



# Plasma-catalytic degradation of anthraquinonic acid green 25 in solution by gliding arc discharge plasma in the presence of tin containing aluminophosphate molecular sieves

H. Hentit<sup>a,\*</sup>, M.R. Ghezzar<sup>b</sup>, M. Womes<sup>c</sup>, J.C. Jumas<sup>c</sup>, A. Addou<sup>b</sup>, M.S. Ouali<sup>a</sup>

<sup>a</sup> Laboratoire de Valorisation des Matériaux, Université Abdelhamid Ibn Badis, BP188, Mostaganem 27000, Algeria

<sup>b</sup> Laboratoire des Sciences et Techniques de l'Environnement et de la Valorisation, Université Ibn Badis, BP188, Mostaganem 27000, Algeria

<sup>c</sup> Institut Charles Gerhardt, Equipe Agrégats Interfaces et Matériaux pour l'Energie (UMR 5253 CNRS), Université Montpellier II, CC 15, Place E. Bataillon, 34095 Montpellier Cedex 5, France

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## ABSTRACT

In this paper, the plasmacatalytic removal of an anthraquinonic dye type acid green 25 (AG 25) in aqueous solutions has been investigated in the presence of microporous tin containing aluminophosphates as catalysts under a gliding arc discharge plasma.

The catalysts have been synthesised in AEL and AFI structure types by static hydrothermal crystallisation. These materials were characterised by elemental analysis, X-ray diffraction, scanning electron microscopy, N<sub>2</sub> adsorption measurements (BET) and Mössbauer spectroscopy. These analyses showed significant catalytic properties of these materials.

The degradation process has been monitored during the experimental runs through UV/visible spectroscopy and chemical oxygen demand (COD). The results showed a good synergetic effect between the tin-containing aluminophosphate particles and the highly reactive plasma species. Moreover, it was found that these catalysts can be reused several times.

Kinetics of the dye removal has also been investigated on the basis of the Langmuir–Hinshelwood (L–H) model.

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## 1. Introduction

A number of advanced oxidation processes (AOP) such as direct ozonation, Fenton's reactions, electrochemical methods, corona discharge (DC), pulse corona discharge, photocatalysis, and UV photolysis, have been applied to degrade organic pollutants in aqueous solutions. Recently, another electrical discharge process called the gliding arc or gliding arc discharge has also been demonstrated to be effective at removing organic compounds from aqueous solutions and gases [1].

The gliding arc discharge (GAD) is an attractive AOP because it generates a plasma at atmospheric pressure and quasi-ambient temperature. In the presence of humid air, it generates •OH and •NO species whose standard oxidation–reduction potentials are  $E^\circ(\bullet\text{OH}/\text{H}_2\text{O}) = 2.85 \text{ V/SHE}$  and  $E^\circ(\text{ONOO}^-/\text{NO}_2) = 2.44 \text{ V/SHE}$ , respectively [2]. These oxidising abilities were used for

decontamination from micro-organisms [3] and surface oxidation of stainless steels [4]. Moreover, the GAD has emerged as an important destructive technology leading to the degradation of most of the organic pollutants such as: synthetic dyes [2,5–7], wastewaters from nuclear industry [8], endocrinal disruptor biphenol A [9].

The GAD process consumes a lot of electric energy when the treatment time is relatively long. To find a remedy to this and to make the electric treatment more efficient, some authors have combined the electric process with photocatalysts such as TiO<sub>2</sub> [10–13] and/or TiO<sub>2</sub> supported on activated modified carbon fibres [14]. In all these works, the strategy was based on the following points: (i) plasmachemical treatment of the pollutant, (ii) plasmacatalytic treatment of the pollutant in the same conditions, and (iii) kinetic study.

Microporous zeolite-like aluminophosphates (AIPOs) molecular sieves have attracted much interest since the first synthesis by Wilson [15] due to their catalytic and sorptive properties [16–21]. The AIPO structure consists of a combination of aluminium and phosphate ions where trivalent aluminium and pentavalent phosphorous are found in a tetrahedral environment in the dehydrated

\* Corresponding author. Tel.: +213 45 20 64 72; fax: +213 45 20 64 86.

E-mail addresses: [hentith@yahoo.fr](mailto:hentith@yahoo.fr), [hentith@univ-mosta.dz](mailto:hentith@univ-mosta.dz), [hentith@gmail.com](mailto:hentith@gmail.com) (H. Hentit).

material [22]. In the field of catalysis, the incorporation of various elements in the AlPO framework leads to the synthesis of new materials with novel acidic or redox properties [23–26].

Up to date, only few studies concerning the tin incorporated aluminophosphate molecular sieves (SnAPOs) have been reported [27–29]. Tin(IV) is one of the few elements besides Si that is expected to substitute for P in the aluminophosphate, producing a negatively charged framework possessing acid sites. Moreover, tin atoms act as excellent catalytic centres for oxidation reactions due to the ability of Sn to reversible changes of its valence (IV → II) [28].

The shape selectivity of the SnAPOs framework and their acid and redox properties allow us to propose the combination of these solids as catalysts with the GAD treatment. Therefore, the isomorphous introduction of tin into an AlPO<sub>4</sub>-5 (AFI) and AlPO<sub>4</sub>-11 (AEL) matrix has been studied in the present work using different physico-chemical methods. The catalytic performances of these tin-containing aluminophosphates were investigated by a plasma-removal process of anthraquinonic dye molecules type acid green 25 (AG 25).

## 2. Experimental

### 2.1. Catalysts synthesis

AlPO<sub>4</sub>-5, AlPO<sub>4</sub>-11, SnAPO<sub>4</sub>-5 and SnAPO<sub>4</sub>-11 have been synthesised hydrothermally according to the literature [15,27]. The aluminium and phosphorous sources were aluminium isopropoxyde (Fluka) and phosphoric acid (85%, Prolabo), respectively. Tin chloride hexahydrate (Aldrich) has been used as the tin source. The templating agents were triethylamine (98%, Fluka) for AlPO<sub>4</sub>-5 and SnAPO<sub>4</sub>-5 and di-*n*-propylamine (98%, Fluka) for AlPO<sub>4</sub>-11 and SnAPO<sub>4</sub>-11. The gel compositions and the synthesis conditions are given in Table 1. The starting mixtures have been heated in a Teflon-coated autoclave for crystallisation. Then, the autoclave has been quenched in water. The solid products have been filtered, washed with distilled water and air dried at room temperature overnight, then at 353 K for 8 h. Calcination of the molecular sieves has been realised in air at 823 K for 6 h to remove the organic template.

### 2.2. Catalysts characterisation

X-ray diffraction (XRD) was carried out on a Philips X'PERT diffractometer using CuK $\alpha$  radiation and a nickel filter. Scanning electron microscopy (SEM) has been performed on a FEI Quanta200 and analyses of the samples were conducted in attached X-rays microanalyser. BET surface area measurements, using nitrogen at 77 K, have been carried out with a micrometrics ASAP2010 equipment. All the samples have been degassed overnight at 493 K before the analysis. The isotherms have been used to calculate the BET specific surface area, total pore volume and average pore diameters according to the Horvath-Kawazoe method. Tin-doped materials were characterised by <sup>119</sup>Sn Mössbauer spectroscopy at room temperature in transmission geometry on a standard spectrometer equipped with components from Ortec & Wissel. Ba <sup>119m</sup>SnO<sub>3</sub> was used as the  $\gamma$ -ray source. The velocity scale has been calibrated using the magnetic sextuplet spectrum of a high purity iron foil absorber. Sn hyperfine parameters were determined by fitting Lorentzian lines to the experimental data using the ISO fit programme [30]. All isomer shifts are given with respect to BaSnO<sub>3</sub>.

### 2.3. Catalytic reaction

Anthraquinonic acid green 25 (AG 25; C<sub>28</sub>H<sub>20</sub>N<sub>2</sub>O<sub>8</sub>S<sub>2</sub>.2Na) was purchased from Agros Organic. Its chemical structure is shown in

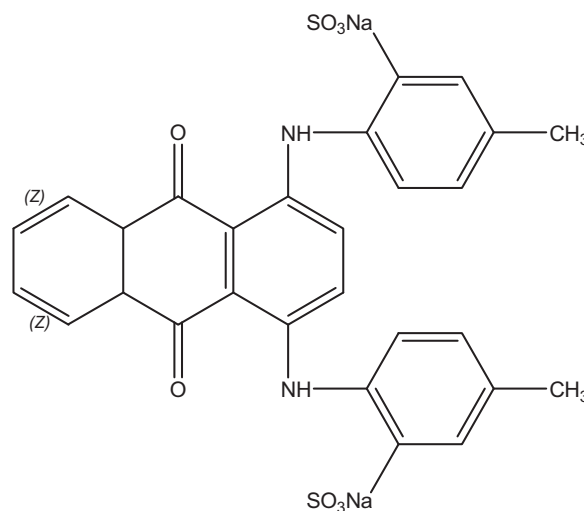


Fig. 1. Chemical structure of AG 25.

Fig. 1. Distilled water was used to prepare a dye solution with a natural pH of 5.1.

The gliding arc system used for this study is shown in Fig. 2. A detailed description can be found elsewhere [31]. A convenient voltage is applied to the two diverging electrodes between which the electric arc is formed. A special transformer (9000 V; 100 mA) provides the electric power. The arc is ignited at the point of minimal distance between the electrodes and is then pushed away from the ignition point by the feeding gas flow until it reaches the maximum of the electrode gap where it forms a large plasma plume. A new arc then appears and develops according to the same procedure. The plasma plume is disposed close enough to the liquid target, so that it licks the liquid surface, and allows the chemical reactions (1)–(10) to take place at the plasma–solution interface.

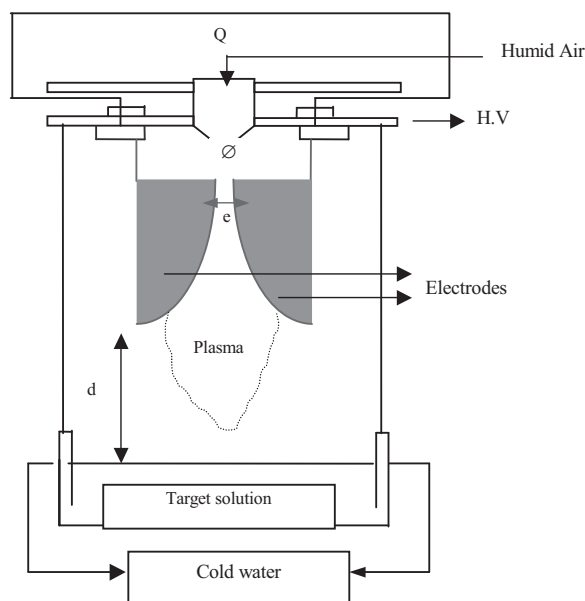
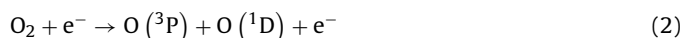


Fig. 2. GAD apparatus.

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