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Review

Emerging sustainable solutions for depollution: Geopolymers

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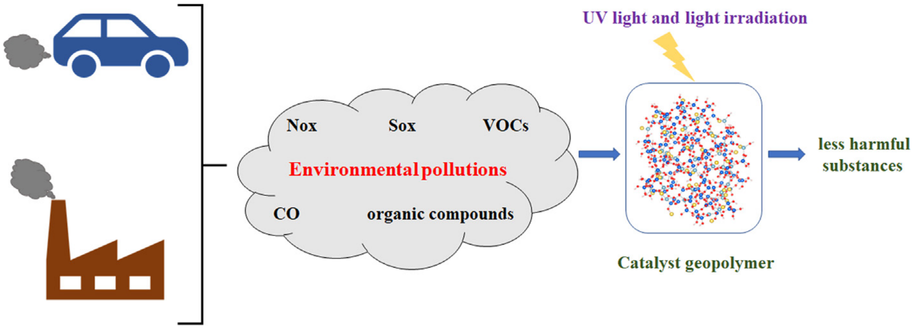
HIGHLIGHTS

- Catalyst geopolymers could utilize for the de-pollution of surrounding environment.
- Recent progress in substituting concrete & substrates with geopolymers is discussed.
- The review highlight studies used waste materials for catalyst geopolymer synthesis.
- Agricultural and industrial wastes have been identified as potential resources.
- Opportunities and Challenges remaining for a wider use are discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Air pollution and water pollution are critical issues because they negatively affect ecosystem and human health, as well as affecting buildings and industries. Utilizing photocatalysts that rely on light irradiation to decompose pollutants is a promising solution to this issue. Geopolymers, when used as a green substitute for cement and other engineered materials, can be synthesized using different agricultural and industrial wastes. These materials exhibit synergies in the depollution process and can be regarded as cost effective and novel in the context of removing organic pollution from water and air to protect and improve the environment. Geopolymer catalytic performance is closely linked to the composition, microstructure, and preparation method of the geopolymer. This review investigates the types of geopolymers and catalysts and their corresponding preparation methods and performances. The limitations of these products and future works are also detailed.

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1. Introduction

Specific concern about a sustainable environment has arisen among policymakers, researchers, industries, and the public as air pollution and water pollution have become serious problems. Pollutants such as a mix of nitrogen oxides (NO_x), sulphur oxide (SO_x), volatile organic compound (VOCs), carbon monoxide (CO), and other organic pollution are mainly due to automotive and other industries, and these pollutants result in acid rain, smog, etc. They not only are hazardous to our health but also can damage buildings and other infrastructure [1–3].

Urbanization renders buildings as some of the most important structures with regard to a sustainable environment. Cement is the main ingredient in concrete, which is the most used material in construction technology. It is highly attractive to researchers. To improve ambient air quality in buildings, the implementation of photocatalysts in construction technology has been explored since the late 1980s [4]. Photocatalysts can remove/convert pollutants to less harmful substances via UV light and light irradiation. Self-cleaning concrete, which contains photocatalysts, could reduce odours and air pollutants, improve indoor air quality, depollute water and eliminate diseases [5]. The application of cement containing photocatalysts for construction and environmental applications has not only been studied at the laboratory scale [4,6] but also been studied in the field in the form of paving elements [7] and coating materials in tunnels [8]. Many examples of self-cleaning concretes exist, including the Church 'Dives in Misericordia', Rome, Italy; Air France Building, Roissy-Charles de Gaulle Airport, France; Ciments Maroc Headquarters, Morocco; parking lane in Antwerp, Belgium; Castorweg Road in the Netherlands; a pavement area in Lysegrå Malmö, Sweden; and a few small structures in Germany comprising outdoor roof applications based on Heidelberg's product TioCem™ from the early 2000s [4].

Geopolymer, as a third generation cement, is regarded as an environmentally friendly substitute for Portland cement due to its very low carbon dioxide emission and low firing/ambient temperature. Different kinds of aluminosilicate materials—kaolinite; feldspar; agricultural, industrial, and mining wastes, such as fly ash (FA); ground granulated blast furnace slag (GGBS); silica fume (SF); palm oil fuel ash (POFA); and rice husk ash (RHA)—can be utilized as precursors to prepare geopolymers [9,10]. The effect of the chemical composition, mineralogical structure, fineness morphology, and glassy phase content of the precursors on the geopolymer development must also be considered [9,11–13]. The composition of geopolymers include a three-dimensional network of AlO_4 and SiO_4 tetrahedra, connected by oxygen corners with Si/Al ratios typically of 1:3, with the negative charge of the network being balanced by extra framework cations. Geopolymers have different chemical composition and ion exchange properties, which could form rings with cationic sites for bare cations. They are regarded as low-silica zeolites, in that they are mostly amorphous The microstructure of the geopolymer consists of an innate nanomaterial with a xerogel-like structure, made up of highly fused nanoparticles (diameter of 10–30 nm), with the core of the nanoparticles most likely being zeolitic [14–16]. The functional potential of geopolymer, which is related to its innate nanostructure, paves the way for a wide range of advanced applications, including acting as drug delivery agents, catalysts, adsorbents of cations from wastewater, and precursors of advanced ceramics [17]. The modification of Na/K/Ca-based geopolymer by other transitional metal ions using ion exchange procedures and controlling the geopolymer nanostructures to realize high surface area and porosity could also pave the way for the synthesis of green and effective catalysts [14]. Geopolymer catalysts result from a simple preparation method because the natural raw material is ubiquitous and inexpensive relative to other advanced catalysts [17].

2. Overview of geopolymer types utilized in catalyst geopolymers preparation

The amorphous zeolitic-type structure of geopolymers consists of three-dimensional tetrahedral silicate and aluminate, bonded via common oxygen atoms with ion exchange properties. The selection of raw materials and processing conditions is well known to determine the characteristic of geopolymers, which results in geopolymers with very different properties. This allows for the preparation of bespoke geopolymers via the manipulation of raw materials, compositions, and processing conditions. For example, fly ash-based geopolymers, which exhibit increased durability and strength, are important in construction relative to metakaolin-based geopolymers, which are more common in adhesives, coatings, and hydroceramics. The growth process, which dictates the structure of the geopolymer, also influences its subsequent microstructure and pore distribution [18]. Researchers developed a porous geopolymer-employing foaming agent to prepare geopolymers [19].

The source of aluminosilicate for geopolymer preparation can be natural minerals, such as metakaolin, laterite, and illite smectite clays, or industrial and agricultural waste materials, such as FA, sludge, and rice husk ash (RHA) [10,20]. The selection of proper precursors, while considering the sufficient amount of reactive silica and alumina for geopolymer preparation, is crucial [19]. Koleżyński et al. [21] performed theoretical studies on geopolymer-SIC structures. Fig. 1 shows the structural model of the geopolymer.

Fig. 2 shows the simplified reaction mechanism for geopolymerisation according to Duxson et al. [18].

Geopolymers used as catalysts are detailed below:

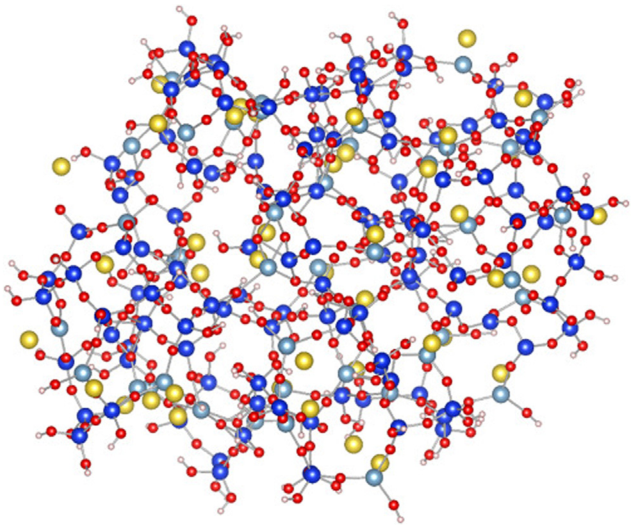

Fig. 1. Structural model for geopolymers (cluster ~ 800 atoms, Si: Al = 2.81). Si, Al, O, Na and H atoms are depicted in blue, silver, red, yellow and beige, respectively [21].(For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

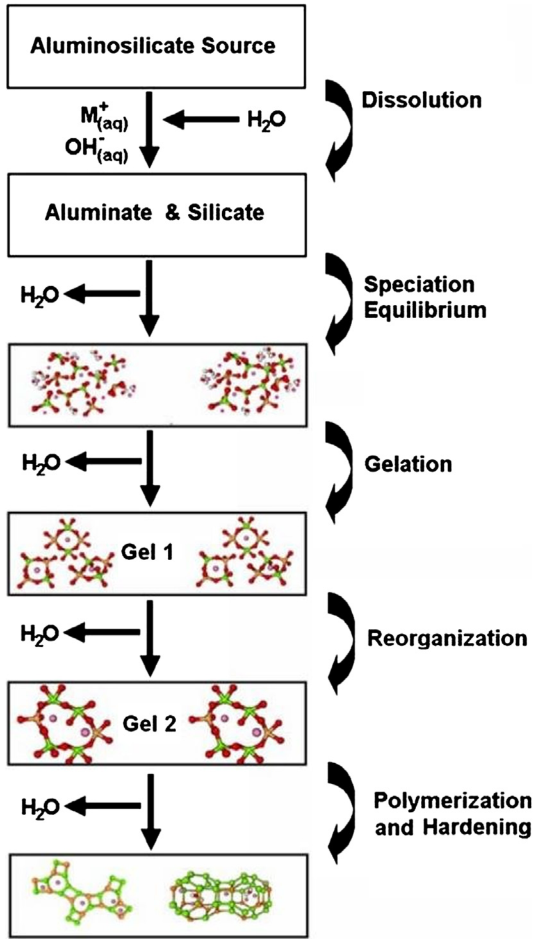

Fig. 2. Conceptual model for geopolymerisation [18].

2.1. Metakaolin-based geopolymers

Duxon et al. [22] determined the link between geopolymer composition, microstructure, and mechanical properties in alkali-activated metakaolin-based geopolymers. They confirmed that the microstructure of these geopolymers is correlated with the corresponding Si/Al ratio. When the ratio of Si/Al is ≤ 1.40, the geopolymer is highly porous, but at Si/Al ≥ 1.65, it is largely homogeneous. Geopolymers with an Si:Al ratio of 1.9 exhibit maximum strength, and increasing the ratio to beyond 1.9 results in decreased strength, with the untreated materials acting as defects within the structure. Geopolymers can be tailored to specific applications, such as for hydrogen storage [23] and as oxygen carriers [24]. Metakaolin can be mixed with different waste materials, such as rice husk, slag, etc., to synthesize geopolymers for use as an adsorbent [25,26].

2.2. Waste material-based geopolymers

Different industrial and agricultural waste materials with aluminosilicate can be used as the raw materials to prepare geopolymers. Their chemical composition, structure, morphology, and glassy phase content are critical to the geopolymer properties [9].

2.2.1. Fly ash-based geopolymers

FA is a biowaste material produced from coal combustion in power plants. It is widely used as a raw material for geopolymer synthesis [27]. It can be mixed with other wastes, such as rice husk ash, fibre, and slag for the preparation of geopolymers [28–30]. Fly ash-based geopolymers not only exhibit increased mechanical and fire resistance but also exhibit excellent encapsulation and solidification capabilities vis-à-vis various toxic and hazardous wastes [31–35]. They can also be used as adsorbents and catalysts [36–39].

2.2.2. Slag-based geopolymers

Slag (GGBS), which is a calcium aluminosilicate waste of iron ore manufacturing (production of pig iron) has been used by itself and mixed with other aluminosilicate materials to synthesize geopolymers [10,20,26,40–43].

2.3. Open cell geopolymer foams

Many researchers have analysed the preparation of porous component-based geopolymers employing the same approach as the cement industry [44,45]. Different methods such as the foaming gaseous method [46,47], solidification procedures [48,49], freeze-casting method [50], replica technique [51,52], gel-casting [53,54], frothing [55], saponification [56], metal powder as foaming agent [57], and a mix of multiple techniques [58,59] have been used to prepare porous geopolymers. However, many of these methods are capable of producing only closed cells, which decrease the permeability of the geopolymers to liquid and gases. In the case of catalyst support with high permeability, the surface area, chemical resistance, and mechanical strength are critical factors [58,59]. These factors are crucial in the development of open-cell geopolymers synthesized via techniques such as gel-casting using surfactant [58,59], saponification/peroxide/gel-casting combination [58,59], mechanical frothing using surfactant or foaming agent [55,56,60], replica technique [61], and polyurethane sponge impregnation [51,52]. The common methods for preparation of porous materials are depicted in Fig. 3.

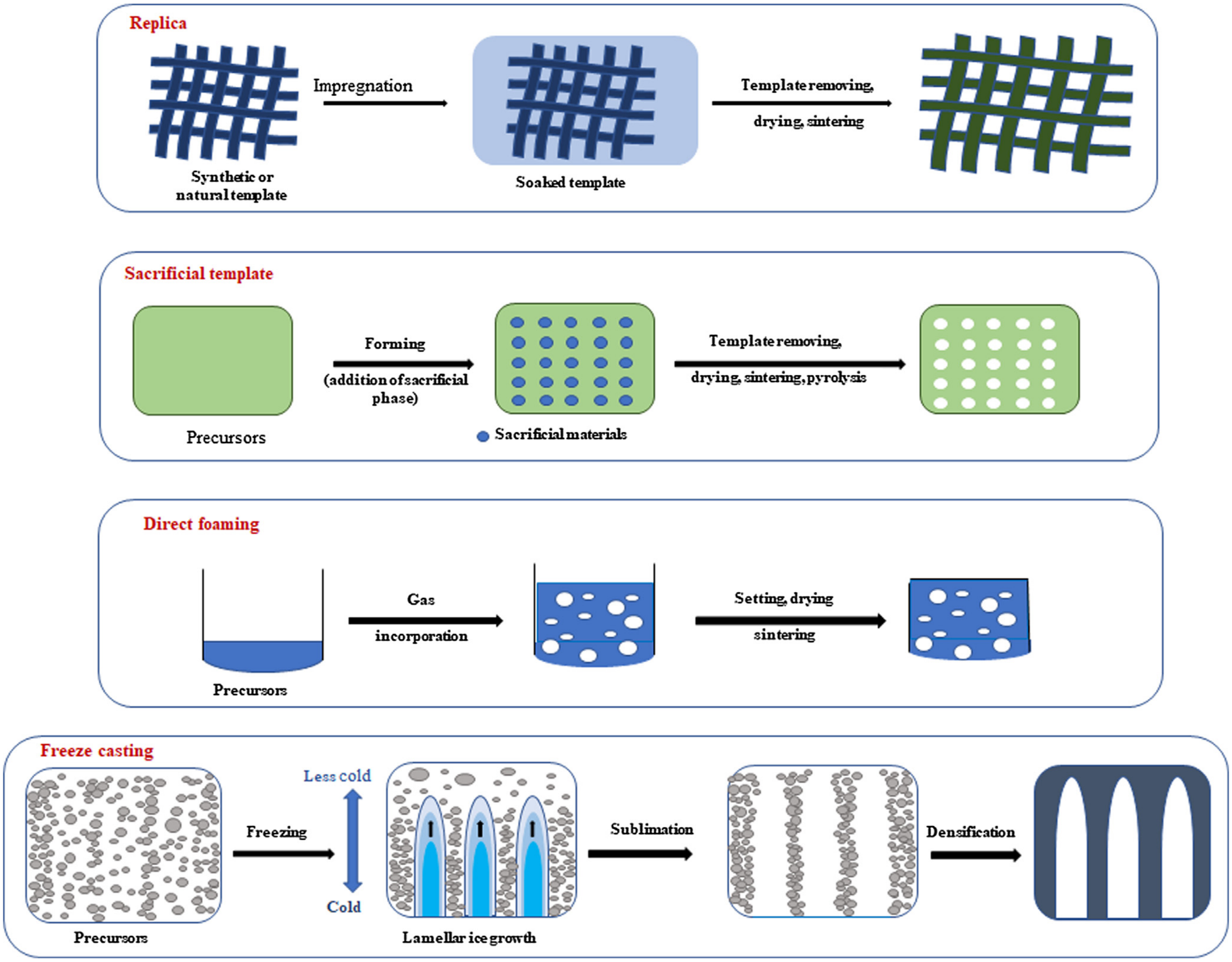

Fig. 3. Some common methods for porous material preparation [114,115].

3. Addition of nanomaterials to geopolymers

The addition of nanomaterials, such as nano-SiO_2, nano-clay, graphene, and carbon nanotubes can modify concrete's geopolymer physico-mechanical properties, such as strength [9,62,63]. However, to realize optimal performance, we need to be aware of the correlation between the interfacing transition zone, microstructure, setting behaviour, mix design characteristics, and the mechanical properties [9]. The uniform dispersion of nanomaterials in geopolymers is one of the most critical aspects of processing, as it dictates the subsequent properties such as physio-mechanical and catalytic properties which are very important for their catalytic performance especially in construction. Uniformity can be realized via multiple techniques [64]. Sumesh et al. [9] reviewed the influence of many nanomaterials on concrete and geopolymer properties, alongside the health and safety issues linked to the utilization of nanomaterials. However, the addition of nanomaterials to geopolymer for catalyst application is a new research approach.

4. Overview of catalyst preparation

The purpose of utilizing geopolymers as the catalyst is to remove contaminants and impurities. In some cases, modifying the geopolymer and creating active sites in its structure could render it a catalyst [17,65], but in most cases, the addition of semiconductors such as metal oxides, metal, alloys and sulphides to a geopolymer structure in the form of coatings results in the catalyst.

To increase catalytic performance in the geopolymers, the catalyst must be homogeneously dispersed and be easily accessed by the pollutants via the geopolymer, which can be realized by controlling the pore size and surface area of the geopolymer. However, the influence of the geopolymer (matrix) chemistry and surface charges must be carefully considered [4]. Fig. 4 shows the common semiconductor valance band (VB) and conduction band (CB) that can be used to instigate the desired chemical reactions [66] to prepare catalytic geopolymers.

The stability of the catalyst and the variation in band gaps and redox coupling potentials must also be accounted for in the quest to improve its performance [67]. The surface chemistry/electrokinetic properties significantly influence the dispersion/agglomeration in the catalyst [4].

4.1. Heterogeneous solid acid catalysts

Interest in solid/base catalysts is increasing due to their ease of preparation, reuse, and regeneration, limited disposal issues, and non-corrosiveness. Zeolite is an interesting material due to its

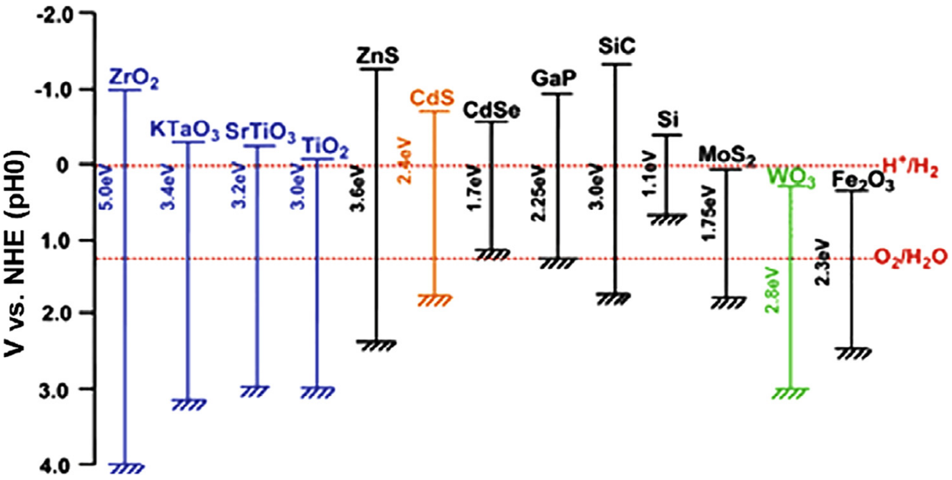

Fig. 4. Bandgap energy, VB and CB positions of several common semiconductors [116].

widespread species and structures that can be used to produce bespoke geopolymers for many applications. Their microporous structures, high concentrations of active acid (Lewis and Brønsted) sites, and affinities for many cations render them ideal candidates for catalyst substrates and absorbents [68,69]. Zeolite with Y-type, mordenite, and beta structures are well known for application as solid acid catalysts and are capable of undergoing ion exchange. CsNaX and CsNaY zeolites can be used as solid base catalysts [69–72].

Aluminium removal by thermal and acid treatments is commonly used to prepare secondary mesopores or remove the extra framework aluminium from the zeolite's structure [73,74]. Desilication via base treatment could be used to prepare intracrystalline mesoporous zeolites [75–77]. Some researchers used both dealumination and desilication as sequential treatments to prepare silanol groups in zeolites so that they could determine the respective catalytic performances [78–80].

Geopolymers are similar to amorphous low-silica zeolites and are excellent substitutes in multiple zeolite applications due to their cost effectiveness and environmentally friendly nature. They are also similar in terms of surface area, pore size and volume, concentration of active sites, good affinity for reactants, and stability [81], most of which are inherent in geopolymers via its synthesis [17].

Alzeer et al. [17] developed kaolin-based geopolymers with active sites in their structures using the ion exchange process with NH^{4+}, followed by thermal treatment. They increased the catalytic performance of geopolymers using sequential treatment to maximize the concentration of the silanol groups. The surface area and pore dimensions were controlled via the starting composition and post-synthesis treatments.

The catalytic performance of the prepared solid acid geopolymer catalyst has been studied via the liquid-phase Beckmann rearrangement of cyclohexanone oxime to ε-caprolactam. In another attempt, geopolymer acid catalysts were evaluated in terms of Friedel-Crafts alkylation reactions [82].

4.2. Preparation and modification process

Due to the structural similarities between zeolites and geopolymers, both can be used to prepare catalysts. The tetrahedra $[SiO_4]^{4-}$ and $[AlO_4]^{5-}$ are randomly linked by oxygen in a geopolymer three-dimensional framework, balanced by extra framework cations of Na^+, K^+, and Ca^{2+}. These cations could be replaced with other transition metal cations and rare metals via processes such as ion exchange, impregnation and mixing with precursor in geopolymer preparation, coupled with different modifications such as surfactant and thermal treatments. These approaches could create new active centres for catalytic activity [40,43,83–87].

The immobilization of the catalyst on the inner and outer surfaces of the substrate could improve the heterogeneous catalyst performance. Post-synthesis treatments, such as thermal, surfactant, dealumination, and desilication treatments, introduce additional mesoporous, macroporous, and active sites to the geopolymer, thereby improving the performance of the catalysts [1,88,89].

Using surfactants such as a large quaternary ammonium ions could dictate the porosity, surface area, size and shape of geopolymers and clay for the specific applications [1,2,90–94].

Some researchers improved the photocatalytic performance using waste glass in cement [95–97]. They claimed that waste glass could enable light to penetrate deeper into the photocatalyst, which increases its activity and performance.

Large surface area/porous structures enhance catalytic performance. However, the size of the pollutants needs to match the size of the pores for the catalysts to be effective [98].

The unique electronic structure of carbon allows it to form different structures based on the hybridization of the orbitals and dimensions. Examples include carbon materials such as graphene, carbon nanotubes (CNT), carbon fibres, glassy carbon, activated carbon, fullerenes, fullerides, carbon black, and diamond [99–101]. Different types of nanostructured carbon materials have been employed as substrates for catalysts due to their potentials in bespoke treatments to produce specific shapes and increase the porosity and thermo-physical stability, all of which influence catalytic performance [101–106].

Loading and coupling of semiconductors with graphene, which has a π-conjugation system and high electron mobility, could improve the separation efficiency of photogenerated electron-hole pairs, which enhances the catalytic performance of the catalysts due to the excellent charge-trapping ability of the graphene [107,108].

Taking into account these facts, Zhang et al. [109] prepared a geopolymer by coupling graphene with CuO for dye degradation. Fig. 5 shows the mechanism of the photocatalytic reaction in the resulting geopolymer [110]. The graphene/CuO geopolymer exhibited promising performance in the context of dye degradation in wastewater.

Table 1 shows the research that has been conducted on the preparation of geopolymer catalysts.

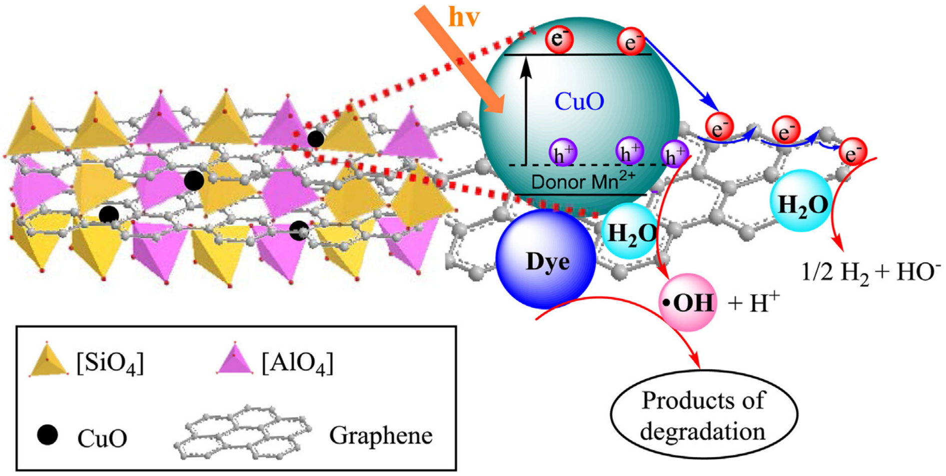

Fig. 5. Mechanism of photocatalytic reaction in geopolymers containing graphene [110].

Table 1
List of the various research on geopolymers as catalysts.

References	Catalyst	Geopolymer type	Reaction	Catalyst Preparation method	Results
[1,117]	Nanocubic Cu_2O and nanospherical Cu_2O/TiO_2	Metakaolinite-based geopolymer; modified metakaolinite-based geopolymer	Methylene blue (MB) dye degradation	Ion-exchange; modification by surfactant	Removing >98% of MB dye under UV irradiation
[14]	Ca^{2+}	Modified metakaolinite-based geopolymer	Conversion of soybean oil to a biodiesel	Ion exchange process	~100% conversion
[118]	TiO_2	Metakaolin based geopolymer	Degradation of 2-butanone	Ion exchange	More efficient
[119]	Chemical compounds in fly ash	Fly ash-based geopolymers	Friedel-Crafts benzylation reactions of benzene		Excellent catalytic reactivity
[17]	The active sites were generated within the structure of the geopolymer	Metakaolin based geopolymer	Liquid-phase Beckmann rearrangement reaction	Ion-exchange with NH^{4+}followed by thermal treatment	Excellent catalytic conversion of the oxime
[120]	Silver	Nanosilica modified geopolymer mortar	Antibacterial	Adsorption	99% mortality for gram-positive and gram-negative bacteria
[121]	TiO_2	Metakaolin and fly ash geopolymers and they foamed hybrid with siloxane	Nitric oxide degradation	Mixing with geopolymer suspension	Different photocatalytic performance depends on the type of binder and the curing process
[39]	TiO_2 and Fe_2O_3 inside structure of fly ash based geopolymer	Coal fly ash-based geopolymer	Methylene blue (MB) dye degradation	Geopolymer preparation	92.79% under UV irradiation
[43]	Ni^{+}	Steel slag-based geopolymer	Methylene blue (MB) dye degradation	Ion exchange	94.39% under UV irradiation
[40]	Fe, Co, Cu metal ions and Pt	Metakaolin-based Na-geopolymer and metakaolin-slag based K, Ca-geopolymer	Reduction of nitrogen oxides by ammonia and the oxidation of decane by oxygen	Ion exchange reactions	
[109]	Mn^{2+} and CuO	Graphene bottom ash-based geopolymer	Sky blue 5B dye degradation and water-splitting	Ion exchange, co-impregnation and calcinations	100% dye degradation for Mn^{2+}-15CuO/GBAG
[122]	CaWO4	Blast furnace slag-based geopolymer	Water splitting	Impregnation and calcination	Significant improvement
[123]	In_2O_3 and NiO	Fly ash-based geopolymer	Water splitting		H_2 evolution of 3180 μmol/g under the irradiation of solar simulation source
[110]	Graphene	Blast furnace slag-based geopolymer	Methyl violet dye degradation	Geopolymer preparation	91.16% dye degradation
[124]	TiO_2	Metakaolinite-based geopolymer	Butane degradation	Mixing with geopolymer suspension	Photocatalytic activities correlate with the values of their surface area
[125]	TiO_2	Metakaolin-based geopolymer	Nleaching of methylene blue (MB)	Ion exchange, treatment with NH_4Cl	
[126]	Ni	Metakaolin-based geopolymer	Ethanol to syngas reforming reaction	Incipient wet impregnation	84 mol% of syngas was produced
[127]	CdS	Chemosynthetic geopolymer sphere	Methyl orange degradation	Photochemical growth method	92.57% for methyl orange degradation
[128]	TiO_2	Metakaolin and fly ash geopolymers	Methylene blue (MB) dye degradation	Sol-gel dip coating process modified by incorporation of Polyvinylpyrrolidone	Good photocatalytic efficiency
[129]	Fe^{+2} spinels	Kaolin based open-celled geopolymer	Antibiotics degradation	Polyurethane sponge impregnation	

5. Suggestions and future work

Geopolymers can be tailored to specific applications using different precursors and preparation conditions according to properties such as the pore size, pore distribution, surface area, shape, morphology, orientation, durability, strength, surface pH, dispersion, size, and band gap. These properties eventually dictate the performance of the resulting catalysts.

Different modifications such as utilizing surfactants and post-synthetic treatment, which influence the catalytic performance of geopolymer-based catalysts (heterogeneous solid acids or geopolymers mixed with semiconductors), require a more in-depth investigation [82].

Developing open cell geopolymers with 3D porous architectures without high temperature treatments which could be modified via alkali-bonding and doping [111], could represent an interesting area of research.

Replacing concrete with catalytic geopolymers in buildings could help mediate pollution, which is economically and environmentally beneficial, especially because the entire process is activated by light (solar), which eliminates the need for an external power source. However, the possibility of human exposure to nanoparticles via inhalation is a health concern and requires further investigation [9].

The use of different varieties of carbon, such as graphene, activated carbon, and carbon nanotubes, as a substrate are promising in the context of geopolymer-based catalysts [110,112,113]. The low-temperature preparation of geopolymers has increased the possibility of using multiple types of carbon, which previously was impossible due to the low decomposition temperature of carbon.

6. Conclusions

Geopolymer, as a future engineering material, is continuously being developed due to its technical and environmental advantages. The potential to control its properties by tailoring key parameters such as surface texture, chemistry, and porosity could also benefit from low-cost preparation methods and the utilization of industrial waste by-products and biomass ash as precursors, all of which makes the study of geopolymers a very interesting research area. Recent progress in substituting concrete and substrates with geopolymers has paved the way for research on the use of geopolymer catalysts for depollution of the surrounding environment.

Replacing concrete with catalyst geopolymers in buildings could also significantly improve human health.

Despite the present work on catalyst geopolymers, much work remains. In addition to the utilization of the different carbon forms in geopolymer—open-cell geopolymers, solid acids, doping, and treatments, the safety and standardization of geopolymer catalysts must be verified prior to their commercialization. Future research could provide the appropriate knowledge to establish a pathway for developing engineered geopolymers that are safe and inexpensive for specific applications.

Conflict of interest

None.

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