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Preparation of Er^{3+}:$Y_3Al_5O_{12}$/$KNbO_3$ composite and application in innocent treatment of ketamine by using sonocatalytic decomposition method

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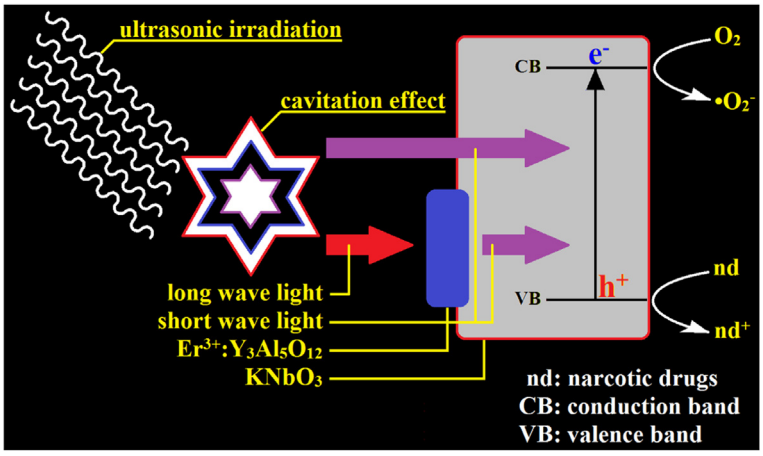
HIGHLIGHTS

- Upconversion luminescence agent Er:YAG can enhance sonocatalytic activity of $KNbO_3$.
- Harmless of narcotic drugs was achieved through sonocatalytic destruction.
- Possible sonocatalytic destruction mechanism on narcotic drugs was proposed.

GRAPHICAL ABSTRACT



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ABSTRACT

A novel sonocatalyst, Er^{3+}:$Y_3Al_5O_{12}$/$KNbO_3$ composite, was synthesized, and then, characterized by X-ray diffractometer (XRD), scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX). In order to evaluate the sonocatalytic activity of prepared Er^{3+}:$Y_3Al_5O_{12}$/$KNbO_3$ composite, the sonocatalytic degradation of ketamine, a kind of narcotic drug, was studied. In addition, some influencing factors such as mass ratio, heat-treated temperature and heat-treated time on the sonocatalytic activity of prepared Er^{3+}:$Y_3Al_5O_{12}$/$KNbO_3$ powders and ultrasonic irradiation time on the sonocatalytic degradation of ketamine were examined by using GC–MS machine. The experimental results showed that the Er^{3+}:$Y_3Al_5O_{12}$/$KNbO_3$ composite is a good sonocatalyst in the field of ultrasonic chemistry and the sonocatalytic degradation was an effective method for the innocent treatment of ketamine.

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1. Introduction

In the present-day world, the flooding of narcotic drugs has become a big threat, which is causing serious damage to the society and all mankind [1–3]. Since the criminals have mastered advanced production techniques, more and more types and amounts of narcotic drugs are being produced. Therefore, for confiscated narcotic drugs the effective destruction and treatment technologies are very necessary. Some traditional treatment methods of narcotic drugs were most often used in many country [4–8]. However, these technologies have still exhibited many disadvantages or limitations [9,10]. In addition, they may also lead to the second pollution [11,12]. The narcotic drugs are generally some special organic compound molecules with stable structure and fixed composition. It is difficult to thoroughly degrade and mineralize them. Of course, for these special organic compounds (narcotic drugs), if their structures or compositions are destroyed just a little, due to the small changes they will lose the toxicity. Considering the particularity of narcotic drugs, some Advanced Oxidation Processes (AOPs) may be feasible methods [13,14].

Recently, the sonocatalytic degradation method has caused attention in the treatment of organic pollutants [15–20]. Sonocatalytic degradation of organic pollutants can be explained by the ultrasonic cavitation effect happened in aqueous medium, which leads to the sonoluminescence and "hot spot" [21–25]. During ultrasonic cavitation the produced "hot spot" can give transient high temperatures (5000 K) and high pressures (1000 atm) [26–29]. The transient high temperatures cause the thermal dissociation of water molecules to produce hydroxyl radicals (•OH) which can degrade and destroy narcotic drugs [30,31]. Besides, the sonoluminescence resulted from ultrasonic cavitation effect can generate light with a comparatively wide range of wavelengths, which can excite the aromatic organic pollutants to become unstable in energy. However, in actual applications, the efficacy using ultrasonic irradiation alone to degrade organic pollutants is not high [28]. In order to efficiently carry out the degradation of organic pollutants under ultrasonic irradiation, the key is selecting some semiconductor materials as sonocatalyst to perform the sonocatalytic degradation reaction [32,33].

In the past, TiO_2 has been used as a sonocatalyst in the sonocatalytic degradation of organic contaminants by most researchers [34,35]. However, compared with some other semiconductor materials with wide-band gap and high oxidation potential, the TiO_2 is not best choice [36]. A lot of research showed that some perovskite niobates can enhance the efficiency of photocatalytic oxygen evolution at least by an order of magnitude [37,38]. Hence, $KNbO_3$ can be considered as a promising sonocatalyst [39,40]. The band gap (3.5 eV) of $KNbO_3$ is slightly wider than that (3.2 eV) of TiO_2, so the $KNbO_3$ may display a high sonocatalytic activity under ultrasonic irradiation [36]. Nevertheless, due to the wide-band gap, the $KNbO_3$ can only absorb high-energy light ($\lambda_{max} \leq 360$ nm) to perform photocatalytic reaction. Using pure $KNbO_3$ the sonocatalytic degradation efficiency will be very low. Thus, it is necessary to provide sufficient high-energy light to maintain the high sonocatalytic activity.

In recent years, we used some up-conversion luminescence agents to combine with wide-band gap semiconductor materials to enhance the sonocatalytic activity and that achieved satisfactory results [41,42]. The up-conversion luminescence agents can absorb two or more low-energy photons generating one high-energy photon. The produced high-energy lights can excite wide-band gap semiconductor materials and help to realize the high sonocatalytic activity. As up-conversion luminescence agent, the Er^{3+}:$Y_3Al_5O_{12}$ has been widely used in many fields due to its high conversion luminescence efficiency and chemical stability [43]. Therefore, in this work, we try to use Er^{3+}:$Y_3Al_5O_{12}$ combined with $KNbO_3$ to prepare a novel Er^{3+}:$Y_3Al_5O_{12}$/$KNbO_3$ sonocatalyst performing the sonocatalytic degradation of ketamine under ultrasonic irradiation.

In this paper, Er^{3+}:$Y_3Al_5O_{12}$ powder was prepared through sol-gel-hydrothermal method, and then combined with $KNbO_3$ by direct mixing and calcination methods as a new sonocatalyst for sonocatalytic degradation of ketamine. Meanwhile, some main influence factors on the sonocatalytic degradation activity of Er^{3+}:$Y_3Al_5O_{12}$/$KNbO_3$ was investigated. Furthermore, the mechanism of up-conversion luminescence process of Er^{3+}:$Y_3Al_5O_{12}$ and the sonocatalytic degradation of ketamine caused by Er^{3+}:$Y_3Al_5O_{12}$/$KNbO_3$ under ultrasonic irradiation were also proposed.

2. Experimental

2.1. Materials and reagents

Niobium pentoxide (Nb_2O_5) and potassium hydroxide (KOH) (Sinopharm Chemical Regent Co, Ltd, China) were used to prepare the potassium niobate ($KNbO_3$). Er_2O_3 (99.999% purity), Y_2O_3 (99.999% purity) and $Al(NO_3)_3{\cdot}9H_2O$ (analytically pure), citric acid (analytically pure) and HNO_3 (65–68%, analytically pure) (Veking Company, China) were used to synthesize the Er^{3+}:$Y_3Al_5O_{12}$ as up-conversion luminescence agent. Ketamine (99.99% purity, the ministry of public security anti-drug information technology center, China) was used to undergo the sonocatalytic degradation for innocent treatment. All the reagents were of analytical purity grade, and were directly used without further purification.

Muffle furnace (SX2-4-10, Great Wall Furnace Company, China) and oven (101-1, Shanghai Experiment Apparatus Company, China) were used to prepare the Er^{3+}:$Y_3Al_5O_{12}$/$KNbO_3$ as sonocatalyst. X-ray powder diffractometer (XRD, D-8, Bruker-axs, Germany, Ni filtered Cu Ka radiation in the range of 2θ from 10° to 70°) and scanning electron microscopy (SEM, JEOL JSM-5610LV, Hitachi Corporation, Japan) were used to determine the crystalline phase and surface morphology. Energy dispersive X-ray spectroscopy (EDX, JEOL JSM-5610LV, Hitachi Corporation, Japan) was used to determine the element type and composition content. GC–MS machine (Agilent 7890A/5975C, Agilent Technologies Inc, USA) was used to inspect the sonocatalytic degradation processes and decomposition products of ketamine in aqueous solution. Controllable Serial-Ultrasonics apparatus (KQ-300, Kunshan Company, China) was adopted to irradiate the ketamine solution, operating at ultrasonic frequency of 40 kHz and output power of 300 W through manual adjust.

2.2. Preparation of $KNbO_3$ powder

$KNbO_3$ particles were prepared by using a hydrothermal method following the work reported by Yan et al. [44]. 3.57 g Nb_2O_5 and 37.69 g KOH were added into 19 mL distilled water, which was stirred vigorously for 30 min. Then the mixture was sealed in a Teflon-lined stainless steel autoclave and heated at 160 °C for 12 h. The products were washed with distilled water and ethanol several times, and then dried at 80 °C overnight. Finally, the $KNbO_3$ particles were obtained and stored for the sonocatalytic degradation.

2.3. Synthesis of Er^{3+}:$Y_3Al_5O_{12}$ as up-conversion luminescence agent

The Er^{3+}:$YAlO_3$ was synthesized by the sol-gel and calcination method [36]. Er_2O_3 and Y_2O_3 powders were dissolved in HNO_3 with magnetic stirring and heated until transparent. Appropriate amount of aluminum nitrate solution was added to the above transparent solution. Then citric acid was added as chelating agent

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