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Microwave-assisted synthesis, third-order nonlinear optical properties, voltammetry cyclic and theoretical calculations of organotin compounds bearing *push*—*pull* Schiff bases



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ABSTRACT

Here we report a green one-pot microwave-assisted synthesis of four *push-pull* organotin complexes derived from Schiff bases in good yields, which provides advancement over conventional method as it is simple, cost-effective and reproducible. All compounds were characterized by UV–Vis, TOF-HRMS, TGA-DTA, cyclic voltammetry, theoretical studies and compound **1** by X-ray single-crystal diffraction. Third-order nonlinear susceptibility $\chi^{(3)}$ of complexes **1**–**4**; ((*E*)-N'-(4-(diethylamino)-2-hydroxysalicylidine) nitrobenzohydrazidatediphenyl-tin (IV) **(1)**, (*E*)-N'-(4-(diethylamino)-2-hydroxysalicylidine) nitrobenzohydrazidate di *n*-butyl-tin (IV) **(2)**, ((*E*)-N'-(4-(methoxy)-2-hydroxysalicylidine) nitrobenzohydrazidate di *n*-butyl-tin (IV) **(3)**, (*E*)-N'-(4-(methoxy)-2-hydroxysalicylidine) nitrobenzohydrazidate di *n*-butyl-tin (IV) **(4)**) were measured via Maker-Fringes technique. In particular, compounds **1** and **2** display larger third-order nonlinearities than their methoxilated counterparts.

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1. Introduction

The development of new advanced materials with nonlinear optical (NLO) properties have garnered much interest in recent years. In particular, great efforts have been focused to design chromophores with large second and third-order NLO properties for their potential use in different photonic applications, including data storage, telecommunications, all-optical switching, optical imagine processing, optical limiting devices and generation of new frequencies [1]. For the design of NLO chromophores is required a strong π -electron donor (D), π -conjugated electronic bridge, and a strong π -electron acceptor (A). Certain combinations and amounts

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of such components allow the observation of nonlinear optical phenomena [2]. By their side, organotin compounds are of particular interest due to their accessible structural design and facile synthesis [3], which applications include organic light-emitting diodes [4], fluorescent bioimaging [5], antitumor agents [6] and optical nonlinear materials [7]. In particular, hexa-coordinated organotin complexes have gained considerable attention due to their large third-order nonlinear properties. Recently, Li et al. reported third-order nonlinear optical properties of an hexacoordinate organotin complex, where a $\chi^{(3)}$ value equal to $4.0 \times 10^{-21} \text{ m}^2/\text{V}^2$ was measured in the infrared region (Fig. 1, I) [8]. Our group has reported a series of hexa-coordinated organotin complexes with second-order nonlinear optical properties in the range of $10-60 \times 10^{-30}$ esu (Fig. 1, II) [9]. Meanwhile, Farfán et al. [10] reported second-order nonlinear optical properties of four diorganotin derivatives from Schiff bases, which first-order hyperpolarizability (β) value were on the order of 1.5 times higher than those of their boron counterparts (Fig. 1, III).

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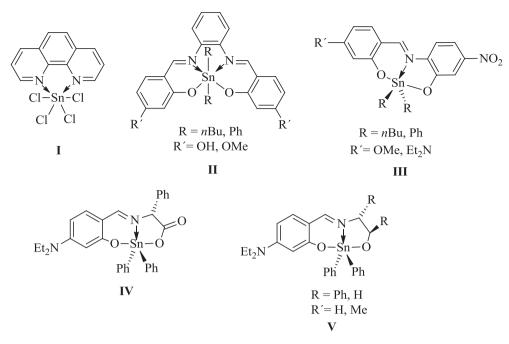


Fig. 1. Organotin compounds.

Furthermore, chiral organotin compounds have showed secondorder nonlinear optical efficiencies up to 8 and 11 times with respect to urea in the solid state [11] (Fig. 1, **IV**, **V**). It is important to note that studies of third-order nonlinear properties of organotin compounds in solid films at infrared wavelengths have been scarcely reported. In fact, there is but one paper, where the authors reported the third-order nonlinear susceptibility $\chi^{(3)}$ for **I** at 1064 nm. However, the literature contains many examples of boron compounds with third-order nonlinear properties (Fig. 2, **VI**, **VII**). For example, Ramos-Ortiz et al. [12] reported the third-order characterization of boron compounds derivatives from Schiff bases, where the authors report that the formation of the N \rightarrow B coordination bond increased $\chi^{(3)}$ three times with respect to the value exhibited by the ligand (Fig. 2, **VI**, **VII**). Recently, our group reported third-order susceptibility $\chi^{(3)}$ studies of boron compounds derived from Schiff bases (Fig. 2, **VIII**), with values of approximately 10^{-12} esu [13].

In continuation of our research, we report the microwaveassisted synthesis of four *push-pull* organotin complexes derived from Schiff bases in good yields providing an advancement over the chemical via and resulting in a simple, effective and reproducible alternative. All compounds were characterized by UV–Vis, TOF-HRMS, TGA-DTA, cyclic voltammetry, theoretical studies and compound **1** by X-ray single-crystal diffraction. Third-order nonlinear responses of complexes **1–4** were measured via Maker-Fringe technique. In particular, compounds **1** and **2** display larger third-order nonlinearities than their methoxilated counterparts.

2. Results and discussion

2.1. Synthesis

Compounds **1–4** were prepared by one-pot synthesis by irradiating the appropriate salicylaldehyde with 4-nitro-benzoylhydrazide and the corresponding diorganotin oxide with microwaves (Scheme 1). Table 1 compares, times and reaction yields in acetonitrile with those obtained by conventional method [5]. Our quantitative yields for traditional methods range from 37 to 96%. The microwave synthesis provides an excellent alternative, with yields ranging between 71 and 96% at reaction times of 12 min; meaning a decrease factor in time of 240 compared to traditional method.

2.2. Molecular structure of 1

Details of the crystal data of **1** and the summary of the collected parameters are given in Table 2, whereas selected bond lengths and angles are summarized in Table 3. X-ray structures for compound **1** showed a penta-coordinated tin atom (Fig. 3) with a distorted bipyramidal geometry, where oxygen atoms occupy axial positions, while nitrogen and two ipso-carbons occupy equatorial ones. The N–Sn distance is 2.137(2) Å, which is shorter compared to diphenylorganotin(IV) complexes derived from 2-hydroxy-1-naphthaldehyde 5-chloro-2-hydroxybenzoylhydrazone, previously reported (N–Sn distance is 2.158(3) Å) [14]. The Sn–O1

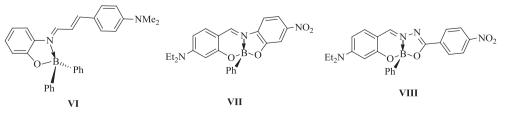


Fig. 2. Organoboron compounds.

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