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CATALYSTS BASED ON POLYMER-CLAY COMPOSITES

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Abstract: *Introduction*. The history of polymer-clay composites dates back to the mid-20th century when the addition of clay minerals such as montmorillonite to polymer matrices was found to significantly enhance their mechanical and thermal properties. They are used in environmentally friendly processes, redox reactions, and pollutant remediation. The introduction of nanotechnology has further advanced these materials by enabling nanoscale dispersion of clay particles, increasing their surface area and interaction with polymers. In recent years, research has focused on environmentally friendly materials, particularly biopolymer matrices such as chitosan and starch. *Objective of the study.* This review aims to analyze the latest developments in polymer-clay composite-based catalysts, highlighting key challenges and identifying promising research directions. *Results and Discussion.* Polymer-clay composites provide structural stability, improved mechanical strength, and high thermal resistance. The integration of clay minerals such as montmorillonite or zeolites enhances adsorption capacity and ion-exchange properties, making these materials highly effective in catalysis and environmental applications. Metal nanoparticles (e.g., Pd, Ni, Cu), embedded in polymer-clay matrices, significantly boost catalytic activity, offering efficient solutions for oxidation, reduction, and energy conversion reactions. However, challenges remain in polymer degradation, catalyst deactivation, and scalability for industrial applications. *Conclusions.* Advancements in hybrid polymer-clay composites with metal nanoparticles offer promising directions for green chemistry and sustainable catalysis. Future research should focus on improving the stability, scalability, and functionalization of these materials to enhance their practical applications in industrial and environmental processes.

Key words: polymer-clay composites, metal nanoparticles, catalysis, composite synthesis, green chemistry.

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1. Introduction

The history of polymer-clay composites dates back to the mid-20th century when it was first recognized that the addition of clay minerals, such as montmorillonite, to polymer matrices could significantly enhance their mechanical and thermal properties [1, 2]. A major milestone in the development of these materials was the introduction of nanotechnology, which enabled the fabrication of composites with clay nanoparticles capable of increasing surface area and interacting with the polymer at the molecular level. In recent decades, research has increasingly focused on environmentally friendly materials, including biopolymer matrices such as starch, chitosan, and others [3–5].

The significance of catalysts based on polymer-clay composites and metal nanoparticles stems from their unique properties and broad application prospects across various scientific and industrial fields [6–8]. These catalysts represent innovative materials with exceptional characteristics, including a high specific surface area, enhanced catalytic activity, and the ability to be modified for various applications [9,10]. This combination of properties makes them highly effective for conducting complex chemical reactions, including redox processes, degradation of organic pollutants, and energy conversion [11].

Polymer-clay composites provide structural stability, improved mechanical strength, and high thermal resistance [12]. The incorporation of clays such as montmorillonite or zeolites enhances their adsorption properties and ion-exchange capacity, making them highly effective in water purification processes and the separation of complex mixtures [13]. Metal nanoparticles, such as gold, palladium, or silver, integrated into the composite matrix, significantly enhance catalytic activity by creating active sites for oxidation, reduction, and hydrogenation reactions [14, 15].

Their potential is particularly important in environmental chemistry, where such composites can be utilized for the purification of water and air from toxic substances. In industrial chemistry, they contribute to reducing the energy intensity of processes and minimizing the formation of by-products [16]. Furthermore, they can serve as the foundation for the development of more accessible and cost-effective catalysts, replacing conventional catalysts based on precious metals and toxic substances. This is especially relevant when utilizing locally available materials and inexpensive polymers, promoting sustainable development and resource conservation.

The objective of this review article is to analyze the latest advancements in the development of catalysts based on polymer-clay composites, identify key challenges, and determine promising directions for further research.

2. Polymer-Clay Composites as a Basis for Catalysts

2.1 Structure and Properties of Polymer-Clay Composites

Clays are used as fillers in composites due to their unique properties, including high specific surface area, interlayer swelling capability, high adsorption capacity, and mechanical strength [17].

Natural clays are layered minerals consisting of alternating layers of silicon and aluminum oxides. These layers contain cavities and interlayer spaces that can retain water molecules, metal ions, and organic compounds. The primary active sites of clays are located either on their surface or within the interlayer regions, where adsorption and ion-exchange processes can take place (Table 1). Due to these properties, clays can interact with various chemical substances, making them effective catalyst supports. Their natural microporous structure allows for the immobilization of active substances, ensuring a high interaction surface, which is crucial for catalysis.

Table 1 - Comparison of the characteristics of smectite clays

Parameter	**Montmorillonite**	**Hectorite**	**Saponite**	**Beidellite**
Chemical formula	$(Na,Ca)_{0.3}(Al,Mg)_2Si_4O_{10}(OH)_2 \cdot nH_2O$	$(Na,Ca)_{0.3}(Mg,Li)_3Si_4O_{10}(OH)_2$	$(Na,Ca)_{0.3}(Mg,Al)_3(Si,Al)_4O_{10}(OH)_2$	$(Ca)_{0.3}(Al,Mg)_2Si_4O_{10}(OH)_2$
Main elements	Si, Al, Mg, Na	Si, Mg, Li	Si, Mg, Al	Si, Al, Mg, Ca
Layer type	2:1	2:1	2:1	2:1
Interlayer spacing (Å)	12-15	12-14	11-13	10-12
Type of interlayer cations	Na$^+$, Ca^{2+}	Na$^+$, Ca^{2+}	Na$^+$, Ca^{2+}	Ca^{2+}
Cation exchange capacity (meq/100 g)	80-150	50-70	80-120	40-60
Specific surface area (m^2/g)	600-800	300-600	500-700	400-500
Thermal stability (°C)	200-600	200-700	200-600	200-700

In catalysis, polymers ensure control over reagent access to active sites. Polymers play a crucial role in catalysis by controlling the accessibility of reactants to active sites and preventing their deactivation under harsh conditions such as high temperatures or aggressive chemical environments. Polymers used in composite materials are classified into synthetic and natural matrices. Examples of synthetic polymers include polyacrylic acid, polyethyleneimine, and polyvinyl alcohol, which can interact with clay particles, enhancing the catalytic performance of the composite. Synthetic polymers form structures where molecules or metal nanoparticles are evenly distributed, preventing aggregation and loss of activity. However, composites based on polyethylene glycol (PEG) and other synthetic polymers are rarely used as catalysts and adsorbents due to their chemical inertness, weak chelating properties, and low thermal stability, which limits their application in catalytic and adsorption processes. Additionally, they exhibit poor mechanical strength and may block active sites in the composite, reducing its overall efficiency.

With the advent of green chemistry in catalysis, natural biopolymers such as cellulose, chitosan, pectin, starch, alginate, gelatin, and casein have increasingly been used as matrix materials. Biopolymers, derived from plant residues and microbial sources, are environmentally friendly [19]. They are widely available, cost-effective, and their composites readily degrade in natural environments [18]. At the molecular level, the interaction between polymers and clays in composites occurs through various physicochemical mechanisms, including adsorption, ion exchange, hydrogen bonding, and covalent interactions. Polymer chains can intercalate into the interlayer spaces of clays, increasing the distance between layers and creating a porous structure. Functional groups in polymers (e.g., amino groups in chitosan or carboxyl groups in polyacrylic acid) can bind to surface ions of clays, enhancing adhesion between phases. This interaction strengthens the composite's stability and promotes uniform distribution of active sites, which is particularly important for catalysis. As a result, polymer-clay composites exhibit a synergistic effect: the polymer protects active particles, while the clay ensures their accessibility to reactants.

2.2. Methods for synthesizing polymer-clay composites

Polymer-clay composites can be synthesized using various methods, each with its specific features and advantages. The choice of a particular method depends on the required properties of the final material and its intended application conditions.

1. Polymer intercalation into clay interlayer spaces

This method involves the insertion of polymer chains between the layers of clay minerals such as montmorillonite. For successful intercalation, the clay is pre-modified with organic compounds that enhance its compatibility with the polymer. As a result, nanocomposites with improved mechanical and thermal properties are formed.

2. Mechanochemical mixing

In this method, the polymer and clay undergo simultaneous mechanical treatment, such as milling in ball mills. Intensive mixing and grinding activate the surfaces of the components, promoting their interaction at the molecular level. This approach allows for the production of homogeneous composites without the use of solvents.

3. Chemical cross-linking and surface modification

This method involves the chemical modification of either the clay or polymer to create strong covalent bonds between them. For example, clays can be functionalized using silane agents, which subsequently react with the polymer’s functional groups, forming a cross-linked structure. This approach improves component compatibility and enhances the stability of the composite.

3. Modification and functionalization of composites

To enhance the catalytic activity of polymer-clay composites, both the polymer matrix and the clay support can be modified using various methods. Polymers are functionalized by introducing active groups (e.g., amino, carboxyl, and sulfo groups), which promote chelation and retention of metal nanoparticles. Chemical cross-linking of polymers with agents such as glutaraldehyde or epichlorohydrin improves their mechanical strength and solvent resistance. Clays can be modified through ion exchange by incorporating active cations (Pd^{2+}, Cu^{2+}, Ni^{2+}), acid activation to increase porosity, or pillaring with polyoxometals (e.g., Al^{3+}, Ti^{4+}), which expands interlayer spacing and increases the number of available active sites [19]. The incorporation of metal nanoparticles via thermal or chemical treatment also creates a synergistic effect between the polymer and clay, ensuring high catalytic activity, selectivity, and stability of the composites.

Composites containing metal nanoparticles exhibit high efficiency in catalytic processes due to the unique physicochemical properties of nanostructured materials. For instance, palladium and platinum nanoparticles embedded in composite matrices serve as highly effective catalysts in photocatalysis and heterogeneous catalysis, providing high selectivity and stability [20].

In green chemistry and environmental applications, these composites are used for water and air purification. Metal nanoparticles such as silver and gold facilitate the degradation of toxic organic compounds, while metal oxides like TiO_2 and ZnO are effective in the catalytic oxidation of toxic gases (e.g., CO, NO_x). These materials contribute to environmentally friendly processes by reducing harmful emissions.

Cation exchange also plays a key role in improving the activity of polymer-clay composites, as it enables the effective incorporation and retention of active metal centers within the interlayer spaces of the clay. The replacement of natural cations (e.g., Na^+ or Ca^{2+}) with active metal cations (Pd^{2+}, Ni^{2+}, Cu^{2+}, etc.) creates stably anchored catalytic sites, promoting their uniform distribution and preventing aggregation (Table 2). As a result, interactions between active components and reactants are enhanced, the active surface area increases, and the adsorption of target molecules improves. Overall, cation exchange contributes to increased catalytic activity, stability of composites, and the possibility of their repeated use in reactions.

Table 2 - Comparison of the Characteristics of Low-Exchange Clays

Parameter	**Kaolinite**	**Illite**	**Halloysite**	**Pyrophyllite**
Chemical formula	$Al_2Si_2O_5(OH)_4$	$(K,H_3O)(Al,Mg,Fe)_2(Si,Al)_4O_{10}[(OH)_2,(H_2O)]$	$Al_2Si_2O_5(OH)_4 \cdot nH_2O$	$Al_2Si_4O_{10}(OH)_2$
Main elements	Si, Al, O, H	Si, Al, Mg, Fe, K, O, H	Al, Si, O, H (примеси Fe, Ti, Cr)	Al, Si, O, H
Layer type	1:1	2:1	1:1	2:1
Interlayer spacing (Å)	~7	~10	~7.2	~9.3–9.6
Type of interlayer cations	No exchangeable cations (layers held by hydrogen bonds)	K^+, with minor Na^+, Ca^{2+}	No exchangeable cations (layers held by hydrogen bonds and water layers)	No exchangeable cations (layers held by van der Waals interactions)
Cation exchange capacity (meq/100 g)	3-15	10-40	10-60	20-50
Specific surface area (m^2/g)	10-20	50-120	50-70	20-40
Thermal stability (°C)	500-700	400-600	500-600	700-900

4. Catalytic applications of polymer-clay composites in modern research

Polymer-clay composites have found extensive applications in heterogeneous catalysis due to the combination of high clay surface area and the functional groups of polymers. Clay-based materials, such as montmorillonite and zeolites, provide structural support and catalytically active sites while enhancing adsorption, the distribution of active species, and resistance to deactivation. These composites are used in transesterification, oxidation, hydrogenation reactions, and pollutant removal processes, demonstrating enhanced selectivity, stability, and the possibility of multiple reuses.

For example, in the transesterification of waste cooking oil (WCO) into biodiesel, Fereidooni L. and Mehrpooya M. developed a heterogeneous catalyst based on zeolite/chitosan/KOH. The zeolite underwent desilication with increased K^+ incorporation, forming hydroxy-potassalite. The use of electrolysis and an acetone co-solvent facilitated a 93% yield of methyl esters [21]. Later, Ahmed et al. synthesized a montmorillonite-chitosan (PAMMT-CH) nanocomposite, which, when used in the catalytic pyrolysis of waste oil, achieved a high bio-jet fuel yield with improved selectivity towards alkanes at 300°C and 0.7% catalyst loading [22].

Abdullah et al. utilized a green synthesis method, incorporating starch as a natural stabilizer and zeolite as a support, eliminating the use of toxic chemicals and solvents. The resulting bio-nanocomposite starch/Fe_3O_4/zeolite (BNC) exhibited high magnetic activity (1.84 emu/g) and uniformly distributed magnetite nanoparticles (average diameter 9.24 nm) on the zeolite surface. The product was effective as a non-precious metal catalyst for the oxygen reduction reaction (ORR) in energy applications [23].

In a study by Zhao et al., a novel hybrid structure was developed, consisting of chitosan (CS) chains and palladium nanoparticles (Pd^0) embedded within the interlayer spaces of montmorillonite (Mt), modified with aluminum or aluminum-iron oxides (Al-Mt or Al-Fe-Mt). These catalysts demonstrated high activity in Sonogashira reactions due to excellent nanoparticle dispersion (down to 3 nm) and a large surface area. Pd^0@CS/Al-Fe-Mt showed superior performance due to its higher adsorption capacity and smaller Pd^0 nanoparticles [24].

In 2024, Sun et al. developed hybrid chitosan-Pd^0 nanocomposites (Pd^0@CS/Al, Co-MMt), where 2–3 nm palladium nanoparticles were chelated by chitosan chains and embedded in montmorillonite, modified with aluminum and cobalt oxides. A simple one-step thermal treatment at 200°C in an inert atmosphere was employed, simultaneously modifying the clay and incorporating the polymer. The nanocomposites exhibited high catalytic activity and stability in cross-coupling reactions (Sonogashira and Heck reactions). A key feature of this study was the synergistic effect of Al, Co-pillaring and chitosan chain encapsulation, leading to an increased surface area of 198.3 m^2/g and improved catalytic performance compared to multi-step synthesis methods [25].

Li Yaqi et al. developed a photocatalyst (Cht/PANI/$MnFe_2O_4$) with high photocatalytic activity and adsorption capacity, synthesized via in-situ polymerization using chitosan, polyaniline, and $MnFe_2O_4$. The composite exhibited a maximum adsorption capacity of 2569.66 mg/g, significantly outperforming pure $MnFe_2O_4$ due to the synergistic effect of the polymers, enhancing charge transfer and material stability [26].

Similarly, Breno F. Ferreira et al. synthesized a hybrid catalyst (Kaol-TRIS-MnTSPP) composed of kaolinite, modified with tris(hydroxymethyl)aminomethane (TRIS) and immobilized metalloporphyrin (MnTSPP) via adsorption. The modification of kaolinite expanded interlayer spacing and improved interactions with MnTSPP, ensuring strong immobilization of porphyrin and preventing its leaching during catalysis. The catalyst demonstrated high efficiency in cyclooctene oxidation (81% conversion) and ε-caprolactone (ε-CL) polymerization, yielding biodegradable polycaprolactone (PCL) under mild temperature and energy conditions. This study highlights the importance of chemical modification of clays for the synthesis of effective catalysts in polymerization and oxidation processes [27].

5. Challenges and limitations of current research

One of the major challenges in the development of polymer-clay composite catalysts is the degradation of the polymer matrix and the leaching of active components during repeated use. Polymers such as polyethylene glycol (PEG) and chitosan have limited thermal stability and may lose their stabilizing properties under high temperatures or chemically aggressive environments, leading to a decline in catalytic activity and selectivity. The introduction of functional groups or chemical cross-linking of polymers can partially address this issue, but the risk of catalyst deactivation over prolonged use remains a critical concern.

Most studies are conducted at the laboratory scale, which poses significant challenges in transitioning to industrial applications. Polymer-clay composites require complex, multi-step synthesis processes, including clay modification, polymer functionalization, and uniform distribution of metal nanoparticles. Scaling up these processes presents difficulties in ensuring uniform distribution of active sites, controlling particle size, and effectively embedding polymers into the clay interlayer spaces. These limitations lead to significant differences in catalytic performance between laboratory-scale and industrial-scale applications.

Furthermore, the effects of high temperatures and chemically reactive environments on polymer-clay interactions remain poorly understood, making it difficult to predict the behavior of composites under real reaction conditions. For example, at temperatures exceeding 200–300°C, polymers may lose their structural stability, while clays may undergo structural transformations, leading to a decline in adsorption and catalytic properties. Additional research is required to improve the understanding of polymer-clay chemical compatibility, especially in the presence of metal nanoparticles, to minimize composite degradation and optimize performance in extreme conditions.

6. Prospective research directions

Recent studies highlight the significant potential of hybrid polymer-clay composites incorporating metal nanoparticles (e.g., Pd, Ni, Cu) and organic ligands, such as polyaniline or amine groups. These systems enable a high degree of functionalization, where metal nanoparticles serve as active catalytic centers, while organic ligands stabilize them and regulate interactions with reactants. This combination enhances catalyst selectivity and activity, while also preventing metal leaching, which is crucial for improving the longevity of catalysts in industrial processes.

Sustainable development necessitates the creation of catalysts effective at low temperatures and pressures, thereby minimizing energy consumption in industrial production. Polymer-clay composites with high surface area and porosity, modified with active sites (e.g., metals or Lewis acids), can efficiently catalyze reactions at moderate temperatures, including transesterification, hydrogenation, and oxidation. The use of such composites in green reactions with minimal energy requirements represents an important research direction in the chemical industry.

In response to environmental challenges, increasing attention is being given to the development of biodegradable polymer-clay composites based on natural materials such as polysaccharides or biodegradable synthetic polymers. Green synthesis methods, including in-situ polymerization in aqueous media or acid activation of clays without toxic reagents, ensure the environmental safety of these composites. Their promising applications include wastewater treatment, photocatalysis, and biomedical technologies, where biocompatibility and non-toxicity are essential.

7. Conclusion

Polymer-clay composites with metal nanoparticles and functional polymer matrices represent one of the most promising areas of modern research in the field of catalysis. A review of recent studies highlights that the synergistic interaction between polymers and clay enhances the stability of active sites, improves the kinetics of catalytic reactions, and increases adsorption capacity. These composites have demonstrated excellent performance in transesterification, wastewater treatment, and photocatalysis, owing to their ability to be modified and adapted to various reaction conditions.

However, several challenges remain, including polymer matrix degradation, scalability issues, and insufficient understanding of component interactions under extreme conditions, which require further investigation. The development of hybrid systems incorporating organic ligands and green synthesis methods will not only enhance the selectivity and stability of these composites but also make their production more environmentally friendly.

Future research directions include the development of biodegradable composites that operate efficiently at low energy consumption and the exploration of new mechanisms of interaction between polymers, clays, and metal nanoparticles. The application of *in situ* and *in operando* techniques to understand these mechanisms will provide a solid foundation for the design of innovative catalysts capable of addressing the challenges of sustainable development and resource conservation. Thus, further research on polymer-clay composites is expected to make a significant contribution to green technologies and industrial chemistry.

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ПОЛИМЕР-ГЛИНАЛЫ КОМПОЗИТТЕР НЕГІЗІНДЕГІ КАТАЛИЗАТОРЛАР

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Түйіндеме. *Кіріспе.* Полимер-балшық композиттерінің тарихы XX ғасырдың ортасынан бастау алады, сол кезде монтмориллонит сияқты балшық минералдарын полимерлік матрицаларға қосу олардың механикалық және термиялық қасиеттерін айтарлықтай жақсартатыны анықталды. Олар экологиялық таза процестерде, тотығу-қалпына келтіру реакцияларында және ластағыштарды тазарту саласында қолданылады. Нанотехнологияларды енгізу глинаны нанодисперсияға түсіруге мүмкіндік беріп, оның беткі ауданын және полимерлермен өзара әрекеттесуін арттырды. Соңғы жылдары зерттеулер экологиялық таза материалдарға, соның ішінде хитозан және крахмал сияқты биополимерлік матрицаларға бағытталған. *Зерттеу мақсаты.* Бұл шолу жұмысы полимер-балшық композиттері негізіндегі катализаторларды әзірлеу саласындағы соңғы жетістіктерді талдау, негізгі мәселелерді анықтау және болашақ зерттеулердің перспективалық бағыттарын белгілеуге бағытталған. *Нәтижелер мен талқылау.* Полимер-балшық композиттері құрылымдық тұрақтылықты, жақсартылған механикалық беріктікті және жоғары термотұрақтылықты қамтамасыз етеді. Монтмориллонит немесе цеолит сияқты балшықтардың қосылуы олардың адсорбциялық қасиеттері мен ионалмасу қабілетін арттырып, бұл материалдарды катализ және экологиялық қолдану үшін тиімді етеді. Полимер-балшық матрицаларына металл нанобөлшектерін (Pd, Ni, Cu) енгізу каталитикалық белсенділікті айтарлықтай арттырып, тотығу, тотықсыздану және энергия түрлендіру процестерін тиімді жүргізуге мүмкіндік береді. Алайда, полимерлердің деградациясы, катализаторлардың дезактивациясы және оларды өнеркәсіптік ауқымда қолдану мәселелері өзекті болып қала береді. *Қорытынды.* Металл нанобөлшектері бар гибридті полимер-балшық композиттерін дамыту жасыл химия және тұрақты катализ үшін перспективалық бағыттар ашады. Болашақ зерттеулер осы материалдардың тұрақтылығын, масштабталуын және функционализациясын арттыруға бағытталуы керек, бұл олардың өнеркәсіп пен экологиядағы практикалық қолдану аясын кеңейтуге мүмкіндік береді.

Түйінді сөздер: полимер-сазды композиттер, металл нанобөлшектері, катализ, композиттерді синтездеу, жасыл химия.

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КАТАЛИЗАТОРЫ НА ОСНОВЕ ПОЛИМЕРНО-ГЛИНИСТЫХ КОМПОЗИТОВ

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Резюме. *Введение.* История полимерно-глинистых композитов берет начало в середине XX века, когда было обнаружено, что добавление глинистых минералов, таких как монтмориллонит, в полимерные матрицы значительно улучшает их механические и термические свойства. Они находят применение в экологически чистых процессах, окислительно-восстановительных реакциях и очистке загрязнителей. Внедрение нанотехнологий позволило добиться нанодисперсии глины, увеличивая площадь поверхности и взаимодействие с полимерами. В последние годы исследования сосредоточены на экологически безопасных материалах, включая биополимерные матрицы, такие как хитозан и крахмал. *Цель работы.* Настоящий обзор направлен на анализ последних достижений в области катализаторов на основе полимерно-глинистых композитов, выявление ключевых проблем и определение перспективных направлений исследований. *Результаты и обсуждение.* Полимерно-глинистые композиты обеспечивают структурную стабильность, улучшенную механическую прочность и высокую термостойкость. Включение глин, таких как монтмориллонит или цеолиты, повышает адсорбционные свойства и ионообменную способность, делая эти материалы эффективными в катализе и экологических приложениях. Внедрение металлических наночастиц (Pd, Ni, Cu) в полимерно-глинистые матрицы значительно увеличивает каталитическую активность, позволяя эффективно проводить процессы окисления, восстановления и преобразования энергии. Однако остаются проблемы деградации полимеров, дезактивации катализаторов и масштабируемости для промышленного применения. *Выводы.* Развитие гибридных полимерно-глинистых композитов с металлическими наночастицами открывает перспективные направления для зеленой химии и устойчивого катализа. Будущие исследования должны быть сосредоточены на повышении стабильности, масштабируемости и функционализации этих материалов для расширения их практического применения в промышленности и экологии.

Ключевые слова: полимер-глинистые композиты, наночастицы металлов, катализ, синтез композитов, зеленая химия.

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