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Environmental applications of titania-graphene photocatalysts

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ABSTRACT

Nowadays, graphene is considered one important achievement as a consequence of its high potential in nanotechnologyand in the development of new environmental and energy processes. Reciently, graphene is receiving great attention in the area of photocatalysis, where is emerging in the next generation of photocatalysts, as a tool for enhancing photocatalytic performance and solar photoefficiency. Titanium dioxide hybridization with graphene has an effect on band gap energy decrease, shifting its absorption threshold to the visible light region and allowing to harness solar energy. So, the conjugation of graphene with semiconductor solid particles such as TiO_2, results in photocatalysts with improved charge separation, reduced recombination of the photogenerated electron-hole pairs, increased specific surface area, and introduces an adequate quantity and quality of adsorption sites, given that enhances their electronic, optoelectronic, electrocatalytic and photocatalytic properties.

This critical review sumarizes the recent progress in the design and synthesis of graphene-based titania semiconductor photocatalysts. Moreover, their applications in wastewater treatments, disinfection and air pollution control have been also discussed. Finally, some perspectives and challenges considered essential to extend the photoefficiency of these new photocatalysts in the visible region, to harvest directly solar light, have been suggested to introduce and elucidate new improvements in environmental photocatalytic processes.

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1. Introduction

In 2003, two researchers at The University of Manchester, Prof. Andre Geim and Prof. Kostya Novoselov, were managed to isolate from graphite "*a magnificent new wonder material that is a million times thinner than paper, stronger than diamond and more conductive than copper*". This strictly two-dimensional material that exhibited exceptionally high crystal and electronic properties was called graphene, and has revolutionized the physics community when the first paper appeared published in Science in 2004 [1]. Six years later, they were awarded the 2010 Nobel Prize in physics for this work.

Graphene is the name given to a single layer of carbon atoms densely packed into a benzene-ring structure, and is widely used to describe properties of many carbon-based materials, including graphite, large fullerenes, nanotubes, etc. (e.g., carbon nanotubes are usually thought of as graphene sheets rolled up into nanometer-sized cylinders) [2–4]. Fascination with this material stems from its remarkable physical properties and the potential applications these properties offer for the future. Nowadays, graphene is emerging on materials sience and condensed-matter physic fields, aiming for a wide range of technological applicacions [5–9],. Its high potential in nanotechnology applications converts to graphene in an important achievement which could change our lifes with the development of new processes in environmental and energy disciplines. Various methods, including thermal expansion [1] micromechanical exfoliation [10], epitaxial growth [11], chemical vapor deposition [12], chemical and electro-chemical reduction of graphite oxide [13,14] and bottom-up organic synthesis have been developed [15,16] from the first report on graphene isolated by manual mechanical cleavage of graphite with a Scoth tape [1]. Reduction of exfoliated graphene oxide (GO) has proven to be an effective and reliable way to obtain graphene nanosheets at low cost and great stability. Besides, via chemical modification can be well adjusted surface properties of graphene, which offers excellent chances for the progress of functionalized graphene-based materials [17,18]. They are very attractive for many potential applications, such as energy storage [19], catalysis [20], biosensors [21], molecular imaging [22], drug delivery [23], nanomedicine and more concretely stem cell-based tissue engineering [24], and recently nanomotors [25]. All of them are posible, fundamentally, because graphene shows distinctive electronic and optical properties with good biocompatibility. Concretely, this material is receiving great attention in the area of

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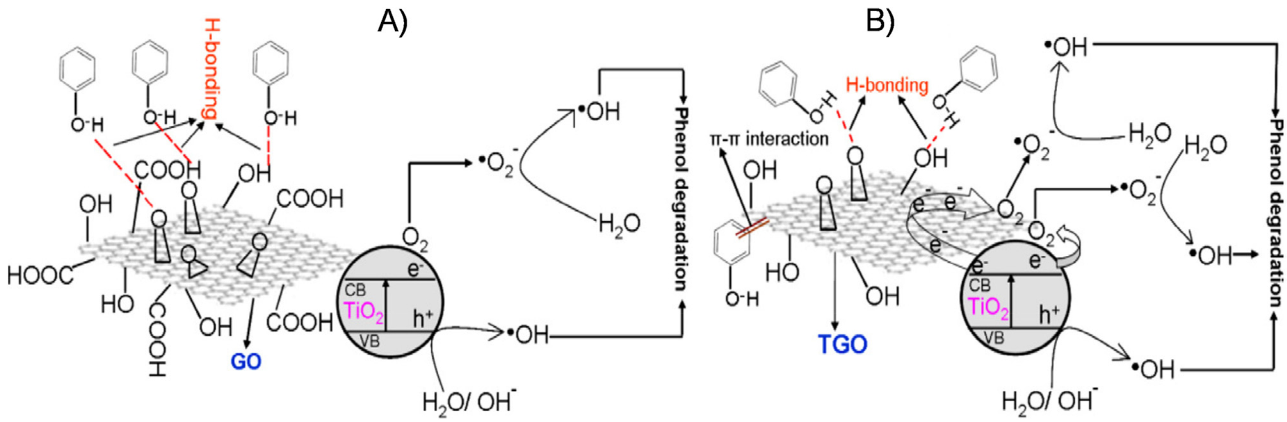

Fig. 1. Proposed mechanisms of phenol degradation in oxygen by TiO_2-graphene composites: A) Without e^- transfer from TiO_2 conduction band (CB) to graphene. B) With e^- transfer. (from [48]).

nanotechnology and photocatalysis due to its outstanding properties, especially those which exploit its high electron mobility and chemical stability [26,27], and its high adsorption capacity [28,29]. In photocatalysis area, semiconductor materials have been the focus of numerous investigations due to its application to the quantitative destruction of undesirable chemical contaminants in water and air [30,31], and to solar energy conversion [32].

Although several oxide semiconductors have photocatalytic properties, the polycrystalline powders of titanium dioxides present unique properties since it is a stable material, high relative photoactivity, high chemical inertness, nontoxicity and low cost [33,34]. However, many problems need to be solved in the TiO_2 photocatalyst system for practical applications, such as narrow UV spectrum response range because of a large band gap energy (~3.0 eV for rutile and 3.2 eV for anatase) and relatively fast recombination of the photo-generated e^-/h^+ pairs [32]. Hole-electron recombination is a serious problem for the development of photo-catalytically based technologies since it severely limits the quantum yields achievable [35]. Therefore, various methods have been developed to extend the absorption range of titania into the visible region and to improve its photocatalytic activity, and there are strategies to reduce electron-hole recombination rates and increase photocatalyst efficiency. For instance, via suitable textural design, doping with noble metals, such as Pt, Ag, Au, Pd, etc, transition metal cations, non-metallic doping or immobilization on adsorbent surface and forming semiconductor composites have been employed [36–44]. In this line, also carbon nanomaterials such as fullerenes, carbon nanotubes and graphene are recently receiving a great attention in photocatalityc process, predominantly graphene, because of its unique properties, including large specific surface area, flexible structure, excellent mobility of charge carriers at room temperature and good electrical and thermal conductivities [45,46]. So, graphene is emerging as one of the most promising materials in the next generation of photocatalysts [47], given that has been embraced in the design of new photodegradation catalysts as a tool for enhancing their photocatalytic performance [48]. The excellent properties of graphene are important features when dealing with the preparation and use of graphene-based materials, thus graphene-based TiO_2 composites are being developed and successfully applied as photocatalysts for the abatement of pollutants, hydrogen production and others applications [44,49–51]. Where the most widely used synthesis procedures with semiconductors such are TiO_2 are in situ growth, solution mixing, sol-gel, hydrothermal and/or solvothermal methodologies [44]. Titanium dioxide hybridization with graphene has an effect on band gap energy decrease, thus shifting the absorption threshold to the visible light region and allowing utilization of solar energy [30]. The excited electrons could be transfered from the conduction band of TiO_2 to the surface of graphene, hence improving the separation of the electron-hole pairs and preventing their recombination [52,53], because it has been already proven that the electron accepting and transport properties of graphene provide a convenient way to direct the flow of photo-generated charge carriers, which thus increases the lifetime of e^-/h^+ pairs generated by TiO_2 upon light irradiation [54,55].

Therefore, graphene is a suitable alternative to noble metals because it has a high conductivity, high surface area, and the ability to favor the electron transfer from the conduction band of TiO_2 to delocalized aromatic structure of graphene [56]. So, the conjugation of graphene with semiconductor solid particles, such as TiO_2, results in a photocatalyst with improved charge separation, reduced recombination of the photogenerated electron-hole pairs, increased specific surface area, and introduces an adequate quantity and quality of adsorption sites, that lead to enhance their electronic [57], optoelectronic [58], electrocatalytic [59] and photocatalytic properties [60].

2. Mechanistic aspects of titania-graphene photocatalysts

It is known carbonaceous materials play an important role in photocatalytic processes given that usually present exceptional adsorption ability to many type of pollutants. In this line, TiO_2-graphene based catalysts can in turn, if photo-generated charges are transferred toward graphene, improve the final efficiency of the process [30,31]. However, there is not a complete agreement on the real role of the presence of graphene on these types of composites along photocatalytic process. Often, different operational parameters and some of the most relevant properties of TiO_2-graphene composites can even affect their final photocatalytic efficiency, such as type of substrates, irradiation light (UV–vis or Vis), etc. In the work of Minella et al. [61] a complete summary of the operational mechanism alternatives acting on TiO_2-rGO composites is presented. They discuss three types of photo-mechanism, concluding as follow:

1) When substrates are not absorbing light, and are hardly being adsorbed on the catalyst surface, a typical UV-based photocatalytic process can happen, supported by UV-activated e^- transition from valence band (VB) to conduction band (CB), which originates e^-/h^+ pairs (Fig. 1A). In this case, no electron is transferred between TiO_2 and rGO, and rGO can act only as competitive light absorber. The extension of light absorption to visible range could happen due to the presence of graphene which can even create states between the VB and CB of TiO_2.

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