relatively low cost [1-10]. Numerical investigations have been carried out to develop the nonlinear optical compounds, especially for the organic or

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metallorganics-based materials [6-7, 10]. A number of organic compounds with relatively enhanced second-order responses have been synthesized or uncovered [1, 9-12]. Recently, Martín group has proved the usefulness of tetrathiafulvalene (TTF) and  $\delta$ -extended TTFs as NLO-phores and different TTF-based push-pull chromophores; moreover, Elangovan has reported the synthesis of bis-(4-methoxyphenyl) hetero-aryl-amino donor-based chromophores and their electrooptic activities [13, 14]. It is well known that typical second-order NLO is consisted by two crucial factors: one is a large polarizable  $\pi$ -conjugated unit, and the other is the conjoint donor or acceptor groups [8, 9]. The first hyperpolarizability ( $\beta$ ) is the most important parameter to demonstrate the performance of the NLO materials which can be increased by introducing the multiple donor (D) and/or acceptor (A) groups [6, 8-9,13-17].



**Figure 1:** Optimized structures of the investigated compounds 1-6 calculated by B3LYP/6-31+G (d,p)

O-Carborane ( $C_2B_{10}H_{12}$ ) clusters exhibit remarkable thermal stability, and have been found to be one extremely qualified candidate for studying the nonlinear optical characters [17-22]. The carbon and boron atoms of the cage are shown in **Fig.1**, as it demonstrated that these carboranes have icosahedral geometry, in which the carbon and boron atoms are tricoordinate. This structural feature can contribute to the stability of such molecules and their derivatives. In 2010, a family of 2D molecules centered with o-Carborane was synthesized by Chujo et al [22]. In this system, the electron-donating or the electron-withdrawing groups are attached to the o-carborane cage via arylacetylenes  $\pi$ -conjugation, which is considered to have strong electron-withdrawing property. On the one hand, those emission properties that can be effectively justified by changing the electronic-donating and electronic-withdrawing substitutions, on the other, the intramolecular charge transfer (ICT) from  $\pi$ -conjugated units to o-carborane should play the other key role [14,21-24].