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Non-catalytic applications of g-C_3N_4: A brief review

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ABSTRACT

The g-C_3N_4 which is well known as a polymeric non-metal semiconductor, has been fabricated by thermal polymerization. It has also been used in catalytic applications including, photo-catalysis, removal and degradation of pollutants in water, Friedel-Crafts reactions, oxygen reduction reaction and etc. It has drawn noticeable research attention due to its economical and affordable fabrication, non-toxicity, biocompatibility, good thermal and electrical conductivity, high hardness, Corrosion resistance, and fireproofing properties. Therefore, the g-C_3N_4 has found non-catalytic applications including composites, cutting tools, improving surface properties, light emitting devices, optical sensors, and solar cells. In the current review, the novel and non-catalytic applications of g-C_3N_4 have been highlighted.

KEYWORDS

Graphitic carbon nitride (g-C_3N_4)
Cutting tools
Composite
Non-catalytic applications

1. Introduction

Among different C_3N_4 carbon nitride allotropes i.e. the g-C_3N_4, α-C_3N_4, β-C_3N_4, cubic C_3N_4, and pseudo-cubic C_3N_4, the g-C_3N_4 (graphitic carbon nitride) which is classified as a polymeric material, is the most stable specie under the ambient condition [1-4]. Blasius et al. (1834) have investigated a material that was named "melon" (linear polymers of connected tri-s-triazines via secondary nitrogen) and had been reported by Liebig [5-8]. They could be synthesized by substituting carbon atoms with nitrogen in carbon materials. Heating up followed by a single-step polymerization of cheap organic feedstock such as dicyandiamide, cyanamide, urea, thiourea, and melamine which is containing C and N, with earth-abundant elements have been used for preparing Graphitic carbon nitrides [6-10]. Unlike conventional organic semiconductors, g-C_3N_4 exhibits unique thermal and chemical stability[11, 12]. Due to other special properties of g-C_3N_4 such as non-toxicity, medium band gap, economical productivity, and environmental compatibility, different studies on g-C_3N_4 have been highly regarded [13-16]. The g-C_3N_4 has been used as a catalyst for different applications such as water treatment [17], Friedel-Crafts reactions [18, 19], and oxygen reduction reactions [20,21]. Wastewater treatment and recycling is one of the most challenging problems in the world, particularly the wastewater produced by chemical industries, because of its remarkable concentration of large organic fragments which are tremendously poisonous and carcinogenic [22, 23]. Polymeric materials have been widely employed in water treatment applications. Graphitic carbon nitride, a two-dimensional organic polymeric material is also used as an effective adsorbent and photo-catalyst for the fast removal and degradation of various pollutants in wastewater [24].

Recently, the g-C_3N_4 has been used in non-catalytic applications for example as a photo-electronic material. It has been used also as a sintering aid and reinforcement agent in manufacturing ultra-high temperature composites (UHTCs) fabricated by spark plasma sintering (SPS). The SPS is a new method used to fabricate the parts through powder technology [25]. The SPS uses the axial pressure and electrical current concurrently based on the Joule heating phenomenon to bond the powder particles. The Joule heat source and the material properties such as thermal conductivity, electrical resistivity, specific heat, and density are important parameters in the SPS process. Spark plasma sintering has been used to fabricate which are difficult to fabricate by more common methods such as casting. Ultra-high-temperature ceramics have been generally fabricated by the SPS process [26-28]. UHTCs have attracted many of attention due to their desirable mechanical properties, thermal and chemical stability at high temperatures, very high hardness, and corrosion resistance [29, 30].

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Using the SPS process in the manufacture of UHTCs will be very useful [31]. The SPS process has faced some challenges in cases in which powder has low sinter-ability, poor oxidation resistance, and low self-diffusion coefficient [32]. Some research has been carried out to solve the mentioned problems in order to improve the properties of parts manufactured with the SPS process by adding the g-C_3N_4 as a sintering aid and reinforcement phase. This review study has highlighted novel and non-catalytic applications of g-C_3N_4 (Fig. 1).

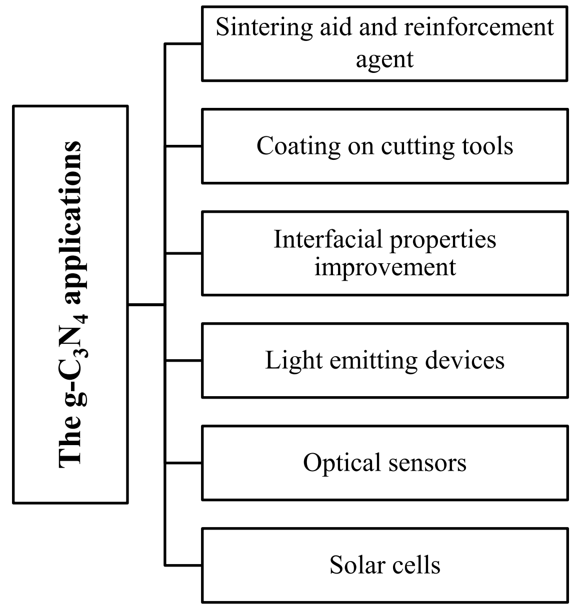

Fig. 1. Non-catalytic applications of g-C_3N_4.

2. Applications

2.1. Sintering aid and reinforcement phase in composites

Ahmadi et al. [9] have used the g- C_3N_4 as a sintering aid and reinforcement phase in the ZrB_2 Matrix fabricated through the SPS process. They have attempted to solve the sintering problems of ZrB_2 and improve the properties. They have reported that by the addition of 5 wt% of g-C_3N_4 to ZrB_2-based composite, the sinter-ability significantly improved. The relative density in g-C_3N_4 doped ZrB_2 ceramics in comparison with monolithic ZrB_2 is reduced from 76.5% to an approximately fully dense ceramic (to about 99.8%).

According to Fig. 2a, porosities are observed in the sintered sample in absence of C_3N_4 which showed incomplete sintering. By adding C_3N_4 to the ZrB_2 matrix, the in-situ synthesis of new phases has taken place in the C_3N_4-ZrB_2 composite which has been propagated in the pore-free ZrB_2 matrix (Fig. 2b).

The XRD analysis indicated that adding 5 wt% C_3N_4 to ZrB_2, has revealed some new peaks in the ZrB_2 XRD pattern. Generally, the ZrB_2, C_3N_4, ZrC, and BN peaks were identified. The oxide impurities also have been detected in the sintered sample after the SPS processing. The new phases could have been formed according to the following reactions [9].

$$C_3N_4 = 3C + 2N_{2(g)} \quad (1)$$

$$ZrB_2 + C + N_{2\,(g)} = ZrC + 2BN \quad (2)$$

$$ZrO_2 + 3C = ZrC + 2CO_{(g)} \quad (3)$$

$$B_2O_3 + 3C + N_{2\,(g)} = 2BN + 3CO_{(g)} \quad (4)$$

The mechanical properties also have been improved by adding 5 wt% C_3N_4 to the ZrB_2 matrix, due to densification enhancement and grain growth reduction. The Vickers hardness, fracture toughness, and flexural strength have been increased from 10.1 GPa, 1.9 MPam^{½}, and 187.6 MPa to 16.2 GPa, 5.4 MPam^{½}, and 516.4 MPa, respectively [33].

2.2. Coating on cutting tools

The C_3N_4 is a very hard and resistant to wear component which has a low coefficient of friction, and acceptable thermal conductivity [34, 35]. Therefore using the C_3N_4 as a coating for cutting tools seems a very attractive idea. Yang et al. [36] have applied a C_3N_4 film on the CN-coated YT15 cutter. They have used a lathe machine to examine the performance of CN-coated YT15 cutting tools on a cylinder sample made of hardened 45 steel. The machining condition has been as follows:

- Dry cutting (without liquid)
- Tool work angles: γ_o = 15 °, α_o = 10 °, K_r = 45 °, and r_ε = 0.12 mm
- Machining parameters: V_c = 40-90 m/min, ap = 0.1–0.4 mm, and f = 0.03–0.12 mm/r.

The tool flank wear-time curve is shown in Fig. 3. The tool life has been increased remarkably with the C_3N_4 coating compared to the uncoated one. The film also has enhanced the tool's thermal resistance which has made it an appropriate choice for dry-cutting processes.

Three types of wear have been investigated, i.e. flank, rake, and crater wear. The main forms have been the rake and flank wear. The rake

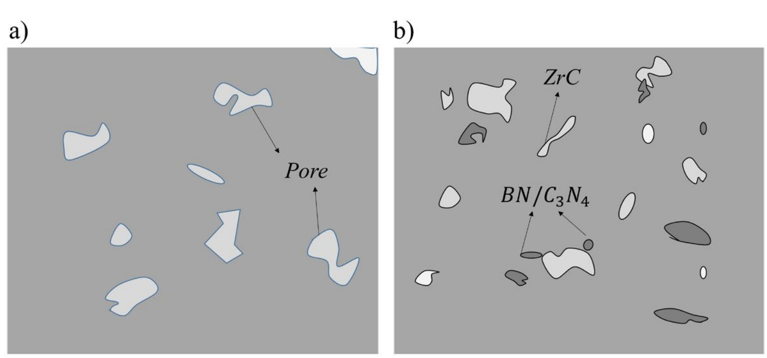

Fig. 2. The schematic representation of the polished surfaces of a) ZrB_2 and, b) C_3N_4-doped ZrB_2 ceramics [9].

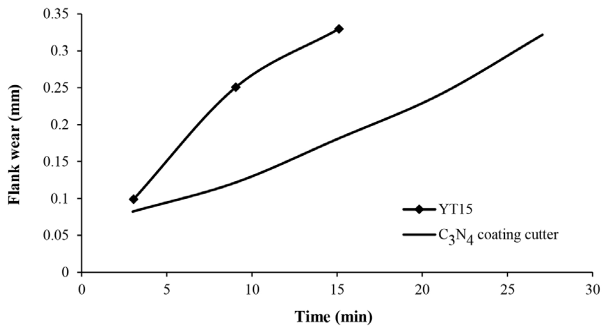

Fig. 3. The tool flank wear-time curve under the condition: V_c = 60 m/min, f = 0.1 mm/r, ap = 0.4 mm [36].

wear has been the dominant one during high-speed cutting with a higher cutting depth. The worn surface of the tool is shown in Fig. 4.

2.3. Agent to improve the interfacial properties of carbon fiber

The application of graphitic carbon nitride in polymer matrix composites is developing a stronger interface between carbon fiber and epoxy [37–40]. The stronger adhesion between the matrix and the reinforcement leads to better mechanical properties, otherwise, the volume occupied by the secondary phase in the matrix will act as a discontinuity which will weaken the properties [41].

Song et al. [40] have increased the roughness and wetting ability of carbon fiber through an in situ precipitation of C_3N_4 on carbon fiber. The nitride changed the surface morphology of carbon fiber (Fig. 5). The addition of C_3N_4, in fact, increased the roughness of the carbon fibers. The inter-laminar shear strength (ILSS) and interfacial shear strength (IFSS) of composites reinforced with the fibers have been increased from 44.3 and 43.1 MPa to 60.7 and 75.9 MPa, respectively. Moreover, the surface free energy of carbon fibers has been increased by 65.6%. The improved interfacial properties have resulted in a higher tensile strength (from 1063 to 1279 MPa) and total absorbed energy of impact examination (from 1.22 to 1.75 J). The dynamic mechanical properties and hydrothermal aging resistance have been enhanced significantly. In addition, the storage modulus has been enhanced from 64.3 to 74.1 GPa.

The presence of nitrogen improves the properties of carbonaceous materials including mechanical properties, structural, and in particular electrical properties. The sp^2 hybridization between nitrogen and carbon forms a π-conjugated electronic structure which endows it with excellent photoelectronic properties [6]. Nowadays, due to the particular electrical properties of C_3N_4, it has found special applications in solar cells, fuel cells, light-emitting devices, and batteries.

2.4. Light-emitting device

Recently, due to the semi-conductivity of g-C_3N_4, numerous investigations have been concentrated on the luminescence properties of g-C_3N_4 [42, 43]. Some studies have been performed to explain the mechanism of visible photoluminescence (PL) of carbon nitride. The photoluminescence area is dominated by the optical band gap and it was indicated that the band gap of g-C_3N_4 is tunable with processing temperature [44, 45]. Zhang et al. [46] have studied the photoluminescence tune-ability of synthesized g-C_3N_4 nanopowder. The powder has been fabricated by low-temperature thermal condensation of melamine. They observed phase transformation from melamine to amorphous g-C_3N_4 by increasing the processing temperature. They reported that the morphology altered from a granular to a layer structure with different dimensions of flakes. The resulting emission area of the samples envelops from 400 nm to 510 nm, varying from blue-violet to green by continuously controlling the processing temperature.

Because of the biocompatibility and non-toxicity of C_3N_4, its application instead of noxious substances in LEDs has drawn considerable attention. Conventional LEDs contain hazardous components such as PbS, CdTe, CuInS, CdSe, and CdS, which raise toxicity and pollution problems. The advantages of white-light-emitting diodes (WLEDs) compared to conventional LEDs, are including high efficiency, low power consumption, extended lifetime, and fast response which has introduced the WLEDs as a suitable alternative to conventional LEDs.

Wang et al. [47] have used the g-C_3N_4 silica-gels to develop WLEDs with non-metal and biocompatible materials. They have synthesized pure g-C_3N_4 nanoparticles by heating up a mixture of 0.4 g of anhydrous citric acid and 0.4 g thiourea in the air at 180 °C for 2 hours. The dip coating technique has been used to precipitate a g-C_3N_4 film on the glass of the LED lamp. The optimal wavelength of 365 nm also has been selected. The fabricated WLED had more flexibility, lower cost, higher quantum efficiency and color rendering index of 70, and also higher transparency.

2.5. The optical sensors

The need for accurate, sensitive, rapid, and simple detectors that have sufficient biocompatibility and reliability becomes more and more every day. Optical sensors are molecular receptors whose optical properties change upon binding to specific guests. One of the most important applications of these sensors is to detect metallic ions (chromogenic/fluorescent receptors) [5, 48]. Many studies have tried to reduce the limitations and enhance the kinetic response of sensors. The g-C_3N_4 could carry functional groups such as NH_2/NH/N on its surface to endow enormously absorption of metal ions via redox or chelation reactions. Regarding the current features of chromogenic/fluorescent receptors, the g-C_3N_4 is an attractive candidate for employment in optical sensors. The sensitivity of the g-C_3N_4 is higher than the sensors with optical receptors because optical receptors hooks on the porous medium while g-C_3N_4 acts as a receptor itself [48, 49].

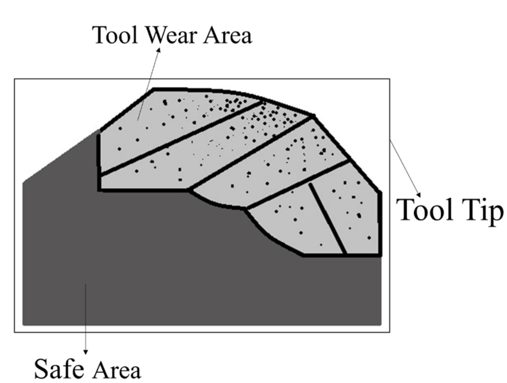

Fig. 4. The schematic image for rake wear in C_3N_4-coated cutter [36].

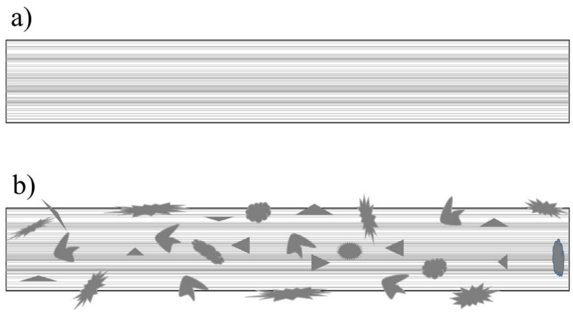

Fig. 5. The schematic micrograph of a) carbon fiber; and b) carbon fiber coated by C_3N_4 [40].

2.6. Solar applications

The use of fossil fuels is facing challenges due to limited resources, pollution, environmental crises, and global warming. Thus, clean energy production from renewable and biocompatible sources especially solar energy become a very important issue. Among various types of solar cells including Bio-hybrid, Buried Contact, Cadmium Telluride, and Concentrated PV Cell, dye-sensitized solar cells (DSSC) are very interesting because of easy fabrication, low cost, and environmentally friendly properties [50]. The g-C_3N_4, as a tenuous band gap semiconductor and sufficient visible light-absorbing abilities, could be an additive to enhance solar cell performance [51]. Xu et al. [52] have fabricated TiO_2@g-C_3N_4 nano-sheets by heating the mixture of urea and TiO_2 nano-sheets employed as a photo-anode material of DSSCs. The g-C_3N_4 nanosheet is a π-conjugated material, which is a proper choice for separating the photogenerated electron-hole pairs. The g-C_3N_4 films could act as a blocking layer that efficiently prevents charge recombination at the TiO_2/electrolyte interface in DSSC. As a consequence, the thin layer of the g-C_3N_4 on the TiO_2 surface can effectively promote electron transportation by prorogating the backward recombination of electrons from TiO_2 and electrolyte and contributing additional electrons to boost the electron concentration in the photo-anodes. This leads to enhancing the performance of DSSC.

3. Conclusions

Graphitic carbon nitride (g-C_3N_4) as an appealing member of materials with attractive properties has been used widely in different applications. The g-C_3N_4 had been mostly used in catalytic applications such as photo-catalysis, removal, and degradation of water pollution, Friedel-Crafts reactions, and oxygen reduction reaction. Recently, it has been used in some non-catalytic applications due to its good thermal and chemical stability, proper mechanical properties, applicable energy band gap, non-toxicity, and cost-effectiveness. It has found special applications in materials engineering including composites reinforcement, sintering aid, coating of cutting tools, solar cells, light-emitting devices, and medical applications.

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