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Application of CO_2 reduction technology based on copper-modified phosphorus-doped carbon nitride catalyst in low-carbon housing construction

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Abstract The world is currently facing a serious carbon emission problem, and the construction industry is one of the major emission sources. The development of efficient and low-cost catalytic technologies for CO_2 reduction has become the key to promote the transition to low-carbon buildings. Carbon nitride has received widespread attention due to its tunable energy band structure and environmental friendliness, and the combined modification strategy of copper modification and phosphorus doping is expected to significantly improve its photocatalytic efficiency. In this paper, we explore the potential of copper-modified phosphorus-doped polymerized carbon nitride (CMPDPCN) materials based on its application in photocatalytic CO_2 reduction and roof photovoltaic carbon-reduction systems in housing construction. In the study, firstly, block PCN, normal PCN, VAEPCN and CMPDPCN were prepared and their optical properties were analyzed by UV-vis spectroscopy and photoluminescence spectroscopy. Comparative CO_2 photodegradation tests were conducted using porous cement composite catalyst samples, and the results showed that the CO_2 degradation rate of the CMPDPCN sample reached 75.64% after 120 min of light exposure, which was much higher than that of the VAEPCN sample at 28.20%. Further roof photovoltaic simulations showed that the CMPDPCN module could enhance the power generation to 38.75 MW-h and the CO_2 degradation rate to 53.84% under 100m² condition. The above results indicate that copper-modified phosphorus-doped carbon nitride has significant application advantages in low-carbon buildings.

Index Terms carbon nitride, photocatalysis, CO_2 reduction, copper modification, building carbon reduction, photovoltaic system

I. Introduction

Improvement of economic conditions has led people to face the serious situation of resource scarcity and environmental deterioration while continuously pursuing living comfort and material enjoyment [1], [2]. Excessive greenhouse gases such as CO2 and N2O emitted in production and life have caused the global greenhouse effect, which has a bad impact on the environment [3]. Among them, construction activities are one of the activities that have the greatest impact on natural resources and ecological environment [4]. Globally, nearly half of the total energy is consumed by construction activities, nearly 40% of natural resources are also used in construction activities, nearly half of the greenhouse gases also come from the emissions of construction activities, and the amount of construction waste is alarming [5]-[7]. It is not difficult to find press, compared to other industries, the construction industry is large and widely distributed, causing a wide range of pollution [8], [9].

The continuous growth of energy and resource consumption as well as the increasing prominence of environmental issues have forced people to start thinking about how to change the development mode and realize the sustainable development of the construction industry [10]-[13]. Therefore, reducing carbon emissions from the production of building materials and the construction and operation of buildings will become the main focus of responding to climate change, and strengthening the promotion and application of low-carbon and carbon-negative technologies, such as CO2 reduction technology, in buildings has become a new way to alleviate the pressure of carbon emissions and solve the climate problem [14]-[17]. At the time of China's comprehensive construction of new socialist cities, the introduction of low-carbon technologies into residential construction and the construction of harmonious human settlements can effectively save resources, reduce energy consumption and minimize environmental pollution.

Under the guidance of the global goal of “carbon peak and carbon neutral”, the green transformation of the construction industry, as one of the major fields of energy consumption and carbon emission, has become a key direction to promote the national dual-carbon strategy. In recent years, the research on photocatalytic materials has continued to advance, providing a new path for the efficient conversion and emission reduction of carbon dioxide. In particular, polymerized carbon nitride (PCN) has received wide attention in photocatalytic CO_2 reduction research due to its stable structure, easy synthesis and good photoresponsive properties. However, PCN still faces problems such as large forbidden band width, insufficient specific surface area, and high photogenerated carrier complexation rate, which limit its catalytic efficiency. Therefore, modification strategies to enhance the photocatalytic performance of PCN are in urgent need of a breakthrough. In this paper, we focus on the synergistic modulation of copper modification and phosphorus doping to construct CMPDPCN catalytic materials with high visible-light response, and systematically analyze their carbon-reducing performance in housing photovoltaic components and porous building materials. The study includes two main aspects: first, at the material level, four material systems, namely, block PCN, ordinary PCN, VAEPCN and CMPDPCN, are constructed to analyze the differences in their optical properties such as spectral response and excited state behavior, and their performance advantages are clarified through the CO_2 photocatalytic degradation test; second, at the application level, the model of the PV-energy storage system of the building roof is constructed to quantitatively assess the CMPDPCN embedded PV power generation and CO_2 emission reduction rate of the degree of enhancement, to verify its technical feasibility and environmental benefits in the actual housing decarbonization path, to provide technical support for the design of green building materials.

II. Construction of carbon nitride nanosheets and its photocatalytic CO_2 reduction study

II. A. CO_2 reduction based on carbon nitride photocatalysts

The photocatalytic properties of semiconductors are related to their own structure. The energy band inside a semiconductor consists of two parts, the valence band (VB) and the conduction band (CB), the valence band is full of electrons while the conduction band contains no electrons, and the gap between the valence band and the conduction band is known as the forbidden bandwidth or bandgap (Eg). When sunlight hits the surface of a semiconductor and the energy is large enough (greater than the semiconductor bandwidth), the valence band of the semiconductor will be excited to produce photogenerated electrons, which will jump across the band gap to the conduction band, and at the same time there will be holes in the valence band, which is the electron-hole pairs (also referred to as photogenerated carriers). Photogenerated carriers can migrate to the surface of carbon nitride photocatalysts [18] and react with pollutants such as dyes, antibiotics, and CO_2 on the surface of the catalyst in an oxidative reaction, degrading them or reducing them with hydrogen ions on the surface of the catalyst to generate hydrogen. However, the uneven distribution of energy levels due to defects inside the semiconductor makes it easy for photogenerated electron-hole pairs to complex inside the semiconductor, releasing light or heat that is wasted. Therefore, inhibiting the compounding of photogenerated electron-hole pairs is one of the keys to the efficient utilization of photocatalytic technology. The potential positions of the valence and conduction bands of photocatalysts determine the strength of their reduction and oxidation ability. In general, the larger the potential of the valence band is, the stronger its oxidizing ability is. Correspondingly, the smaller the potential of the conduction band, the stronger the reduction ability. When the forbidden band width is small, only a small amount of excitation energy is needed to be excited. When a semiconductor has a strong redox capability, its forbidden band width is relatively large, and a high energy is required to excite the semiconductor.

Covalently linked hybrids consisting of cobalt porphyrin and carbon nitride reduce CO_2 to CO under visible light, and the photocatalytic activity of the hybrids is 13 times higher than that of the pristine carbon nitride, and twice as high as that of a heterojunction system in which the cobalt porphyrin is directly loaded on the carbon nitride. The enhanced photocatalytic performance is attributed to the addition of photogenerated electron transfer at the Co active site and the affinity of cobalt porphyrin for CO_2. In addition, carbon nitride nanosheets with oxygen and carbon co-doping show high selectivity of the photocatalytic system for CO2 reduction. This enhanced photocatalytic reduction performance can be attributed to the oxygen and carbon co-doping, which accelerates the photogenerated carrier generation and transfer and improves the visible light absorption. A study has demonstrated the significant aspect effect of Pd co-catalysts on CO_2 photoreduction from graphitic carbon nitride (g-C3N4). The surface atomic structure of the Pd co-catalysts can be precisely controlled by adjusting the number of {111} and {100} facets to regulate the interfacial charge carrier transfer, CO_2 adsorption and CH_3OH desorption processes.

II. B. Preparation of PCN

Polymerized carbon nitride (PCN) [19] is a synthetic organic polymer, of which three common preparation methods and the properties of PCN obtained are briefly described below:

(1) Thermal polymerization

Thermal polymerization refers to the heating of carbon and nitrogen-containing precursors to more than 50 ℃ to fully polymerize the PCN material, increasing the polymerization temperature will make the polymerization degree of PCN increase and reduce the content of hydrogen atoms, which can effectively reduce the accumulation of inactive electrons, which is more conducive to the enhancement of photocatalytic activity. The temperature of PCN prepared with thiourea as precursor is significantly lower than that of other precursors, because the presence of sulfur atoms promotes the polymerization reaction. Of course, the PCNs prepared with different precursors have large differences in morphology, texture and photoelectric properties.

(2) Solvent thermal method

Solvent thermal method refers to the use of organic solvents as a reaction medium in a closed reactor, heating and pressurizing the reaction system (or self-generated vapor pressure) to create a high-temperature, high-pressure reaction environment, so that the reaction solvent is in a critical or supercritical state, thereby enhancing the reaction activity. The solvent heat method has the advantages of operability and adjustability, and is a common method for material synthesis.

(3) Molten salt method

Molten salt method refers to the molten salt as a reaction medium, in the high temperature (usually higher than the melting point of the medium) conditions, the molten salt to form an ionic melt to play the role of a solvent, changing the kinetics of polymerization reaction, so as to obtain PCN materials.

II. C.Deficiencies of PCN photocatalysts

Although PCN has the advantages of high chemical stability and high thermal stability, there are still some problems to overcome if it is to be widely used. First, due to the high recombination probability of the photogenerated carriers in PCN polymer semiconductors, the photocatalytic performance is very limited although it is responsive to sunlight, which is one of the main reasons limiting its wide application. In order to improve the utilization efficiency of the photogenerated carriers and achieve an efficient photocatalytic process, the photogenerated electron-hole pairs must be effectively separated. In addition, the large forbidden bandwidth, which makes it underutilized for the visible part of sunlight, leads to the poor photocatalytic performance of PCN. Moreover, the specific surface area of PCN differs from the theoretically calculated value, resulting in few active sites, which is not conducive to the inter-interfacial reaction, and there is still much room for improvement.

II. D.Carbon nitride photocatalyst preparation

II. D. 1) Block PCN

Block PCN was prepared by pyrolyzing melamine in air atmosphere. The specific preparation method was as follows: weighing 3 g of melamine (MA) in a porcelain boat, mounting the lid and wrapping it with aluminum foil, placing it in a muffle furnace, and heating it from room temperature to 550 °C at a rate of 3 °C min^{-1} in an air atmosphere and keeping it at 550 °C for 4 h. After natural cooling down to room temperature, a yellow color was obtained, and the sample was fully ground and marked as BBPCN.

II. D. 2) General PCN

Ordinary PCN nanosheets were prepared by pyrolysis of melamine-cyanuric acid (CA) supramolecular polymerization intermediates at 550°C under air atmosphere. The specific preparation method was as follows: 1.51 g of melamine and 1.55 g of cyanuric acid were weighed and dissolved in a flask containing 40 mL of deionized water at 90 ℃ with vigorous stirring. When the melamine was completely dissolved, the melamine solution was poured into the cyanuric acid solution, and a large amount of white precipitate was produced at the moment of pouring. After continuing stirring in a 90℃ water bath for 40min, the white precipitate was filtered. It was washed sufficiently with deionized water, centrifuged and separated, and then dried in a vacuum oven at 60°C. The dried samples were fully ground, and subsequently, the precursor samples were placed in a porcelain boat and roasted in an air atmosphere at 550°C for 3 h to obtain fluffy light yellow samples labeled as NORPCN.

II. D. 3) Preparation of PCN nanosheets with enhanced visible light absorption

Visible light absorption-enhanced PCN nanosheets were prepared by pyrolysis of melamine-cyanuric acid supramolecular polymerization intermediates at 670 °C under argon atmosphere. The specific preparation method was as follows: the MCA precursor was placed in a porcelain boat, fitted with a lid and wrapped with aluminum foil, placed in a tube furnace, and evacuated by argon gas for 1h. After that, it was roasted in argon atmosphere at 670°C for 2h, and after it was naturally cooled down to room temperature, the sample obtained was labeled as VAEPCN.

II. D. 4) Copper-modified phosphorus-doped PCNs

The NORPCN sample prepared above was placed in 50 ml of deionized water solution and ultrasonically dispersed for 20 min, after which copper-modified phosphorus with different mass ratios was added to the well-dispersed NORPCN solution and stirred for 2 h. The resulting product was filtered, washed, dried, and naturally cooled to room temperature to obtain a light-yellow powder, which was noted as CMPDPCN.

III. Optical properties of carbon nitride photocatalysts and applications for carbon reduction in housing

III. A. Catalyst optical properties

Ultraviolet-visible diffuse reflectance spectroscopy (UV-Vis-DRS) is used to study the optical properties and energy band structure of materials. By measuring the degree of absorption of UV-visible light by a material, information such as absorption fringes, energy band gaps, and energy band structure of the material can be understood.

In this study, UV-vis diffuse reflectance spectroscopy (VV-DRS) tests were carried out on bulk PCN (BBPCN), normal PCN (NORPCN), copper-modified phosphorus doped PCN (CMPDPCN), and PCN with enhanced visible light absorption (VAEPCN) to evaluate the light absorption ability of the materials.

The UV-visible absorption spectra of several materials are shown in Fig. 1. The light absorption of both UV and visible parts of ordinary PCN, copper-modified phosphorus-doped PCN and PCN nanosheets with enhanced visible light absorption are enhanced compared to bulk PCN. The absorption edge of bulk PCN is around 500 nm, which has better absorption of all UV light and part of visible light, while the absorption edge of normal PCN extends to around 520 nm, and both UV and visible parts of the light absorption ability are enhanced. The UV and visible light absorption of copper-modified phosphorus-doped PCN and PCN with enhanced visible light absorption are further improved, especially the visible part has a very obvious improvement. It can be seen that the light utilization of copper-modified phosphorus-doped PCN is well improved.

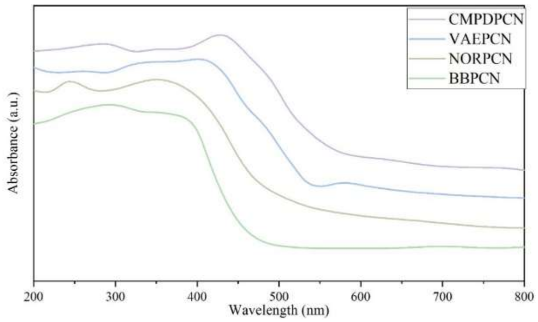

Figure 1: Ultraviolet - visible light absorption spectrum of several materials

The photogenerated electron-hole complexation and migration behaviors in bulk PCN, ordinary PCN, Cu-modified phosphorus-doped PCN and visible light absorption-enhanced PCN were investigated using photoluminescence spectra, and the photoluminescence spectra of different materials are shown in Figure 2. The excitation wavelengths of bulk PCN and normal PCN and visible light absorption-enhanced PCN are located at 400, 425, and 440 nm, respectively, while the excitation wavelength of Cu-modified phosphorus-doped PCN produces a drastic change and is located at 490 nm, which may be due to the change of the bandgap of PCN after carbon doping. In addition, the copper-modified phosphorus-doped PCN has the smallest photoluminescence intensity, which proves that the copper-modified phosphorus doping slows down the compounding degree of photogenerated electron holes more effectively, making more photogenerated electrons have the opportunity to participate in the catalytic reaction.

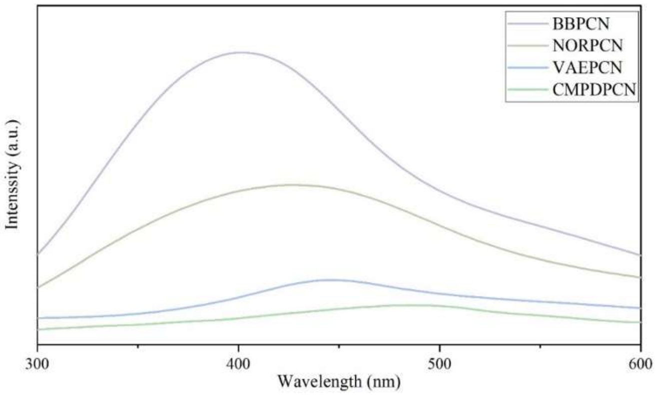

Figure 2: Photoluminescence spectra of different materials

III. B. Photocatalytic performance analysis

Photocatalytic reactions are generally carried out by generating free radicals, which can degrade organic pollutants or inorganic pollutants into harmless inorganic substances.This paper focuses on the catalytic degradation of $C0_2$ in housing construction by copper-modified phosphorus-doped carbon nitride catalysts.

In order to investigate the degradation ability of copper-modified phosphorus-doped carbon nitride catalysts for CO_2 degradation in housing construction, a self-assembled photocatalytic degradation device was used to test the photocatalytic degradation of porous cement concrete loaded with copper-modified phosphorus-doped carbon nitride catalysts, and PCN nanosheets with enhanced visible-light absorption were also selected for the comparison of the catalytic performance of the above prepared visible-light-absorbing PCN nanosheets.

The photocatalytic degradation rate was defined as the initial CO_2 concentration c_o minus the CO_2 concentration after 120 min c_{120} divided by the initial CO_2 concentration:

$$P = \frac{c_o - c_{120}}{c_0} \times 100\% \tag{1}$$

The photocatalytic degradation rate with time is given in Figure 3. The blank control in the figure is porous cement without carbon nitride photocatalyst, VAEPCN is porous concrete doped with visible light absorption enhanced PCN nanosheets, and CMPDPCN is porous concrete doped with copper-modified phosphorus doped carbon nitride catalyst. As can be seen from the figure, the copper-modified phosphorus-doped carbon nitride catalyst is far better than the visible light absorption-enhanced PCN nanosheets for CO_2 photocatalysis in housing construction, and under the irradiation of UV light, the CO_2 degradation rate of the copper-modified phosphorus-doped carbon nitride catalyst can reach 75.64% after 2h, while the CO_2 degradation rate of visible light absorption-enhanced PCN nanosheets is only 28.20%.

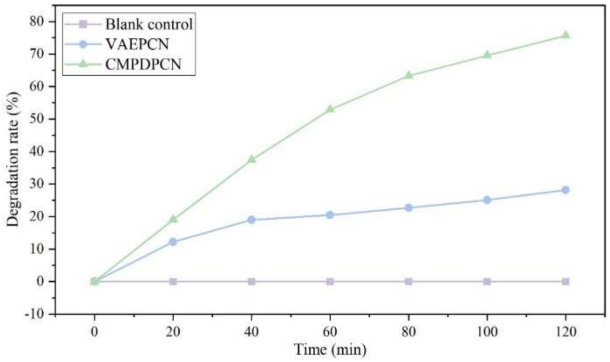

Figure 3: Photocatalytic degradation rate is at any time

Table 1 shows the variation of degradation rate of porous cement concrete loaded with carbon nitride catalyst with $g\text{-}C_3N_4$ doping for 2 h. All the porous cement concrete doped with $g\text{-}C_3N_4$ can effectively degrade CO_2 gas. The degradation efficiency of porous cement concrete doped with copper-modified phosphorus-doped carbon nitride catalysts was much greater than that of the samples doped with visible-light-absorption-enhanced PCN nanosheets at the same concentration of g-C_3N_4. This is due to the larger pores and greater surface area of g-C_3N_4 after doping modification. And the higher the doping amount, the higher the degradation rate, but the doping amount should not be too large. In this experiment, when the doping amount of photocatalyst reaches 7%, the CO_2 degradation rate of both VAEPCN and CMPDPCN reaches the maximum, which is 35.95% and 79.03%, respectively. Continuing to increase the photocatalyst will lead to a decrease in the degradation rate, so the optimal amount of photocatalyst incorporated into porous cement in housing construction is 7%.

Table 1: The degradation rate varies with the admixture of g-C_3N_4

g-C_3N_4 admixture (%)	VAEPCN	CMPDPCN	g-C_3N_4 admixture (%)	VAEPCN	CMPDPCN
0	0.00	0.00	6	32.09	67.90
1	9.82	17.10	7	35.95	79.03
2	14.67	32.34	8	29.66	69.36
3	19.99	48.76	9	27.24	61.61
4	24.34	52.90	10	25.06	52.90
5	28.45	59.90			

III. C. Evaluation of Carbon Reduction Rate of Rooftop PV Technology

In this section, carbon nitride catalysts are applied to photovoltaic-energy storage systems in housing construction, where the catalysts are utilized to drive the conversion of CO_2 to produce carbons that can be used as raw materials for construction and clean energy. In this paper, it is assumed that all of the building roof PV power generation is used by all types of energy-using systems in the building body, and it is assumed that 60% of the building roof area can be used for placing PV panels. The rated power of the PV panels is based on the technical parameters of a mature product from a leading manufacturer in the PV industry, and the equivalent rated power generation time is based on actual project data in a city.

The calculation results of the carbon dioxide reduction effect of roof PV for housing construction are shown in Table 2. As can be seen from the table, the building roof PV power generation and carbon dioxide emission reduction are enhanced with the increase of PV area, but the carbon dioxide emission reduction rate contributed by the application of roof PV decreases with the increase of building volume due to the overall energy consumption of the building rises rapidly with its volume at the same time. For the typical housing building model constructed in this paper, the blank control generates 26.17 MW-h of PV power when the area of roofing PV is 100 m2, which can offset 36.21% of its own carbon emissions. The incorporation of carbon nitride catalyst in the PV module can greatly increase the PV power generation and improve the CO2 emission reduction. In the case of PV module, after incorporating PCN nanosheets with enhanced visible light absorption, the power generation capacity was 30.44 MW-h in 100 m2 PV area, and the CO2 emission reduction rate was increased to 45.64%. In addition, when the carbon nitride catalyst was replaced with copper-modified phosphorus-doped carbon nitride catalyst, the power generation and emission reduction rate of PV were further enhanced to 38.75 MW-h and 53.84%, respectively. It can be seen that the copper-modified phosphorus-doped carbon-nitride catalyst based on PV-energy storage system in housing construction can effectively reduce CO2 emission and realize low-carbon construction.

Table 2: The results of the calculation of photovoltaic carbon reduction in roofing

Photovoltaic area (m^2)		100	250	500	1000
Blank control	Generating capacity (MW·h)	26.17	65.43	130.85	261.70
	Carbon dioxide emissions (t)	7.98	19.95	30.90	79.80
	Carbon dioxide reduction rate (%)	36.21	30.72	22.45	9.18
VAEPCN	Generating capacity (MW·h)	30.44	76.10	152.20	304.40
	Carbon dioxide emissions (t)	10.29	25.73	51.45	102.90
	Carbon dioxide reduction rate (%)	45.64	36.95	28.47	14.35
CMPDPCN	Generating capacity (MW·h)	38.75	96.88	193.75	387.50
	Carbon dioxide emissions (t)	12.71	31.78	63.55	127.10
	Carbon dioxide reduction rate (%)	53.84	41.95	32.75	20.96

IV. Conclusion

Copper-modified phosphorus-doped carbon nitride catalysts showed superior performance in photocatalytic CO_2 degradation with roof photovoltaic systems. In the porous cement concrete composite system, the catalyst doping ratio significantly affected the CO_2 degradation rate, and CMPDPCN showed the highest degradation rate of 79.03%

at 7% doping, which was 43.08 percentage points higher compared to VAEPCN. Further studies showed that doping above 8% led to a decrease in performance, indicating the existence of an optimal loading ratio. In the roof PV system, CMPDPCN embedding enhances the carbon reduction efficiency of the building model at all stages. Taking 250m² of PV area as an example, the CO_2 reduction rate of CMPDPCN is 41.95%, which is higher than the 30.72% of ordinary PV system, an improvement of 11.23 percentage points. This catalytic reduction of CO_2 to useful compounds combines both energy conversion and environmental treatment, and is suitable for synergistic systems incorporating building shells and energy modules. Overall, the catalytic strategy is highly efficient, sustainable and practical, providing an empirical basis and engineering path for the development of low-carbon building materials.

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