

Original Research Article

Granulated Biochar–MMT–Zn/Al LDH Composite as a Durable Adsorbent for Column Applications

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Abstract

Granular adsorbents are essential for fixed-bed column systems because they reduce the pressure drop and minimize the material loss commonly associated with powdered adsorbents. However, transforming powdered materials into mechanically stable granules without compromising their structural integrity remains a significant challenge. In this study, a granulated composite consisting of biochar, montmorillonite (MMT), and Zn/Al layered double hydroxide (LDH) was developed as a potential adsorbent for column application. Biochar derived from *Avicennia marina* biomass was combined with MMT and Zn/Al LDH to produce a composite material, which was subsequently granulated using bentonite, tapioca starch, and polyvinyl alcohol (PVA) as binding agents. The resulting granules (0.5–1.5 cm in diameter) were evaluated for mechanical durability under static water immersion and dynamic water flow conditions. The results indicate that the binder composition strongly influences granule stability. Granules prepared without PVA rapidly disintegrated in aqueous environments, whereas the inclusion of PVA significantly improved structural durability. The optimized formulation exhibited durability of up to 8208 min under static conditions and maintained its structural integrity under continuous water flow. These results demonstrate the potential of Biochar–MMT–Zn/Al LDH granules as durable adsorbents for fixed-bed column water treatment systems

Keywords: Adsorbent; composite materials; environmental remediation; wastewater treatment

1. Introduction

Adsorption is widely recognized as an effective and flexible method for removing contaminants from water because of its operational simplicity, high efficiency, and adaptability to various pollutants (Naswir et al., 2021; Wibowo et al., 2022; Wibowo, et al., 2025). A wide range of adsorbent materials have been developed, including activated carbon (Budihardjo et al., 2021), biochar (Wibowo et al., 2022), clay minerals (Wibowo et al., 2025), montmorillonite (MMT) (Taher et al., 2023), and its modification using layered double hydroxides (LDHs) (Taher et al., 2024), iron (Liu et al., 202), and other modified materials (Cheng et al., 2021). Among these materials, biochar-based composites have attracted considerable attention because of their high surface area, abundant functional groups, and relatively low production cost derived from biomass waste (Wibowo et al., 2023). Furthermore, combining biochar with mineral materials such as MMT and LDH can significantly enhance the adsorption capacity through synergistic physicochemical interactions (Viglašová et al., 2018).

MMT is a layered aluminosilicate clay mineral with a high cation exchange capacity and large specific surface area, making it a suitable support material for composite adsorbents (Taher et al., 2023). LDH are a class of anionic clays with a unique lamellar structure capable of exchanging interlayer anions, which enables the effective adsorption of various pollutants, including heavy metals, phosphates, and organic contaminants (Kameliya et al., 2023; Tiwari and Syväjärvi, 2014). Therefore, the integration of biochar, MMT, and LDHs materials into a single composite system can create multifunctional adsorbents with improved adsorption performance and structural stability. Despite the increasing number of studies on biochar-based composite adsorbents, most research has primarily focused on adsorption performance under batch conditions (Wibowo et al., 2023; Rianjanu et al., 2024; Wibowo et al., 2025), with limited attention given to the mechanical durability and hydraulic stability required for practical applications in columns. In real water treatment systems, adsorbents must withstand continuous water flow, mechanical stress, and prolonged contact with aqueous environments without undergoing disintegration. Although powdered materials often exhibit high adsorption capacities, they are unsuitable for fixed-bed column operations because they can cause excessive pressure drops, clogging, and material loss. Consequently, the development of mechanically stable granular adsorbents is a critical challenge for bridging the gap between laboratory-scale adsorption studies and practical water treatment applications (Shanmugam, 2017).

The novelty of this study lies in the integration of biochar, MMT, and Zn/Al LDHs into a granulated composite system, combined with a synergistic binder network consisting of bentonite, tapioca starch, and polyvinyl alcohol. Unlike previous studies that mainly emphasize adsorption capacity, this study specifically investigated the mechanical durability and hydraulic stability of the granules under both static immersion and dynamic water-flow conditions. Understanding the role of binder composition in maintaining structural integrity is essential for designing adsorbents that remain stable during continuous operations. Therefore, this study not only develops a multifunctional composite adsorbent but also addresses the critical issue of structural durability, which is essential for the practical implementation of biochar-based adsorbents in fixed-bed column water treatment systems.

Various binders have been explored to improve the structural stability of granular adsorbents, including clay minerals (e.g., bentonite and kaolinite), starch-based polymers, and synthetic binders such as polyvinyl alcohol (PVA), which enhance particle cohesion and mechanical strength during granulation or pelletization (Aghaei et al., 2022; Yoosuk et al., 2017). These binders can enhance particle cohesion and durability in aqueous environments while maintaining the functional properties of adsorbent materials. However, selecting an appropriate binder composition remains a challenge, as excessive binder content may block the adsorption sites or reduce the mass transfer efficiency within the granules. In recent years, research on biochar-based composite adsorbents has increased significantly, particularly in the development of hybrid materials that combine biochar with clay minerals and LDH (Wibowo et al., 2025). However, most studies have primarily focused on adsorption performance, while limited attention has been given to the structural

durability of granular composites designed for use in columns. Mechanical resistance and water stability are critical factors that determine the practical applicability of granular adsorbents in continuous flow systems.

Therefore, this study aimed to develop a granulated composite consisting of biochar, MMT, and Zn/Al LDHs as a potential adsorbent for column applications. Biochar derived from *Avicennia marina* biomass was combined with MMT and Zn/Al LDH to form a composite material, which was subsequently granulated using a mixture of bentonite, tapioca starch, and polyvinyl alcohol as binding agents. The combination of bentonite, starch, and PVA was selected to create a synergistic binder network in which the clay minerals provide structural reinforcement, starch facilitates granule formation, and PVA forms a polymeric matrix that enhances water resistance and mechanical durability. The mechanical durability of the resulting granules was evaluated under static water immersion and dynamic water-flow conditions to assess their structural stability in aqueous environments. Additionally, a literature-based analysis was conducted to examine current research trends related to biochar composite adsorbents. The findings of this study provide insights into the development of mechanically stable granular adsorbents suitable for fixed-bed column applications in water treatment.

2. Materials and Method

2.1 Materials

Biochar was produced from *Avicennia marina* biomass, locally known as Bakau Api-Api, collected from the Petengoran Mangrove Forest, Pesawaran District, Lampung Province, Indonesia (5°34'06" S, 105°14'24" E). MMT clay was obtained from a local commercial supplier and used as is. Zinc–aluminum LDH (Zn/Al LDH) was synthesized following the procedure described by Wibowo et al. (2025). Several binding agents were used during the granulation process to improve the particle cohesion and mechanical stability. Bentonite clay, tapioca starch, and polyvinyl alcohol (PVA) were selected as binders because of their ability to enhance the structural durability of granular materials in aqueous environments (Bertolini et al., 2022). All chemicals and materials used in this study were of non-analytical grade and used without further purification. Deionized water was used in all experiments.

2.2 Preparation of Materials

The collected biomass was washed with distilled water to remove adhering impurities and then dried at room temperature. A pretreatment step was performed using torrefaction at 180 °C for 2 h in a Memmert oven to reduce the moisture content and remove volatile impurities from the biomass (Azzaz et al., 2022; Ivanovski et al., 2022). After pretreatment, the material was pyrolyzed in a Nabertherm muffle furnace at 300 °C for 6 h under limited oxygen conditions to produce biochar. The resulting biochar was cooled to room temperature and ground into a fine powder. The powdered biochar was then sieved using a 200 mesh shaking sieve to obtain a uniform particle size suitable for the preparation of the composite. The composite material was prepared by mixing biochar, MMT, and Zn/Al LDH according to the formulation described by Wibowo et al. (2025). The powdered components were homogenized to ensure a uniform distribution of mineral phases within the biochar matrix. The resulting composite powder was used as the base material for subsequent experiments.

2.3 Preparation of Granular Materials

The powdered composite was converted into a granular form to improve its mechanical stability and suitability for column adsorption systems, as granular adsorbents exhibit better mechanical strength and operational performance in fixed-bed applications (ThiMinh et al., 2016). The granulation process was performed using a mixture of binders consisting of bentonite, tapioca starch, and polyvinyl alcohol (PVA). These binders were selected to enhance particle cohesion and structural durability without significantly affecting the physicochemical properties of the composite material (Bertolini et al., 2022; Denduyver et al.,

2024). The powdered composite and binder materials were mixed according to predetermined mass ratios, followed by the addition of a small amount of distilled water to form a homogeneous paste. The paste was then manually shaped into spherical granules with diameters of approximately 0.5, 1.0, and 1.5 cm using a manual pelletizing technique. The formed granules were air-dried for 6 h under ambient conditions and subsequently oven-dried at 105 °C for 12 h to remove residual moisture and stabilize the granular structure. The dried granules were stored in sealed containers before durability testing.

2.4 Mechanical Testing in Water and Water-Flow

The mechanical durability of the Biochar–MMT–Zn/Al LDH granules was evaluated under both static and dynamic aqueous conditions to assess their suitability for column applications. Granules with diameters of 0.5–1.5 cm were immersed in 500 mL of deionized water at room temperature (approximately 29 °C) for 24 h without agitation. After immersion, the granules were carefully removed from the water, gently dried with tissue paper, and visually inspected for structural damage, such as cracking, surface erosion, or disintegration. The durability of the granules was evaluated based on the percentage of mass loss, using the following equation:

$$\text{Weight loss (\%)} = \frac{W_0 - W_t}{W_0} \times 100$$

where W_0 and W_t are the initial and final dry weights of the granules before and after immersion, respectively.

The resistance of the granules under dynamic conditions was evaluated using a vertical acrylic column with a height of 30 cm and an internal diameter of 2.5 cm. The column was packed with a single layer of granules and connected to a peristaltic pump. Deionized water was continuously pumped through the column at controlled flow rates for 48 h. During the experiment, the structural integrity of the granules was periodically monitored to observe potential deformations, cracks, or fragmentation. After the flow experiment, the granules were removed from the column, dried at 105 °C for 12 h, and weighed again to determine the extent of structural degradation. These tests provided an assessment of the mechanical stability and water resistance of the composite granules under conditions simulating column adsorption operations.

2.5 Literature Review and Bibliometric Analysis

A literature review was conducted to evaluate the current research trends related to biochar-based composite adsorbents containing MMT and LDH. A literature review was conducted following the methodology described in a previous study (Wibowo et al., 2026). Scientific publications published between 2010 and 2025 were retrieved from the Scopus database on November 10, 2025. The search keywords included combinations of “biochar composite,” “montmorillonite,” “layered double hydroxide,” “granular adsorbent,” and “column adsorption.” The inclusion criteria comprised studies that (i) reported the synthesis or modification of biochar-based composites containing MMT or LDH, (ii) investigated the physicochemical or mechanical properties of the composites, or (iii) evaluated the adsorption performance under batch or column conditions. Bibliometric analysis was performed using VOSviewer software (version 1.6.20) to analyze the keyword co-occurrence networks and identify the dominant research themes. The visualization results were used to examine the evolution of research trends and position the present study.

3. Result and Discussion

3.1 Effect of Binder Composition on Granule Durability

The durability of the Biochar–MMT–Zn/Al LDH granules was strongly influenced by the binder composition used during granulation. The type and proportion of binders play crucial roles in determining the particle cohesion, mechanical strength, and structural stability of granulated materials, thereby affecting their

resistance to mechanical stress during handling and operation (Alsaqoor et al., 2022; Bertolini et al., 2022; Denduyver et al., 2024). As summarized in Table 1, the granules prepared using only bentonite as a binder exhibited extremely low durability values under aqueous conditions. For example, compositions with Bio–MMT–LDH: bentonite ratios of 1:2.34 and 1:1.5 showed durability values of only 0.58 min and 0.52 min, respectively. This rapid disintegration indicates that bentonite alone could not maintain sufficient particle cohesion when exposed to water. Bentonite is a swelling clay mineral dominated by MMT layers that readily absorb water molecules within its interlayer structure (Clegg et al., 2014). When hydrated, clay layers expand and generate internal stress within the granule matrix, which weakens interparticle bonding and promotes structural fragmentation (Clegg et al., 2014; Dai et al., 2020). Consequently, the granules lose their mechanical integrity and rapidly disintegrate in aqueous environments.

Table 1. Durability performance of Biochar–MMT–Zn/Al LDH granules prepared with different binder compositions

Bio (g)	Bnt (g)	PVA (g)	Bio : Bnt : PVA ratio	Water (mL)	Durability			
					Init. Dur (min)	8 mL/min (min)	16 mL/min (min)	32 mL/min (min)
0.3	0.7	-	1 : 2.34	1	0.58	0.77	0.66	0.67
0.4	0.6	-	1 : 1.5	1	0.52	0.72	0.52	0.36
0.6	0.4	-	1.5 : 1	1	0.25	0.5	0.28	0.21
0.7	0.3	-	2.34 : 1	1	0.11	0.42	0.27	0.2
0.6	0.15	0.25	4 : 1 : 1.67	1	243	15	15	15
0.6	0.25	0.15	4 : 1.67 : 1	1	258.6	15	15	15
0.6	0.3	0.1	6 : 3 : 1	1	4312	15	15	15
0.6	0.35	0.05	12 : 7 : 1	1	8208	15	15	15
0.65	0.3	0.05	13 : 6 : 1	1	195.4	15	15	15
0.65	0.25	0.1	6.5 : 2.5 : 1	1	189.7	15	15	15
0.7	0.1	0.2	7 : 1 : 2	1	134.3	15	15	15

The introduction of PVA significantly improved the granule durability. Compositions containing both bentonite and PVA exhibited markedly higher durability values than those prepared using only bentonite. The improvement in mechanical stability can be attributed to the polymeric nature of PVA, which enhances particle cohesion and forms a flexible binding network within the granule matrix. This polymer network strengthens the interparticle bonding and reduces structural fragmentation in aqueous environments (Denduyver et al., 2024; Hadi and Yoon, 2026). In addition, PVA has been widely reported as an effective binder for the preparation of granular adsorbents, providing improved mechanical integrity and resistance to disintegration during operation in aqueous systems (G. Zhang et al., 2020).

For instance, the Bio–MMT–LDH:bentonite:PVA ratios of 4:1:1.67 and 4:1.67:1 showed durability values of 243 and 258.6 min, respectively. The improvement in durability can be attributed to the polymeric nature of PVA, which forms strong hydrogen bonds with the hydroxyl groups present on the surfaces of biochar, clay minerals, and LDH particles. These interactions promote the formation of a cohesive polymer–mineral network that enhances interparticle adhesion within the granular matrix (S. Zhang et al., 2021). The presence of this polymeric binding phase reduces the susceptibility of the granules to swelling-induced fractures and improves their resistance to mechanical stress in aqueous environments (Hadi and Yoon, 2026).

Further optimization of the binder composition resulted in a substantial increase in the granule durability. The formulation with a Bio–MMT–LDH:bentonite:PVA ratio of 6:3:1 achieved a durability of 4312

min, while the ratio of 12:7:1 exhibited the highest durability value of 8208 min. These results suggest that an optimal balance between the inorganic clay and polymeric binders is required to produce mechanically stable granules. In this system, bentonite contributes structural rigidity through its mineral framework, whereas PVA provides flexible polymeric bridges that bind the composite particles together. The combination of these two binders creates a synergistic binding mechanism in which clay particles act as structural scaffolds, while the polymer phase enhances cohesion and elasticity within the granule matrix (Kim et al., 2025; G. Zhang et al., 2020).

However, excessive deviation from the optimal binder ratio resulted in reduced durability. For example, compositions with ratios of 13:6:1 and 7:1:2 exhibited significantly lower durability values of 195.4 and 134.3 min, respectively. This decline in durability suggests that either insufficient clay content or excessive polymer loading may weaken the granule structural framework. When the clay content is too low, the mineral skeleton required to support the granular structure is insufficient. Conversely, excessive polymer content may produce a softer matrix that lacks rigidity required to withstand mechanical stress. Therefore, the results demonstrate that the durability of the biochar–MMT–Zn/Al LDH granules depends on the formation of a balanced hybrid binding system that combines the rigidity of inorganic clay minerals with the flexibility of polymeric binders. Overall, the findings highlight the critical role of the binder composition in determining the mechanical stability of granular adsorbents. The synergistic interaction between bentonite and PVA significantly enhanced the granule cohesion and resistance to hydration-induced disintegration. This improved structural stability is particularly important for adsorption materials intended for fixed-bed column systems, where adsorbent particles are continuously exposed to flowing water and mechanical stress during operation (Alsaqoor et al., 2022).

Therefore, the results demonstrate that the durability of the Biochar–MMT–Zn/Al LDH granules depends on the formation of a balanced hybrid binding system that combines the rigidity of inorganic clay minerals with the flexibility of polymeric binders. Such hybrid binder systems have been reported to improve the mechanical stability of granular materials through synergistic interactions between the mineral frameworks and polymer networks (Kim et al., 2025). Overall, the findings highlight the critical role of the binder composition in determining the mechanical stability of granular adsorbents. The synergistic interaction between bentonite and PVA significantly enhanced the granule cohesion and resistance to hydration-induced disintegration. This improved structural stability is particularly important for adsorption materials intended for fixed-bed column systems, where adsorbent particles are continuously exposed to flowing water and mechanical stress during operation (ThiMinh et al., 2016).

3.2 Structural Stability under Dynamic Water Flow

In addition to static immersion tests, the structural stability of the Biochar–MMT–Zn/Al LDH granules was evaluated under dynamic water flow conditions to simulate the hydraulic stresses typically encountered in fixed-bed adsorption columns. During column operation, adsorbent particles are continuously subjected to shear forces, particle–particle collisions, and hydrodynamic pressure generated by the flowing water. These forces can gradually weaken the interparticle bonding within the adsorbent matrix, leading to particle erosion, fragmentation, and eventual material loss from the column. Therefore, evaluating the resistance of granulated adsorbents under dynamic flow conditions is essential for assessing their suitability for continuous water treatment applications (Landström et al., 2023; ThiMinh et al., 2016). Therefore, evaluating the resistance of granulated adsorbents under dynamic flow conditions is essential for assessing their suitability for continuous water treatment. Granular adsorbents with sufficient mechanical stability are particularly required for column systems to prevent particle disintegration, pressure drop fluctuations, and adsorbent loss during long-term operation (Hadi and Yoon, 2026).

The experimental results revealed a clear difference in the mechanical performance between the granules prepared with and without the polymeric binder. Granules containing only bentonite exhibited extremely poor stability when exposed to flowing water, with durability values generally below 1 min at all tested flow rates. This rapid degradation suggests that the clay binder alone cannot maintain sufficient cohesion between the composite particles under hydraulic stress. When water flows through the packed bed, the penetration of water molecules into the bentonite structure causes rapid swelling of the clay layers owing to the hydration of montmorillonite interlayers (Clegg et al., 2014). The resulting expansion generates internal stress within the granule matrix, weakening the interparticle contact and accelerating the structural breakdown (Dai et al., 2020). In addition, the absence of a polymeric bridging network renders the granules more susceptible to particle detachment and surface erosion caused by fluid shear forces within the column system (ThiMinh et al., 2016; G. Zhang et al., 2020).

In contrast, incorporating PVA significantly enhanced the resistance of the granules to hydraulic stress. Granules prepared with optimized binder compositions maintained their structural integrity throughout the dynamic flow experiment, reaching the maximum experimental durability of 15 min across all tested flow rates. This improvement can be attributed to the formation of a polymer–mineral hybrid network within the granule. PVA molecules form extensive hydrogen bonding interactions with the hydroxyl groups present on the surfaces of biochar, MMT, and LDH particles, which strengthens the adhesion between particles (S. Zhang et al., 2021). These interactions create polymeric bridges that connect adjacent particles, reinforcing the internal structure of the granules and improving their resistance to fragmentation under shear stress (Hadi & Yoon, 2026; Zhang et al., 2020). Such polymer–mineral hybrid structures are known to enhance the mechanical stability of granular materials by combining the rigidity of inorganic frameworks with the flexibility of polymer networks (Kim et al., 2025)

The enhanced durability observed under dynamic conditions also indicates that the binder system effectively distributed mechanical stress throughout the granule matrix. In the presence of PVA, external hydraulic forces were partially absorbed by the flexible polymer network rather than being concentrated at individual particle contact points. Such polymer networks are known to dissipate mechanical energy and distribute stress more evenly within composite materials, thereby reducing the likelihood of localized fractures (Kim et al., 2025). Bentonite contributes to mechanical rigidity by forming a mineral scaffold that supports the composite framework (Bertolini et al., 2022). PVA molecules can also form hydrogen bonds with hydroxyl groups on mineral and carbon surfaces, strengthening particle adhesion and enhancing the cohesion of the granule matrix (S. Zhang et al., 2021). Therefore, the synergistic interaction between the inorganic clay structure and the flexible polymer binder produces a hybrid structure that combines rigidity with elasticity, enabling the granules to withstand both compressive and shear stresses generated during water flow (Hadi and Yoon, 2026).

From an engineering perspective, the ability of granules to remain stable under dynamic flow conditions is critical for their application in fixed-bed adsorption systems. In practical water treatment processes, adsorbents are often exposed to prolonged hydraulic loading, and mechanically unstable materials may undergo attrition or fragmentation, leading to an increased pressure drop and loss of adsorption capacity (Landström et al., 2023; ThiMinh et al., 2016). Granular adsorbents with sufficient mechanical strength are essential for maintaining stable column operations and preventing particle loss during continuous treatment processes (Hadi and Yoon, 2026). The results obtained in this study demonstrate that the optimized Biochar–MMT–Zn/Al LDH granules possess sufficient mechanical resistance to withstand short-term hydraulic stress, indicating their potential applicability in continuous flow systems. The improved durability achieved through the combined use of bentonite and PVA represents an important step toward bridging the gap between laboratory-scale adsorbent synthesis and practical implementation in column-based water treatment technologies (Kim et al., 2025).

3.3 Implications for Column Adsorption Applications

The development of mechanically stable granular adsorbents is critical for the practical implementation of adsorption processes in continuous water treatment systems (Afolabi et al., 2022; Ren et al., 2020). Unlike batch adsorption systems, fixed-bed columns operate under continuous flow conditions, where adsorbent particles are subjected to hydraulic pressure, interparticle friction, and prolonged contact with aqueous environments (Patel, 2019, 2022). Under these conditions, powdered adsorbents often exhibit several operational limitations, including excessive pressure drop, bed clogging, and material loss through particle entrainment (Afolabi et al., 2022; Balasubramanian et al., 2025). These challenges significantly reduce the process efficiency and complicate the separation of adsorbent materials from treated water. Therefore, converting powdered adsorbents into mechanically stable granular forms is an important strategy for improving the applicability of adsorption technologies in real treatment systems.

The results obtained in this study demonstrate that the granulation of biochar–MMT–Zn/Al LDH composites using bentonite and polyvinyl alcohol binders can significantly improve the structural durability of the material (Alsaqoor et al., 2022). The optimized granules maintained structural integrity under both static immersion and dynamic water flow conditions, indicating that the granulation process effectively strengthened the internal particle network of the composite (Hu et al., 2025; J. Zhang et al., 2025). This structural reinforcement is essential for maintaining a stable packed bed during column operations. Mechanically stable granules help preserve bed porosity and prevent excessive compaction, allowing water to flow uniformly through the adsorbent bed while maintaining efficient contact between the adsorbent surface and the dissolved pollutants. The particle size and mechanical strength are also important factors influencing the column performance (Majors, 1973; Patel, 2022; Paul Chen et al., 2003). Granules with diameters ranging from 0.5 to 1.5 cm provide several advantages for column systems. Larger particle sizes generally reduce the hydraulic resistance within the packed bed, thereby minimizing the pressure drop across the column. Larger particle sizes generally reduce the hydraulic resistance within the packed bed, thereby minimizing the pressure drop across the column (Allen et al., 2013; Koekemoer and Luckos, 2015). This behavior is consistent with packed-bed flow theory, where pressure drop is inversely related to particle diameter due to increased void space and reduced flow resistance in beds composed of larger particles (Keyser et al., 2006; Koekemoer and Luckos, 2015)

Simultaneously, sufficient mechanical strength prevents particle fragmentation, which could generate fine particles capable of clogging the pore spaces within the bed. Therefore, maintaining an optimal balance between particle size and structural stability is essential for achieving efficient mass transfer and long-term operational stability in column adsorption systems. In addition to mechanical stability, the composite structure of the Biochar–MMT–Zn/Al LDH material offers potential advantages for adsorption performance. Biochar contributes a porous carbon matrix with abundant surface functional groups that can interact with contaminants through physical adsorption and surface complexation. MMT has a high cation exchange capacity and layered structures that facilitate the adsorption of metal ions and other positively charged species. Meanwhile, Zn/Al LDH introduces anion exchange capability through positively charged hydroxide layers and exchangeable interlayer anions. Therefore, the integration of these materials within a single composite structure creates a multifunctional adsorbent capable of interacting with a wide range of pollutants through multiple adsorption mechanisms.

From an engineering perspective, the durability of the granules observed in this study suggests that the developed composite material may be suitable for application in fixed-bed adsorption columns for water treatment. The ability of the granules to withstand hydraulic stress indicates that they can maintain structural stability during prolonged column operation, which is essential for preventing adsorbent loss and maintaining consistent adsorption performance. Furthermore, the use of low-cost and widely available binders, such as

bentonite and starch-based materials, provides an economically attractive approach for producing granular adsorbents on a larger scale. Overall, the findings of this study highlight the importance of combining material design with particle engineering strategies to improve the practical applicability of adsorbent materials. By integrating biochar, MMT, and LDH within a mechanically stable granular structure, the developed composite offers promising potential for application in continuous adsorption systems. Future research should focus on evaluating the breakthrough behavior, adsorption kinetics, and regeneration performance of granules in fixed-bed column experiments to fully assess their effectiveness in real-world water treatment applications.

3.4 Recent Trends in Granular Adsorbent in Column Sorption

The analysis of publication trends related to granular adsorbents for column sorption revealed a substantial and continuous increase in research activity over the past two decades. Based on the Scopus database analysis, 1,287 documents were identified between 2007 and 2026, indicating rapidly expanding research interest in adsorption technologies designed for continuous water treatment systems. The annual publication trend shown in Figure 1 demonstrates a clear evolution in the research field, reflecting the growing importance of granular adsorbent materials in environmental remediation.

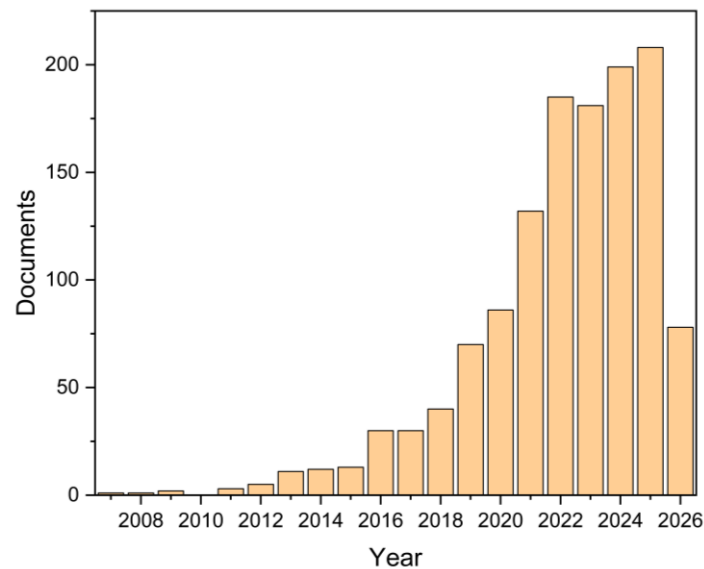


Figure 1. Trends in publication according to Scopus database based on years (updated March 11, 2026)

During the early development stage (2007–2014), the research output remained relatively limited, with fewer than 15 publications per year. This period can be considered the foundational stage of adsorption research, during which most studies focused on fundamental adsorption mechanisms, material synthesis, and batch adsorption experiments. At that time, many adsorbent materials were still being investigated in powdered form because batch experiments allow rapid evaluation of adsorption capacity and surface interactions. However, these experimental systems do not fully represent the operational conditions encountered in practical water treatment processes.

A gradual increase in publication activity was observed between 2015 and 2019, where the number of studies began to rise from approximately 30 to nearly 70 publications annually. This increase suggests the growing recognition of adsorption as a viable technology for environmental remediation, particularly for the removal of emerging contaminants, such as dyes, pharmaceuticals, and heavy metals. During this period, researchers began exploring composite adsorbent materials that combine carbon-based materials with mineral components to enhance adsorption performance.

The most significant growth in research activity occurred after 2020, when publication output increased sharply and exceeded 130 documents per year. The number of publications reached approximately 180–210 documents annually between 2022 and 2025, representing the peak research activity period. This rapid expansion reflects the increasing global attention toward sustainable water treatment technologies and the development of advanced adsorbent materials for wastewater treatment. In particular, biochar-based composites and hybrid materials incorporating clay minerals, metal oxides, and LDH have gained increasing interest owing to their multifunctional adsorption properties and relatively low production costs.

The distribution of document types further illustrates the maturity of the field. As shown in Table 1, research articles (526 records) and review articles (514 records) dominate the literature, accounting for most of the identified publications. The high number of research articles indicates active experimental investigations aimed at developing new adsorbent materials and evaluating their performances. Meanwhile, the substantial number of review articles suggests that the field has reached a level of maturity, where researchers are synthesizing existing knowledge to identify emerging research directions, technological challenges, and opportunities for innovation.

Other document types, such as book chapters (113 records) and conference abstracts (68 records), also contribute to the dissemination of scientific knowledge, particularly in early stage research and interdisciplinary collaboration. These publications often present preliminary findings, new material synthesis approaches, or emerging adsorption technologies that may later be developed into full research articles. In contrast, fewer case reports, editorials, and short communications indicate that these formats play a minor role in the research field's development.

Despite the rapid growth in research output, bibliometric analysis indicates that many studies still emphasize adsorption performance under batch experimental conditions. Fewer studies have investigated the mechanical durability, hydraulic stability, and structural robustness required for adsorbents to function effectively in fixed-bed column systems. Because column adsorption processes involve continuous hydraulic loading and prolonged operational periods, adsorbent materials must possess sufficient mechanical strength to resist attrition, swelling, and particle fragmentation. Therefore, the development of mechanically stable granular adsorbents remains an important area of research.

In this context, the present study contributes to the growing research area of granular composite adsorbents by focusing specifically on the structural durability of biochar–MMT–Zn/Al LDH granules. By evaluating the mechanical stability of the granulated composite under both static immersion and dynamic water-flow conditions, this study addresses a key challenge in the practical implementation of adsorption technologies. These results provide valuable insights into the design of durable granular adsorbents suitable for fixed-bed column systems in water treatment applications.

Table 1. Trends in publication according to article types

Document Type	Number of Records
Review articles	514
Research articles	526
Encyclopedia	16
Book chapters	113
Conference abstracts	68

Document Type	Number of Records
Case reports	3
Correspondence	1
Editorials	1
Short communications	2
Other	43

3.5 Bibliometric Analysis

The keyword co-occurrence network presented in Figure 1 illustrates the research landscape of biochar-based composite materials for adsorption applications. The mapping was generated using VOSviewer based on publications retrieved from the Scopus database between 2010 and 2025. The visualization highlights the relationships between frequently used keywords, where the node size represents the frequency of occurrence and the link strength indicates the degree of association between research topics.

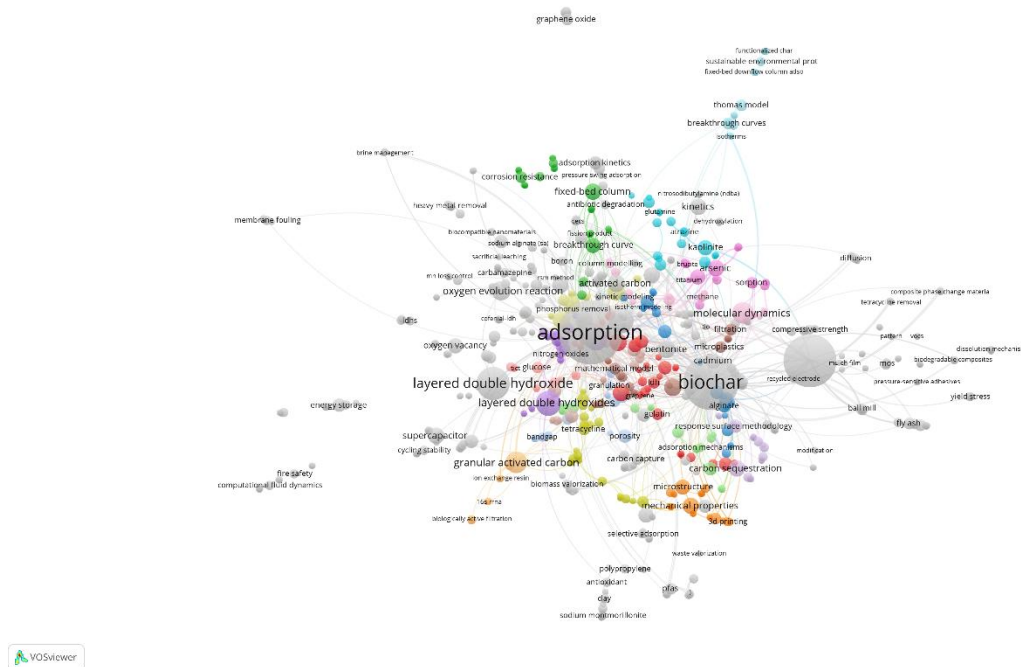


Figure 2. Keywords mapping for this issue

The map reveals that the keyword “adsorption” is the most dominant term and is positioned at the center of the network. This indicates that adsorption remains the primary mechanism investigated in studies on biochar and related composite materials. The strong connections between “adsorption,” “biochar,” and “layered double hydroxide” demonstrate that these materials are frequently studied in combination to enhance pollutant removal performance. Several thematic clusters were identified in the visualization. One cluster was strongly associated with biochar-based materials, including keywords such as biochar, activated carbon, carbonization, and surface modification. This cluster reflects research focusing on biomass-derived carbon materials and their structural modifications to improve adsorption properties. Biochar is widely used because

of its high porosity, surface functional groups, and relatively low production cost derived from biomass resources.

Another cluster is related to LDH materials, represented by keywords such as layered double hydroxide, LDH, and metal hydroxides. This cluster highlights the increasing interest in LDH materials as adsorbents because of their layered structure, high anion exchange capacity, and tunable chemical composition. The presence of LDH-related keywords close to biochar suggests that hybrid materials combining biochar and LDH have become an important research direction for improving adsorption performance. A separate cluster was associated with column and dynamic adsorption systems, which included keywords such as fixed-bed column, breakthrough curve, and kinetics. These terms indicate that research on adsorption has gradually expanded from traditional batch experiments to continuous flow systems. However, the relatively small number of keywords in this cluster suggests that column-based studies remain less explored than batch adsorption research.

In addition, several keywords related to molecular dynamics and computational modeling were observed in the network. This indicates that theoretical approaches are increasingly being used to understand adsorption mechanisms at the molecular level. Computational studies can provide insights into adsorption energy, surface interactions, and adsorption pathways, complementing experimental investigations. Overall, the bibliometric analysis demonstrates that research on biochar-based composite adsorbents is rapidly expanding, particularly in the development of hybrid materials that combine carbon-based structures with mineral components such as LDH. However, the mapping also indicates that studies focusing on granular adsorbents and column applications remain relatively limited. Most existing studies still emphasize adsorption performance under batch conditions rather than evaluating the structural durability and mechanical stability required for practical column operations. Therefore, the present study addresses this research gap by developing a granulated biochar–MMT–Zn/Al LDH composite and evaluating its mechanical durability under both static and dynamic water conditions. By focusing on the structural stability of granular adsorbents, this study bridges the gap between laboratory-scale adsorption studies and practical implementation in continuous water treatment systems.

3.6 Recommendation and Limitation Study

Although the present study demonstrates the feasibility of producing mechanically stable biochar–MMT–Zn/Al LDH granules for potential column adsorption applications, several limitations should be acknowledged. First, this study primarily focused on evaluating the structural durability of the granulated composites under static immersion and dynamic water-flow conditions. While durability is an essential parameter for fixed-bed adsorption systems, the adsorption performance of the developed granules toward specific contaminants was not experimentally investigated in this study. Therefore, further research is necessary to evaluate the adsorption capacity, selectivity, and removal efficiency of granules for common water pollutants, such as heavy metals, nutrients, and organic contaminants. Another limitation of this study is the absence of a detailed physicochemical characterization of the composite material after the granulation process. Granulation using binders, such as bentonite and polyvinyl alcohol, may influence the surface area, pore structure, and accessibility of adsorption sites within the composite. Future studies should therefore include comprehensive material characterization techniques such as scanning electron microscopy (SEM), X-ray diffraction (XRD), Brunauer–Emmett–Teller (BET) surface area analysis, and Fourier transform infrared spectroscopy (FTIR) to better understand the structural and chemical properties of the granulated composites.

In addition, the dynamic durability test performed in this study was limited to water flow resistance within a laboratory-scale column system. Although the results demonstrate that the optimized binder composition significantly improves the structural stability of the granules, the long-term operational stability under continuous column operation was not examined. In practical water treatment applications, adsorbents

are often subjected to prolonged hydraulic stress, repeated adsorption–desorption cycles, and varying water chemistry conditions. Therefore, future investigations should assess the long-term mechanical stability, regeneration potential, and reusability of granulated adsorbents under realistic operating conditions. Furthermore, the granulation process in this study was conducted using manual pelletization, which may have led to variations in granule size and morphology. For large-scale applications, more controlled granulation techniques, such as extrusion, spray granulation, or pelletizing reactors, may be required to ensure a uniform particle size distribution and consistent mechanical strength. Therefore, process optimization and scale-up considerations should be addressed in future studies.

Despite these limitations, the present study provides an important preliminary evaluation of the structural durability of biochar–MMT–Zn/Al LDH granules and highlights the critical role of binder composition in improving mechanical stability. These findings contribute to the development of mechanically robust granular adsorbents suitable for fixed-bed column systems in water treatment applications. Future studies should integrate adsorption performance evaluation, advanced material characterization, and long-term column studies to fully assess the potential of this composite material for practical environmental remediation.

4. Conclusion

In this study, a granulated biochar–MMT–Zn/Al LDH composite was successfully developed as a mechanically durable adsorbent for potential column adsorption applications. The results demonstrated that the binder composition plays a critical role in determining the structural stability of the granules. Granules prepared using bentonite alone exhibited extremely poor durability and rapidly disintegrated in aqueous environments. In contrast, the addition of polyvinyl alcohol (PVA) significantly improved the mechanical strength of the granules by forming a cohesive binding network within the composite structure. The optimization of the binder ratio revealed that an appropriate balance between the inorganic clay and polymeric binders is required to produce stable granules. The optimized composition achieved a durability of up to 8208 min under static conditions and maintained its structural integrity under dynamic water-flow tests. These results indicate that the combined use of bentonite and PVA has a synergistic effect that enhances granule cohesion and resistance to hydraulic stress.

These findings highlight the importance of granulation strategies in improving the practical applicability of powdered adsorbent materials in fixed-bed column systems. By converting biochar-based composite powders into mechanically stable granules, the operational challenges associated with powder adsorbents, such as clogging, pressure drop, and material loss, can be minimized. Overall, the developed biochar–MMT–Zn/Al LDH granules demonstrated promising structural durability for column adsorption systems. Future research should focus on evaluating the adsorption performance of granules toward specific contaminants, conducting detailed physicochemical characterization of the composite materials, and assessing long-term stability and regeneration performance under continuous column operation.

Declaration of Generative AI and AI-assisted Technologies in the Writing Process

During the preparation of this work, the author(s) used ChatGPT to improve the readability and fix grammatical errors. After using these tools/services, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the published article.

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Ethics Declaration

This study did not require ethical clearance.

CRediT authors statement

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