


Cite this: *RSC Adv.*, 2025, 15, 3928

Magnetically recoverable catalysts for efficient multicomponent synthesis of organosulfur compounds

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This manuscript introduces a groundbreaking study on the development and application of magnetically recoverable catalysts for the efficient multicomponent synthesis of organosulfur compounds. Capitalizing on the unique advantages of magnetic recovery, these catalysts streamline the synthesis process, offering an innovative solution that marries efficiency with environmental sustainability. By facilitating the multicomponent reaction of key precursors in the presence of sulfur sources, the catalysts enable the straightforward synthesis of various valuable organosulfur compounds, crucial in numerous pharmaceutical, agricultural, and material science applications. Key findings demonstrate a significant enhancement in reaction yields and selectivity and the remarkable ease with which the catalysts can be recovered and reused, thereby reducing both waste and operational costs. Magnetic catalysts, often based on magnetic iron nanoparticles, facilitate rapid and efficient reactions under mild

Received 16th December 2024

Accepted 22nd January 2025

DOI: 10.1039/d4ra08769k

rsc.li/rsc-advances

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conditions, offering superior atom economy, reduced solvent use, and the potential for scalable processes. Additionally, magnetically separating the catalysts from the reaction mixture enables multiple recycling cycles, reducing waste and operational costs. The review also discusses the mechanistic insights, challenges, and recent advancements in this field alongside future directions for developing more robust and versatile magnetic catalytic systems. This research embodies a significant step forward in the field of catalysis, highlighting the potential of magnetically recoverable catalysts to revolutionize the synthesis of complex molecules. Future perspectives discussed in the manuscript focus on expanding the scope of these catalysts to broader applications, optimizing catalyst design for enhanced performance, and further aligning chemical synthesis processes with the principles of green chemistry. This review covers the literature from 2010 to the end of 2024, and it encompasses the different one-pot protocols for synthesizing various heterocyclic organosulfur compounds based on magnetically recoverable catalysts.

1. Introduction

1.1. Catalysis

Multicomponent synthesis (MCS) has emerged as a transformative approach in organic synthesis, whereby multiple reactants are combined to create a single product through a one-pot reaction. This innovative methodology presents several advantages over conventional synthetic strategies. Primarily, it significantly minimizes the need for purification and isolation steps, which are often time-consuming and resource-intensive. By allowing the integration of various reactants into a single reaction vessel, MCS streamlines the overall synthesis process, saving both time and materials.^{1–3} Moreover, this synthesis approach promotes more excellent atom economy, a key principle in sustainable chemistry that emphasizes the efficient use of atoms in chemical processes. By maximizing the incorporation of reactants into the final product, MCS reduces waste generation, making it a more environmentally friendly option. These characteristics align closely with the principles of green chemistry, which aim to improve the sustainability of chemical processes by reducing toxicity and environmental impact.^{4,5}

A critical element of multicomponent reactions (MCRs) is catalysis, which plays a vital role in facilitating the formation of complex organosulfur compounds. Catalysis enhances reaction rates and selectivity, allowing reactions to occur under milder conditions that are often more efficient and safer than traditional methods. The integration of catalysis into MCRs marks a significant evolution in synthetic chemistry, as it empowers chemists to generate diverse and intricate molecular architectures with high efficiency.⁶ Recent advancements in the field underscore the utility of metal catalysts, such as rhodium and palladium—that have demonstrated exceptional effectiveness in promoting reactions involving sulfur-containing substrates. These catalysts are adept at influencing various reaction pathways and selectivity, thus enhancing the overall efficiency of the synthesis process. Their ability to operate under mild conditions is particularly noteworthy, as traditional synthetic methods frequently require harsh environments—high temperatures or extreme pressures—that may lead to undesirable side reactions or degradation of sensitive substrates.⁷

Furthermore, the strategic development of these metal catalysts has addressed many challenges historically associated with organic synthesis, thereby enabling the selective formation

of desired products. The recent trend toward using sustainable and catalytic approaches reflects a broader commitment within the chemical community to innovate while minimizing the ecological footprint of synthetic processes.⁸ Additionally, ongoing research continues to explore new catalytic systems and methodologies that will expand the repertoire of available tools for chemists, further enhancing the versatility and applicability of MCRs. Through these innovations, multicomponent synthesis is poised for ongoing growth, providing exciting opportunities for developing novel materials, pharmaceuticals, and other valuable chemical entities.^{9,10}

Catalysts are fundamental players in the realm of chemical reactions, functioning as crucial agents that significantly enhance the rate at which these reactions occur without undergoing any permanent change themselves. This unique ability to facilitate reactions without being consumed is what sets catalysts apart from other reactants.¹¹ Catalysts operate by providing an alternative reaction pathway, which possesses a dramatically lower activation energy compared to the standard uncatalyzed process. Activation energy is the energy barrier that must be overcome for reactants to be converted into products, and by lowering this barrier, catalysts enable reactions to proceed more readily.^{12–14} This means that at any given temperature, a larger proportion of the reactant molecules can achieve the energy necessary to reach the transition state, the critical point at which reactants can transform into products. Consequently, this increased accessibility to the transition state results in higher reaction rates, allowing for a more efficient conversion of starting materials into desired products.^{15,16}

The significance of catalysts in chemistry cannot be overstated. Their essential role is evident across a vast array of industrial processes, where they expedite reactions and enhance process efficiency.^{17,18} In various sectors, including petrochemicals, pharmaceuticals, and materials science, catalysts contribute to synthesizing a wide range of substances, from everyday items like plastics and fuels to more complex compounds used in drug development.¹⁹ Catalysts play a crucial role in conserving energy by enabling reactions to occur more rapidly or at reduced temperatures. This is particularly important in global sustainability efforts, as reducing energy consumption directly correlates with decreased environmental impact. Additionally, using catalysts leads to substantial cost savings in industrial operations.²⁰ By allowing for more efficient reactions, companies can optimize resource use, minimize



waste, and achieve greater throughput, all of which translate into lower production costs and enhanced profitability. The influence of catalysts extends beyond mere economic benefits; they are pivotal in driving innovation and efficiency within the field of chemical manufacturing. The ongoing development and optimization of new catalytic processes open doors to novel reactions and applications, often allowing chemists to explore previously untenable synthetic pathways.²¹

Catalysts play a transformative role in green chemistry, fundamentally altering chemical reaction approaches to foster more environmentally sustainable methods.²² This shift toward sustainability is crucial in addressing the environmental challenges faced by the chemical industry, where traditional processes often result in excessive waste and pollution. By facilitating reactions that generate significantly less waste and capitalize on renewable resources, catalysts embody the core principles of sustainability, promoting processes that are efficient and low in environmental impact.^{23–25} In nature, enzymes serve as natural catalysts indispensable for life. These remarkable proteins catalyze an array of complex biochemical reactions essential for the functioning of living organisms, such as digestion, metabolism, and DNA replication.²⁶ Enzymes excel in their specificity and efficiency, showcasing nature's ability to fine-tune reactions in a way that supports life. This inherent ability to accelerate reactions is crucial for maintaining vital biological processes, demonstrating the importance of catalysts beyond the confines of the laboratory.^{27,28}

Another essential characteristic of catalysts is their selectivity, which refers to their capacity to direct chemical reactions toward the formation of specific desired products while minimizing the production of unwanted byproducts.²⁹ This selectivity is particularly valuable in the pharmaceutical industry, where the demand for high-purity substances is paramount for producing effective medications. High selectivity ensures that the desired therapeutic compounds are generated in reliable quantities and reduces the cost and complexity associated with purifying reaction mixtures, which can significantly enhance the overall efficiency of drug development processes.³⁰ Furthermore, catalysts are at the forefront of groundbreaking research in renewable energy. They play a pivotal role in developing innovative technologies, such as solar fuels, where catalysts facilitate the conversion of sunlight into energy-storing molecules, mimicking photosynthesis. This area holds immense potential as a sustainable energy source, directly contributing to the quest for cleaner energy alternatives.³¹ Additionally, catalysts are instrumental in pioneering processes to convert carbon dioxide—a significant greenhouse gas—into beneficial chemicals and fuels, effectively recycling emissions into valuable products.³² By enabling these transformations, catalysts not only enhance efficiency in energy production but also play a crucial role in efforts to combat climate change and minimize our carbon footprint.³³ The positive impact of catalysts on sustainable practices extends to various fields, including waste treatment, sustainable agriculture, and the production of biodegradable materials. Their ability to streamline reactions while conserving resources makes them

integral to developing more sustainable processes that align with global environmental goals.

The primary objective of this review manuscript is to explore and critically analyze the recent advancements in the field of magnetically recoverable catalysts engineered explicitly for the efficient multicomponent synthesis of organosulfur compounds. To achieve this, the following key objectives have been established:

- **Comprehensive overview:** provide a detailed overview of the current state of research in magnetically recoverable catalysts, highlighting their structural designs, functional principles, and material compositions that facilitate their application in synthesizing organosulfur compounds.
- **Synthesis strategies:** examine the various synthetic strategies and reaction pathways employed in multicomponent reactions involving magnetically recoverable catalysts, emphasizing how these methods enhance reaction efficiency, selectivity, and yield compared to traditional catalytic approaches.
- **Environmental and economic benefits:** evaluate the environmental and economic advantages offered by these catalysts, including their contribution to greener synthesis processes through recyclability, reduced energy consumption, and diminished chemical waste.
- **Challenges and limitations:** identify the current challenges, limitations, and technical barriers associated with using magnetically recoverable catalysts in organosulfur compound synthesis, providing a balanced perspective on areas requiring further research and development.
- **Future directions:** propose future research directions and innovative approaches to overcome existing challenges, enhance catalytic performance, and broaden the applicability of these catalysts in diverse chemical synthesis processes.
- **The review aims to synthesize current knowledge, inspire continued innovation in the field, and reinforce the significance of magnetically recoverable catalysts in developing sustainable and efficient chemical synthesis methodologies.**

In summary, catalysts are vital in accelerating chemical reactions, enhancing selectivity, saving energy, and enabling sustainable industrial processes. Their use spans across various sectors, making them indispensable tools in modern chemistry and industry. The ongoing research and development of new and improved catalysts continue to open up possibilities for future innovations and advancements in science and technology.

1.2. Magnetic catalysts

Magnetic catalysts have emerged as a groundbreaking advancement in the realm of green chemistry and modern catalyst science, presenting a sustainable and environmentally-friendly strategy for facilitating chemical reactions (Fig. 1).^{34,35} Typically constructed from metal complexes that are meticulously supported on magnetic nanoparticles, these innovative catalysts offer the unique advantage of being easily separable from the reaction mixture.³⁶ By applying an external magnetic field, the catalysts can be swiftly removed without the need for traditional separation methods, such as filtration or centrifugation.^{37,38} This simplified separation process not only



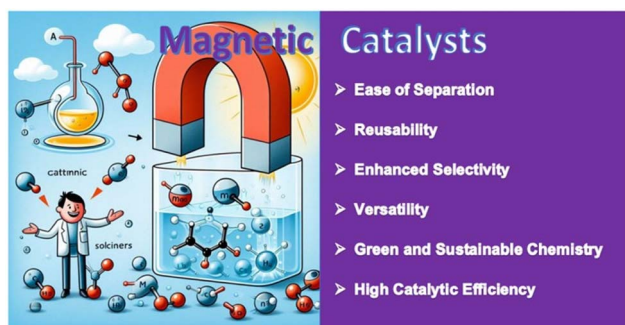


Fig. 1 Highlights of magnetic catalysts.

significantly diminishes the consumption of solvents and energy but also substantially reduces waste generation, making it a key player in promoting sustainable practices.³⁹ The design and operation of magnetic catalysts align seamlessly with the central tenets of green chemistry, which prioritize minimizing environmental impact while enhancing safety for both the processes involved and the products produced.^{40,41} Through this integration of efficiency and sustainability, magnetic catalysts are paving the way for a more responsible approach to chemical synthesis and industrial applications.⁴²

The reusability of magnetic catalysts presents a crucial advantage in contemporary chemistry, as these innovative materials can be effectively recovered and employed multiple times without experiencing a significant decline in their catalytic activity.⁴³ This characteristic not only enhances cost-effectiveness but also promotes resource conservation in chemical processes.⁴⁴ In the realm of modern catalyst science, the design and synthesis of magnetic catalysts are meticulously crafted to elevate their catalytic performance, stability, and selectivity.^{45–48} Cutting-edge advancements in material science have paved the way for the development of magnetic nanocatalysts, which possess tailored surface properties and functionalities.⁴⁹ These properties can be precisely adjusted to optimize their effectiveness for specific reactions, thereby broadening the potential applications of magnetic catalysts across a diverse array of chemical transformations.^{50,51}

Moreover, magnetic catalysts facilitate the development of novel reaction pathways and mechanisms, potentially leading to more efficient and less energy-intensive processes.⁵² For instance, the magnetic properties of these catalysts can influence reaction kinetics and dynamics, offering a unique control over the reaction by simply applying an external magnetic field.⁵³ This aspect opens up new possibilities for catalysis, such as magnetic field-driven technologies that can change the catalytic output or enable the use of magnetic fluidized-bed reactors for improved mass and heat transfer.⁵⁴ In the pharmaceutical industry, magnetic catalysts have been instrumental in the synthesis of valuable compounds with significant medicinal applications.⁵⁵ The ability to perform reactions under milder conditions and with higher yields is particularly beneficial for the production of pharmaceuticals, where purity and efficacy are paramount.⁵⁶ The environmental benefits of magnetic catalysts are also noteworthy, as they support the shift towards greener processes in the

chemical industry.⁵⁷ For example, they have been used to enhance hydrogen production and water splitting, which are critical for the development of clean energy technologies.⁵⁸

The versatility and efficiency of magnetic catalysts are not limited to organic synthesis but also extend to material science, where they play a crucial role in the production of dyes, plasticizers, and fiber-reinforced composite materials.^{59,60} Their application in creating vulcanization accelerators for the rubber industry further exemplifies their industrial significance. In summary, magnetic catalysts represent a transformative approach in green chemistry and modern catalyst science, offering a harmonious blend of environmental stewardship, economic viability, and scientific innovation.

1.3. Magnetic catalysts in comparison with conventional catalysts

Magnetic nanocatalysts offer several advantages over conventional catalysts, making them increasingly popular in various chemical processes. Here are some key benefits:

- ✓ **Ease of separation:** magnetic nanocatalysts can be easily separated from reaction mixtures using an external magnetic field. This eliminates the need for filtration or centrifugation, making the process cleaner and more efficient.⁶¹
- ✓ **Reusability:** due to their magnetic properties, magnetic catalysts can be recovered and reused multiple times without significant loss of activity. This enhances their cost-effectiveness and sustainability.³⁵
- ✓ **High surface area:** magnetic nanocatalysts have a high specific surface area, which increases the number of active sites available for reactions. This leads to higher catalytic activity and efficiency compared to conventional catalysts.⁶²
- ✓ **Enhanced selectivity:** the high surface area and the ability to functionalize the surface of magnetic nanocatalysts allow for better control over reaction pathways, improving the selectivity of the desired products.⁶³
- ✓ **Environmental benefits:** magnetic separation avoids the use of additional chemicals and solvents, reducing the environmental impact of the catalytic process. This makes the process greener and more sustainable.⁶⁴
- ✓ **Versatility:** magnetic nanocatalysts can be used in a wide range of reactions, including biofuel production, organic synthesis, and environmental remediation.⁶⁵

In summary, magnetic nano catalysts offer improved efficiency, selectivity, and reusability, along with environmental benefits over conventional catalysts.⁶⁶ These properties make them an attractive option for modern chemical processes, driving the continuous advancements in catalysis and green chemistry.⁶⁷ The ongoing development of magnetic nanocatalysts is expected to further enhance their performance and application scope, solidifying their role in the future of catalytic science.

1.4. Methods for the construction of magnetic catalysts

There are several methods for preparing magnetic catalysts, each with its own advantages and applications. Here are some of the most common methods:



✓ **Co-precipitation:** this is one of the simplest and most widely used methods. It involves the simultaneous precipitation of magnetic nanoparticles from a solution containing metal salts, usually under basic conditions. This method is cost-effective and can produce large quantities of nanoparticles.⁶⁸

✓ **Thermal decomposition:** this method involves the decomposition of organometallic compounds at high temperatures in the presence of surfactants. It allows for precise control over the size and shape of the nanoparticles, resulting in highly uniform particles.⁶⁹

✓ **Hydrothermal and solvothermal methods:** these methods use high-temperature and high-pressure conditions to synthesize nanoparticles in aqueous or non-aqueous solutions. They are particularly useful for producing well-crystallized and highly pure nanoparticles.⁶³

✓ **Sol-gel method:** this involves the transition of a system from a liquid “sol” (mostly colloidal) into a solid “gel” phase. It is versatile and can be used to produce a wide range of materials, including magnetic nanoparticles.³⁹

✓ **Microemulsion:** this method uses microemulsions as nanoreactors to control the size and shape of the nanoparticles. It is particularly useful for producing monodisperse nanoparticles with narrow size distributions.³²

✓ **Ball milling:** this mechanical method involves grinding bulk materials into fine powders using a ball mill. It is a straightforward and scalable method, though it may result in a broad size distribution of particles.⁷⁰

✓ **Biological methods:** these involve the use of biological organisms or biomolecules to synthesize nanoparticles. This eco-friendly approach can produce nanoparticles with unique properties, though it is still under extensive research.⁴²

The preparation of magnetic catalysts involves various methods, each offering specific advantages in terms of control over particle size, composition, stability, and catalytic activity.^{71,72} The choice of method depends on the desired properties of the final catalyst, the type of reaction, and the scale of production.⁷³ Regardless of the method, magnetic catalysts are particularly advantageous due to their ability to be easily separated and recycled, making them ideal for many catalytic processes, including environmental and industrial applications.

1.5. Multicomponent reactions

Multicomponent reactions (MCRs) are a powerful and increasingly important class of reactions in modern chemistry, particularly in the context of green chemistry.⁷⁴ These reactions involve the simultaneous combination of three or more reactants to form a complex product, typically in a single reaction vessel and under relatively simple conditions.⁷⁵ Here are several reasons why MCRs are beneficial:

1.5.1. Efficiency and atom economy. ✓ **Reduced waste:** MCRs are known for their high atom economy, meaning that the atoms from the reactants are effectively incorporated into the final product with minimal waste. This reduces the generation of byproducts and waste materials, making MCRs more sustainable.⁷⁶

✓ **Fewer steps:** instead of performing multiple individual reactions to form a desired product, MCRs allow chemists to assemble complex molecules in a single step. This reduces the number of synthetic steps, simplifying workflows and saving both time and resources.⁷⁷

1.5.2. Cost-effectiveness. ✓ **Reduced reagents and solvents:** since MCRs typically require fewer reagents, solvents, and steps compared to traditional methods, they help reduce costs in terms of both materials and energy. This is especially important in industrial applications where the cost of reagents and solvents can be a significant factor.⁷⁸

✓ **Simplified purification:** fewer steps often mean fewer intermediate compounds, leading to simpler purification processes, which further reduces costs and environmental impact.⁷⁹

1.5.3. Diversity of products. MCRs allow for the synthesis of a broad range of complex molecules with varied functional groups and structural diversity.⁸⁰ This makes them invaluable in fields like drug discovery, materials science, and natural product synthesis, where structural complexity is often essential.⁸¹ By varying the choice of reagents and conditions, different products can be obtained in a single reaction setup, offering flexibility and versatility.⁸²

1.5.4. Environmental benefits (green chemistry). ✓ **Less energy consumption:** MCRs often proceed under mild conditions (ambient temperature, solvent-free conditions, *etc.*), which minimizes the need for excessive heating or pressure. This reduces energy consumption, making the process more environmentally friendly.⁷⁵

✓ **Safer processes:** MCRs can sometimes replace hazardous reagents or energy-intensive processes with safer alternatives. Additionally, the use of fewer solvents reduces the potential for harmful solvent waste and emissions.⁷⁵

✓ **Reduced use of toxic reagents:** the efficiency of MCRs can enable the use of more benign reagents, thereby minimizing the generation of harmful byproducts and reducing environmental impact.⁸²

1.5.5. Complexity from simplicity. MCRs provide a way to synthesize complex molecules from simple starting materials. This concept is particularly valuable in the synthesis of pharmaceuticals, agrochemicals, and specialty materials, where the complexity of the product is often difficult to achieve through traditional synthetic methods.⁸³

1.5.6. High stereoselectivity and regioselectivity. Many MCRs offer good control over stereochemistry (spatial arrangement of atoms in molecules) and regioselectivity (selectivity for certain positions on a molecule).⁸⁴ This makes them suitable for the precise construction of molecules required in the synthesis of biologically active compounds or materials with specific properties.⁷⁶

1.5.7. Potential for automation and scalable synthesis. Due to their efficiency and the fact that they often require fewer steps and reagents, MCRs are well-suited for automation, which can streamline large-scale production processes. This is advantageous in industrial settings, where the scalability of a synthetic route is crucial.⁸⁵



In summary, multicomponent reactions are a cornerstone of modern synthetic chemistry, offering multiple benefits that align well with the principles of green chemistry. They provide more efficient, cost-effective, and environmentally friendly synthetic routes, reduce waste, and enable the synthesis of complex molecules. As a result, MCRs are crucial not only in academic research but also in the development of sustainable industrial processes.

1.6. Organosulfur compounds

Organosulfur compounds are a broad class of compounds that contain carbon (C) and sulfur (S) atoms bonded together, and they have significant medicinal, biological, therapeutic, and industrial importance due to their unique chemical properties.^{86–89} These compounds are present in many natural substances and have a wide range of applications in various fields.

1.6.1. Medicinal importance. Organosulfur compounds are of great interest in the pharmaceutical industry because they exhibit diverse bioactivity.⁹⁰ Some well-known organosulfur compounds used in medicine include:

- ✓ **Antibiotics:** many organosulfur compounds are essential in the development of antibiotics. A classic example is penicillin, where the sulfur atom plays a crucial role in its antibacterial activity by inhibiting bacterial cell wall synthesis.^{86,91}

- ✓ **Anticancer agents:** compounds like sulforaphane, found in cruciferous vegetables (such as broccoli), have shown promising anticancer properties by inducing phase II detoxifying enzymes and inhibiting carcinogenic activity. Other organosulfur compounds, like diallyl disulfide (from garlic), have also demonstrated potential anticancer effects in preclinical studies.^{92,93}

- ✓ **Antioxidant activity:** some organosulfur compounds possess antioxidant properties, which can help protect cells from oxidative stress. These include compounds like alpha-lipoic acid, which is involved in mitochondrial energy production and acts as a powerful antioxidant.^{94,95}

- ✓ **Antiviral and antimicrobial:** garlic-derived organosulfur compounds, such as allicin, have been shown to possess broad-spectrum antimicrobial and antiviral activities. These compounds are thought to inhibit the growth of pathogens through their ability to modify the thiol groups in microbial enzymes and proteins.^{96,97}

- ✓ **Cardiovascular health:** organosulfur compounds, particularly from garlic (ajoene), can have cardiovascular benefits by reducing blood pressure, lowering cholesterol, and improving circulation.⁹⁸

1.6.2. Biological importance. Organosulfur compounds are crucial in various biological processes. Some examples include:

- ✓ **Amino acids and proteins:** certain amino acids that contain sulfur, such as cysteine and methionine, are essential for protein synthesis, enzyme function, and cellular metabolism.^{99,100} These sulfur-containing amino acids are involved in the formation of disulfide bonds that stabilize protein structures.¹⁰¹

- ✓ **Vitamins:** thiamine (vitamin B1) and biotin (vitamin B7) are examples of vitamins that contain sulfur atoms in their

structures. These vitamins are important for energy metabolism and various enzymatic processes in the body.¹⁰²

- ✓ **Enzyme co-factors:** several important coenzymes like coenzyme A and lipoic acid contain sulfur. These molecules are involved in key metabolic processes, such as the citric acid cycle and fatty acid metabolism.¹⁰³

- ✓ **Detoxification:** organosulfur compounds, especially glutathione (a tripeptide composed of glutamine, cysteine, and glycine), play a critical role in cellular detoxification.¹⁰⁴ Glutathione is a key antioxidant and detoxifier, helping neutralize reactive oxygen species and other harmful molecules in the body.

1.6.3. Therapeutic applications. Organosulfur compounds are increasingly recognized for their therapeutic properties, especially in alternative and complementary medicine. Some examples of therapeutic uses include:

- ✓ **Anti-inflammatory:** organosulfur compounds like diallyl sulfide (from garlic) have anti-inflammatory effects and are thought to modulate the immune response. They may help reduce inflammation in conditions such as arthritis.^{105,106}

- ✓ **Antidiabetic:** certain organosulfur compounds show potential in managing diabetes. For example, sulforaphane has been studied for its ability to improve insulin sensitivity and reduce blood glucose levels.¹⁰⁷

- ✓ **Neurological health:** organosulfur compounds, particularly alpha-lipoic acid, have neuroprotective effects. Lipoic acid has been used as a supplement for managing neurodegenerative diseases like Alzheimer's disease due to its ability to reduce oxidative stress and improve mitochondrial function.¹⁰⁸

- ✓ **Detoxification:** compounds such as sulforaphane and cysteine (often in the form of *N*-acetylcysteine) are used therapeutically to support detoxification processes in the liver and improve overall cellular health.¹⁰⁹

Overall, the multifaceted importance of aryl sulfides underscores their value in advancing health and industrial processes (Fig. 2).^{110–115}

1.6.4. Industrial importance. Organosulfur compounds have widespread applications in various industrial sectors due to their unique chemical reactivity, odor characteristics, and other functional properties:

- ✓ **Petrochemical industry:** in the oil and gas industry, sulfur is a significant byproduct. Organosulfur compounds, such as thiols and sulfides, are found in crude oil and natural gas and need to be removed during the refining process due to their undesirable odors and their role in causing corrosion. Sulfur compounds are also used in the production of hydrogen sulfide (H₂S) and sulfuric acid (H₂SO₄), both of which are crucial in a wide range of chemical processes.^{116,117}

- ✓ **Agriculture:** organosulfur compounds, such as dimethyl disulfide and methyl isothiocyanate, are used as fumigants and pesticides. These compounds are especially effective at controlling pests and fungi in agricultural settings.¹¹⁸

- ✓ **Rubber industry:** in the production of synthetic rubber, sulfur is used in a process called vulcanization. Sulfur is used to crosslink the rubber molecules, making the material more durable, elastic, and heat-resistant.¹¹⁹



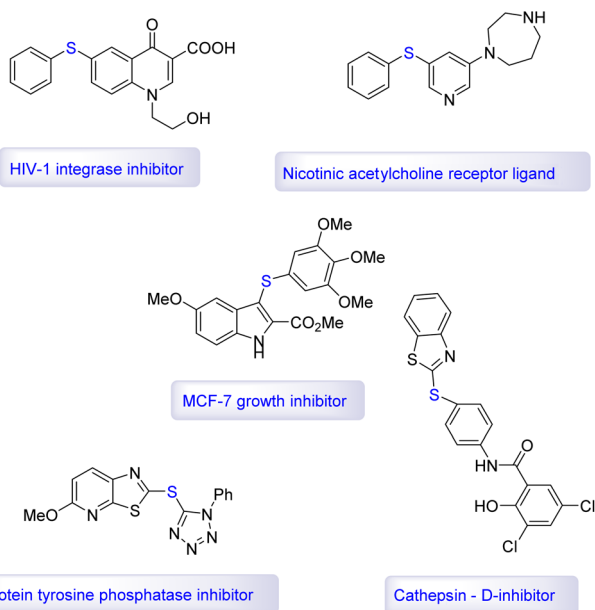


Fig. 2 Some important molecules containing sulfide scaffold.

✓ **Food industry:** some organosulfur compounds, such as those in garlic and onions (*e.g.*, allyl sulfides), are used to flavor food and are known for their health benefits. Additionally, sulfur-containing preservatives like sulfur dioxide are used to prevent spoilage in dried fruits, wines, and other food products.¹²⁰

✓ **Cosmetic industry:** organosulfur compounds are also utilized in cosmetics for their antioxidant, antimicrobial, and anti-aging properties. For example, sulfur is used in topical treatments for skin conditions like acne and seborrheic dermatitis.¹²¹

In summary, organosulfur compounds are an essential class of chemicals with a wide range of applications across various fields, from medicine to industry. Their biological and medicinal properties, including anticancer, antimicrobial, and antioxidant activities, make them valuable in therapeutic contexts. Additionally, their utility in industrial processes like petroleum refining, rubber production, and agriculture highlights their versatility and importance in daily life. As research continues, it is likely that new uses and benefits of organosulfur compounds will emerge, further cementing their significance in modern science and industry.^{122–125}

2. Magnetic catalysts in the multi-component preparation of organosulfur compounds

The field of chemical synthesis is perpetually evolving, with significant emphasis on enhancing catalytic processes' efficiency, selectivity, and sustainability. Magnetically recoverable catalysts have emerged as a pivotal innovation in this realm, particularly for the multicomponent synthesis of organosulfur compounds. These catalysts combine the reactive versatility

required for complex chemical transformations with magnetic recovery's environmental and operational benefits.^{71–75}

Magnetically recoverable catalysts are designed to offer a straightforward, economical, and eco-friendly approach to chemical synthesis. These catalysts typically consist of a magnetic core material, such as iron oxide (Fe_3O_4), functionalized with various catalytic active sites. The core-shell structure enables the catalyst to participate actively in the reaction to form organosulfur compounds while allowing for an easy magnetic separation process post-reaction, thereby eliminating the need for filtration or centrifugation.^{76–80}

Key advantages in multicomponent syntheses:

- **Enhanced catalytic activity:** magnetic catalysts often display superior catalytic activity due to the high surface area and easy accessibility of active sites on the magnetic nanoparticles.
- **Selectivity and yield:** these catalysts facilitate improved selectivity and higher yields in multicomponent reactions due to their tailored surface chemistry, which can be engineered to favor specific reaction pathways.
- **Operational efficiency:** these catalysts' magnetic properties allow for quick and easy recovery using an external magnetic field, significantly reducing downtime between batches and minimizing catalyst loss.
- **Reusability:** one of the standout features of magnetically recoverable catalysts is their reusability, which significantly reduces cost and environmental impact by reducing chemical waste and the need for fresh catalysts (Fig. 3).

The use of magnetic catalysts in the multicomponent synthesis of organosulfur compounds is a promising approach that combines the efficiency of MCRs with the benefits of magnetically separable catalysts. This method improves sustainability, reduces environmental impact, and offers practical advantages in terms of catalyst recovery and reuse. By leveraging the unique properties of magnetic materials, researchers can optimize reaction conditions and broaden the scope of organosulfur compound synthesis. Continuing our research on magnetic catalysts, this review explores the

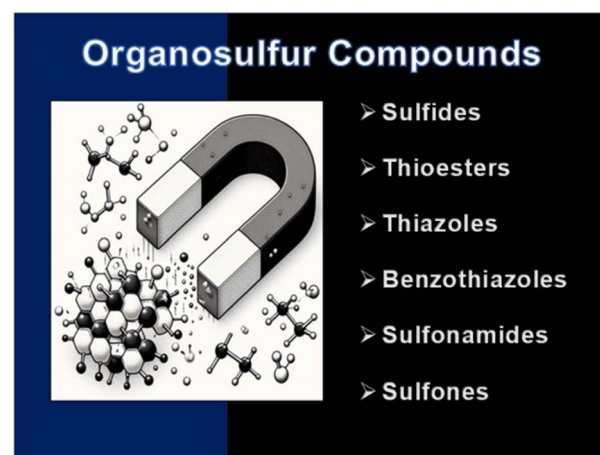


Fig. 3 Magnetic catalysts in multicomponent preparation of organosulfur compounds.



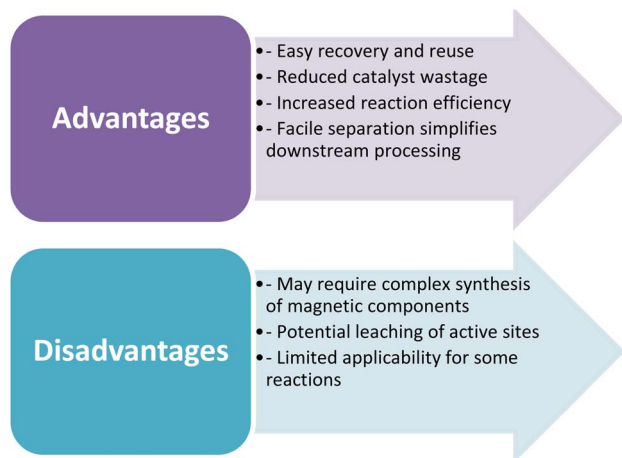


Fig. 4 The advantages and disadvantages of magnetically recoverable catalysts.

advantages of employing magnetic catalysts in the preparation of multicomponent organosulfur compounds, emphasizing their enhanced catalytic activity, ease of separation, and environmental sustainability. The advantages and disadvantages of using magnetic catalysts can be summarized in Fig. 4.^{75–85}

2.1. Environmental implications of magnetically recoverable nanocatalysts

Integrating environmentally sustainable practices within chemical synthesis is increasingly important in contemporary research. Magnetically recoverable nanocatalysts, particularly in synthesizing organosulfur compounds through multicomponent reactions, exemplify a forward-thinking approach to merging catalytic efficiency with environmental stewardship. These innovative catalysts streamline synthetic processes and significantly mitigate the ecological footprint associated with traditional chemical manufacturing.^{40–45}

Magnetically recoverable nanocatalysts are engineered with magnetic properties that facilitate easy separation and recovery using a magnetic field, thus dramatically reducing waste and eliminating the need for chemical solvents or extensive energy inputs typically used in catalyst recovery. This feature is pivotal in minimizing the environmental impact traditionally associated with catalyst filtration, washing, reduction of hazardous waste, energy conservation, and disposal processes.^{46–50}

Research demonstrates that magnetically recoverable nanocatalysts sustain high catalytic activity and promote greener reaction environments. For instance, in synthesizing thiophenes and thiazoles, which are important organosulfur compounds, magnetic nanocatalysts have been shown to facilitate reactions efficiently at room temperature, thereby conserving energy that would otherwise be needed for heating. Furthermore, these catalysts can often operate in water or other green solvents, which are significantly less toxic and more biodegradable than the organic solvents typically used in such syntheses. This reduces VOC emissions and decreases the risk of environmental contamination associated with solvent use.^{51–56}

Magnetically recoverable nanocatalysts represent a significant breakthrough in green chemistry, particularly for synthesizing organosulfur compounds through multicomponent reactions. By reducing waste, conserving energy, and facilitating the use of greener solvents, these catalysts contribute positively to environmental conservation efforts. Ongoing research and development will be key to overcoming existing challenges and unlocking the full potential of these catalysts in sustainable industrial chemistry.^{57–60}

2.2. Challenges directions

(1) Catalyst stability and degradation: one of the primary concerns with magnetically recoverable catalysts is their stability under typical reaction conditions. Repeated magnetic recovery and exposure to reactive environments can degrade the catalyst, reducing its effectiveness over time. Research is required to develop more robust materials to withstand the mechanical and chemical stresses of multiple reaction cycles.

(2) Magnetic saturation and aggregation: due to magnetic attraction, magnetized nanoparticles tend to aggregate, which can decrease the surface area available for reactions and, consequently, reduce catalytic activity. Addressing issues of nanoparticle aggregation and ensuring uniform dispersion during the reaction is crucial for maintaining the high efficiency of these catalysts.

(3) Leaching of active species: leaching metal or active sites from the catalyst surface into the reaction medium is a significant challenge. It diminishes the catalyst's efficacy and contaminates the product, complicating purification processes. Innovative anchoring strategies and stronger chemical bonds between the active sites and magnetic support are needed.^{115–120}

2.3. Future directions

(1) Advanced material engineering: research into new materials that can enhance the durability and reusability of magnetically recoverable catalysts is essential. Exploring alloys or composite materials that resist corrosion and physical wear could extend the life and efficiency of these catalysts.

(2) Innovative catalyst systems: developing 'smart' catalysts that can adjust their activity in response to environmental changes or specific reaction stages could revolutionize their application in complex syntheses. Incorporating stimuli-responsive materials or molecular switches could allow for more precise control over the catalytic process.

(3) Scale-up and commercialization: scaling up the production and use of magnetically recoverable catalysts from laboratory to industrial scale remains challenging. Studies focused on the engineering aspects of catalyst manufacture, including the design of large-scale magnetic separation systems and continuous flow reactors fitted with magnetic recovery units, will be critical.

(4) Environmental impact assessment: comprehensive life-cycle assessments are necessary to evaluate the environmental impacts associated with the production, use, and disposal of magnetically recoverable catalysts. These studies will help identify potential environmental risks and guide the development of practices to mitigate them.



(5) Integration with renewable energy sources: using magnetically recoverable catalysts with renewable energy sources in chemical syntheses could further enhance their environmental profile. Research into systems that utilize solar or wind energy to drive reactions catalyzed by these advanced materials could set new benchmarks for sustainability in the chemical industry.^{115–120}

2.4. MCRs preparation of sulfides

The three-component synthesis of aryl sulfides from a sulfur source using magnetic catalysts is a fascinating area of research in organic chemistry, particularly due to the importance of aryl sulfides in pharmaceuticals and materials science.^{126–130} This process typically involves the use of a magnetic nanocatalyst, which facilitates the formation of the C–S bond through a coupling reaction.¹³¹ The use of magnetic catalysts offers several advantages, including easy separation by an external magnetic field and recyclability, which makes the process more environmentally friendly and cost-effective. Additionally, the presence of a base can enhance the catalytic activity and suppress side reactions, leading to higher yields of the desired aryl sulfides. This method represents a significant advancement in the field of synthetic chemistry, providing a more efficient route to synthesize valuable sulfur-containing compounds (Fig. 5).

In a pioneering study, Chang and his research group unveiled a novel catalyst system utilizing Fe₃O₄-supported 3-amino-4-mercaptobenzoic acid copper complex (Fe₃O₄@AMBA-CuI) magnetically recoverable catalyst [MRC-1] (Fig. 4).¹³² This innovative catalyst proved to be both effective and magnetically recoverable, facilitating the synthesis of a wide array of heteroaryl-aryl and di-heteroaryl sulfides. The method involves reacting various heteroaryl halides with aryl or heteroaryl boronic acids, utilizing elemental sulfur (S₈) as the source of sulfur, all under environmentally-friendly conditions. The catalytic efficiency of this system is particularly noteworthy, as it

successfully accommodated a diverse range of heteroaryl substrates, including challenging compounds like benzothiazole, benzoxazole, benzimidazole, oxadiazole, benzofuran, and imidazo[1,2-*a*]pyridine. Remarkably, the desired diaryl and di-heteroaryl sulfides were synthesized in high yields, showcasing the utility and versatility of this approach. While the products synthesized—heteroaryl-aryl and di-heteroaryl sulfides—are known compounds with previous reports, the scale and variety achieved in this methodology are unprecedented and have not been documented by any other synthetic routes (Scheme 1). This achievement not only highlights the effectiveness of the catalyst but also contributes significantly to the field of organic synthesis. Reusability tests confirmed that the Fe₃O₄@AMBA-CuI catalyst [MRC-1] (Fig. 4) has high catalytic activity despite being reused 6 times. To assess the stability and performance of the Fe₃O₄@AMBA-CuI nanocatalyst, various techniques such as VSM and ICP-OES techniques were employed. These analyses demonstrated that even after six cycles of use, the catalyst retained its magnetic properties and overall stability, which is crucial for practical applications.

The three-component synthesis of aryl sulfides through C–H bond sulfonylation is a significant advancement in organic chemistry, allowing for the creation of sulfide compounds *via* a direct and selective functionalization process.¹³³ This method typically involves the reaction of an aromatic precursor with a sulfur dioxide surrogate and a reagent that facilitates the formation of the C–S bond.¹³⁴ The process is advantageous due to its step and atom economy, often proceeding under mild conditions, which is crucial for maintaining the integrity of sensitive molecules.¹³⁵ Recent developments have even enabled asymmetric sulfonylation, providing access to chiral sulfones with high regio- and enantioselectivity.

In a research publication authored by Saleh Shafik and his research team, the use of CuFe₂O₄ nanoparticles as a highly

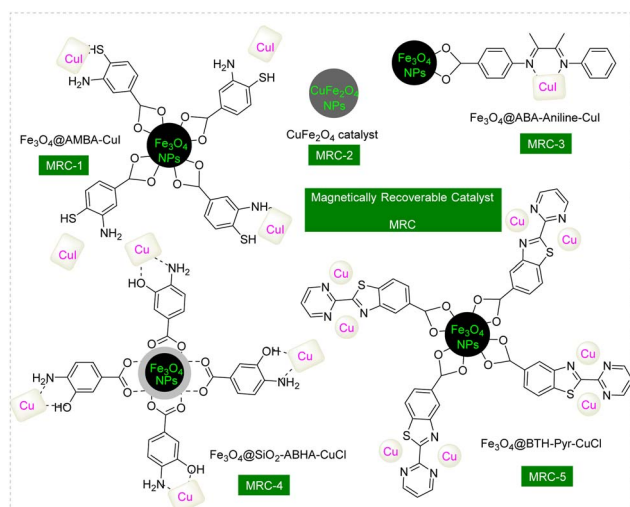
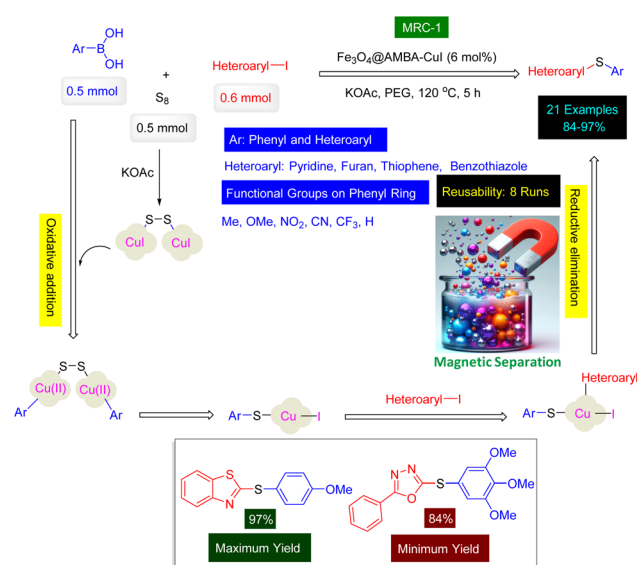


Fig. 5 Structure of several magnetic catalysts in preparation of sulfides.



Scheme 1 Fe₃O₄@AMBA-CuI (MRC-1) catalyzed synthesis of heteroaryl-aryl and di-heteroaryl sulfides from S₈.

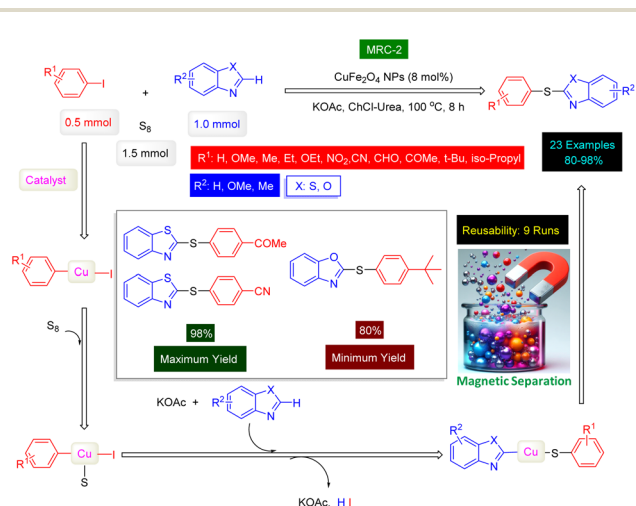


effective nanocatalyst for the synthesis of 2-thio-benzothiazoles and 2-thioaryl-benzoxazoles in CHCl_3 -urea was detailed. The size distribution of CuFe_2O_4 particles was assessed using TEM analysis, revealing an average nanoparticle diameter of 15 nm. The results clearly demonstrate that the CuFe_2O_4 NPs/ CHCl_3 -urea system, in the presence of KOAc, serves as a versatile and effective catalytic platform for the one-pot three-component coupling reaction involving benzoxazoles or benzothiazoles with aryl iodides and S_8 as the sulfur donor.¹³⁶ It is important to note that substituents on the aromatic ring do not significantly influence the reaction progress. Following this mechanism, the initial reaction between the aryl halide and the catalyst leads to the formation of an intermediate. Subsequently, the addition of S_8 produces another intermediate (Scheme 2). In the next step, this intermediate reacts with either benzoxazole or benzothiazole in the presence of a base, yielding an additional intermediate. Finally, the compound is synthesized from this intermediate through a reductive elimination step. Reusability tests confirmed that the CuFe_2O_4 catalyst [MRC-2] has high catalytic activity despite being reused 9 times.

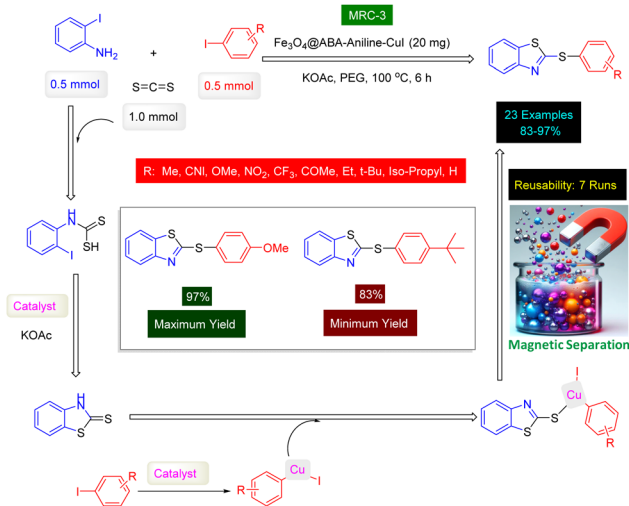
The Fe_3O_4 @ABA-aniline-CuI nanocomposite operates as a highly effective and reusable nanocatalyst, playing an essential role in the production of benzothiazole-sulfide compounds including both aryls and heteroaryls. This innovative material demonstrates remarkable catalytic activity, making it a valuable tool in organic synthesis processes. The Fe_3O_4 @ABA-aniline-CuI nanocomposite [MRC-1] (Fig. 4) was expertly characterized using a range of advanced techniques. SEM and TEM images convincingly demonstrated that the particles are spherical and optimally sized, measuring around 12 to 25 nanometers.¹³⁷ The incorporation of both the catalyst and the base was crucial for successfully synthesizing the desired product. In a one-pot, three-component reaction, 2-iodoaniline was reactively combined with carbon disulfide and a variety of aryl or heteroaryl iodides. This reaction was efficiently catalyzed by the Fe_3O_4 @ABA-aniline-CuI nanomaterial, while potassium acetate (KOAc) acted as the base, with the reaction occurring in

PEG-400 serving as the solvent (refer to Scheme 3 for a visual representation). This approach not only streamlines the synthesis process but also enhances overall efficiency and yield. Furthermore, Shen and his research team proposed a detailed tentative mechanism for the synthesis of benzothiazole scaffolds that integrate aryl and heteroaryl sulfides, elucidated clearly in Scheme 3 to provide deeper insight into the reaction pathway. Reusability tests confirmed that the Fe_3O_4 @ABA-aniline-CuI catalyst [MRC-3] has high catalytic activity despite being reused 7 times.

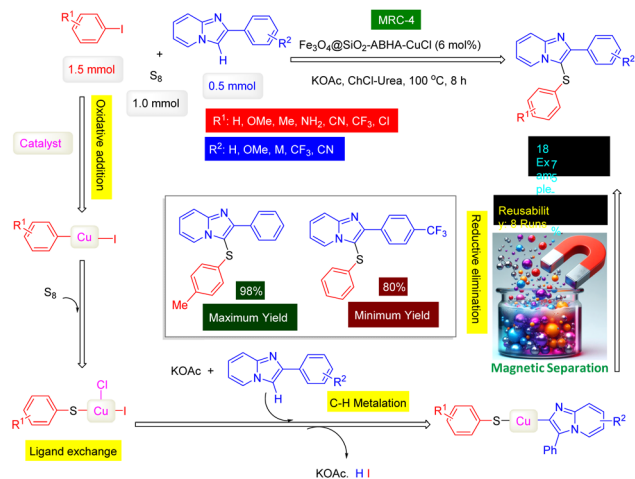
Recently, Chen made a significant advancement in green chemistry by developing an innovative, eco-friendly magnetic nanocatalyst, designated as Fe_3O_4 @ SiO_2 -ABHA-CuCl nanocomposite (MRC-4) (Fig. 4). This cutting-edge catalyst facilitates the efficient synthesis of diaryl sulfides infused with intricate imidazo[1,2-*a*]pyridine scaffolds, showcasing its potential in organic transformations.¹³⁸ The synthesis of this remarkable catalyst involved the strategic immobilization of copper(II) chloride (CuCl) onto magnetic Fe_3O_4 nanoparticles, which were artfully modified using 4-amino-3-hydroxybenzoic acid. Characterization techniques revealed that the resulting nanoparticles exhibited a captivating spherical morphology, with sizes ranging from 15 to 30 nanometers, which is ideal for maximizing surface area and catalytic efficiency. In practical applications, the catalyst demonstrated outstanding catalytic activity by promoting the sulfenylation of C-H bonds in imidazopyridines, utilizing a green solvent for the reaction, identified as PEG-400 (Scheme 4). This not only underscores the catalyst's effectiveness but also highlights its alignment with sustainable practices in chemical synthesis. Notably, substrates decorated with electron-donating groups, such as iodobenzene and phenyl groups in the derivatives of 2-phenylimidazo[1,2-*a*]pyridine, yielded particularly higher reaction rates, emphasizing the importance of these functional groups in enhancing reactivity. Reusability tests confirmed that the Fe_3O_4 @ SiO_2 -ABHA-CuCl catalyst [MRC-4] has high catalytic activity despite



Scheme 2 CuFe_2O_4 NPs (MRC-2) catalyzed synthesis of benzothiazole or benzoxazole-sulfide aryls from S_8 .



Scheme 3 Fe_3O_4 @ABA-aniline-CuI nanomaterial (MRC-3) catalyzed preparation of benzothiazole sulfides from CS_2 .



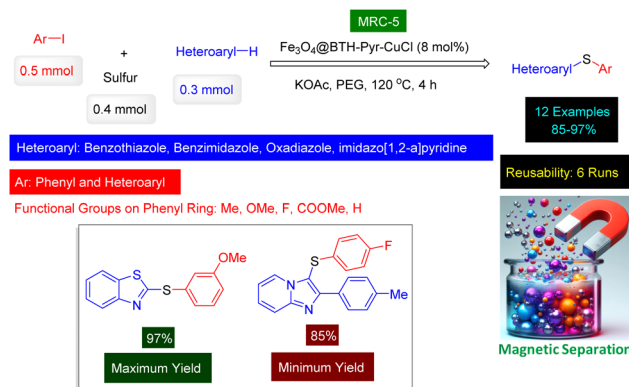
Scheme 4 $\text{Fe}_3\text{O}_4@(\text{SiO}_2\text{-ABHA-CuCl})$ nanomaterial (**MRC-4**) catalyzed synthesis of diaryl sulfides infused with intricate imidazo[1,2-*a*]pyridine scaffolds from S_8 .

being reused 8 times. A comprehensive analysis after repeated use confirmed that the catalyst not only retained its magnetic properties but also its structural integrity, suggesting that it can be effectively employed in continuous processes without compromising its function.

In a compelling and efficient discovery, Chang and Wang introduced a novel catalyst for the formation of carbon–sulfur (C–S) bonds. This catalyst denoted as $\text{Fe}_3\text{O}_4@(\text{BTH-Pyr-CuCl})$ nanomaterial (**MRC-5**) (Fig. 4), is composed of copper(i) chloride immobilized on magnetic nanoparticles that have been modified with a unique benzothiazole-pyrimidine ligand.¹³⁹ The innovative design allows for the catalyst to be magnetically recovered following its use in reactions involving a specific set of heterocyclic compounds in combination with aryl iodides, as well as sulfur and selenium reagents. The structural characteristics of the $\text{Fe}_3\text{O}_4@(\text{BTH-Pyr-CuCl})$ nanocatalyst were thoroughly analyzed using a range of advanced techniques, including FT-IR, SEM, TEM, EDX, elemental mapping, TGA, XRD, VSM and ICP-OES techniques. Significantly, the recycling tests demonstrated that the $\text{Fe}_3\text{O}_4@(\text{BTH-Pyr-CuCl})$ nanocatalyst maintained its effectiveness through six consecutive uses, exhibiting only a minimal decrease in catalytic activity. This method stands out in comparison to other reported methodologies for C–S coupling of heterocycles due to several advantages: it employs an environmentally friendly solvent, achieves high product yields, utilizes a catalyst that can be easily separated and reused, and facilitates reactions within a significantly shorter time frame. Reusability tests confirmed that the $\text{Fe}_3\text{O}_4@(\text{BTH-Pyr-CuCl})$ catalyst [**MRC-5**] has high catalytic activity despite being reused 6 times (Scheme 5).

2.5. MCRs synthesis of thioesters

Thioesters play a crucial role in various biological and medical applications due to their involvement in fundamental biochemical processes.¹⁴⁰ In the realm of medicinal chemistry, thioesters are known for their therapeutic potential, exhibiting



Scheme 5 $\text{Fe}_3\text{O}_4@(\text{BTH-Pyr-CuCl})$ nanomaterial (**MRC-5**) catalyzed synthesis of heteroaryl-aryl sulfides from sulfur.

a range of biological activities including antibacterial, antioxidant, anticancer, anti-inflammatory, anti-Alzheimer, antituberculosis, and antimalarial properties (Fig. 6).^{141,142} These compounds serve as intermediates in metabolic pathways, particularly in the synthesis and degradation of fatty acids and steroids, such as in the formation of acetyl-CoA, a key molecule in energy production and biosynthesis.¹⁴³ The carbonyl reaction is integral to the synthesis of thioesters, providing a reactive site for nucleophilic attack.¹⁴⁴ This reactivity is harnessed in the preparation of thioesters, where the carbonyl carbon of a carboxylic acid reacts with a thiol group.¹⁴⁵ The increased reactivity of the carbonyl group in thioesters compared to oxygen nucleophiles allows for the formation of amides, which are pivotal in peptide synthesis through native chemical ligation methods.¹⁴⁶ This enhanced reactivity also facilitates the synthesis of thioesters from aldehydes and thiols, a process that

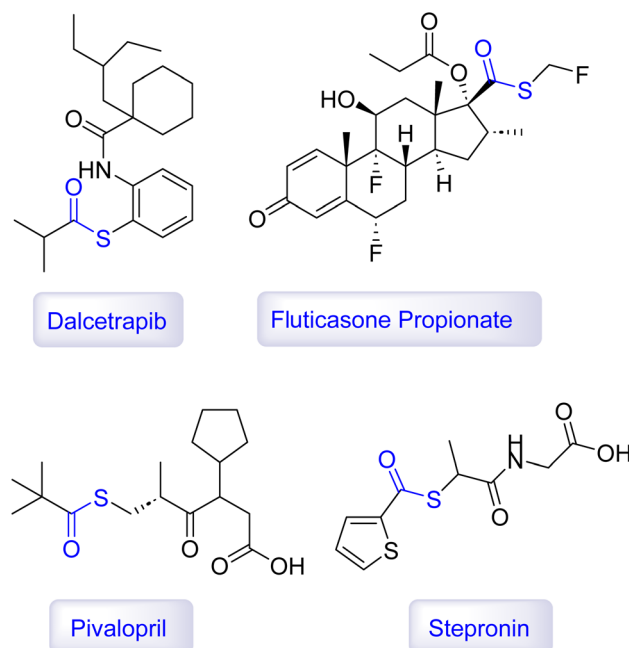


Fig. 6 Several bioactive examples of thioester molecules.



is critical for the production of biologically active molecules and polymers with applications ranging from bioconjugates to biodegradable materials. The versatility and reactivity of thioesters underscore their importance in both biological systems and synthetic chemistry, making them valuable targets for drug development and material science.¹⁴⁷

To synthesize thioester derivatives, Wei's research group developed a nickel magnetic catalyst $[\text{Fe}_3\text{O}_4@\text{BA}/\text{Pyrim-carboxamide-NiCl}_2]$ and examined its catalytic performance in three-component reactions involving aryl iodides, aryl or benzyl phenols or thiols, and $\text{Cr}(\text{CO})_6$ as the carbonyl source.¹³⁸ Initially, Fe_3O_4 nanoparticles were surface modified using 4-(aminomethyl)benzoic acid to create the magnetic nickel catalyst, followed by aminolysis through the reaction of the amine with pyrimidine-2-carbonyl chloride, which resulted in the formation of the magnetic $\text{Fe}_3\text{O}_4@\text{BA}/\text{Pyrim-carboxamide-NiCl}_2$ catalyst [MRC-6] (Fig. 7). The Vibrating Sample Magnetometry (VSM) analysis clearly indicated that the $\text{Fe}_3\text{O}_4@\text{BA}/\text{Pyrim-carboxamide-NiCl}_2$ catalyst possesses significant magnetic potential. The structure and functional groups of the $\text{Fe}_3\text{O}_4@\text{BA}/\text{Pyrim-carboxamide-NiCl}_2$ catalyst [MRC-6] were confirmed through X-ray Diffraction (XRD) and Fourier Transform Infrared (FT-IR) analyses. The authors performed a range of experiments to optimize the conditions for the synthesis of esters and thioesters, finding that both the type of base and the amount of catalyst were critical to the optimization process. By evaluating different solvents, the authors discovered that the CHCl_3 -urea solvent was more effective than others for synthesizing ester and thioester derivatives. Reactions involving aryl iodides, aryl or benzyl thiols, and $\text{Cr}(\text{CO})_6$ as the carbonyl source were catalyzed by the $\text{Fe}_3\text{O}_4@\text{BA}/\text{Pyrim-carboxamide-NiCl}_2$ nanocomposite in the presence of KOAc, resulting in various thioester derivatives being synthesized with high yields in CHCl_3 -urea under mild conditions and air. Based on recycling tests, the $\text{Fe}_3\text{O}_4@\text{BA}/\text{Pyrim-carboxamide-NiCl}_2$ catalyst [MRC-6] was successfully separated *via* magnetic decantation and could be reused up to eight times without a decrease in its catalytic efficiency (Scheme 6).¹⁴⁸

To synthesize thioester derivatives, Khalili's research team introduced an intriguing and practical method that involves the

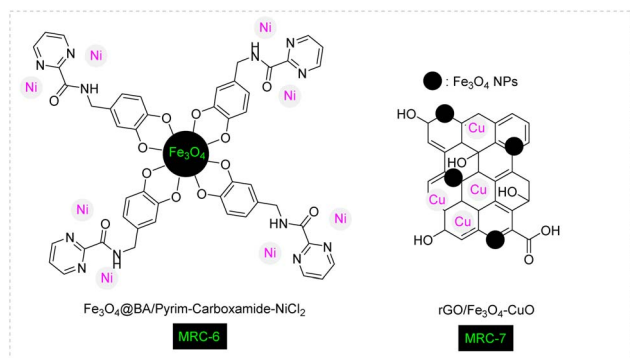
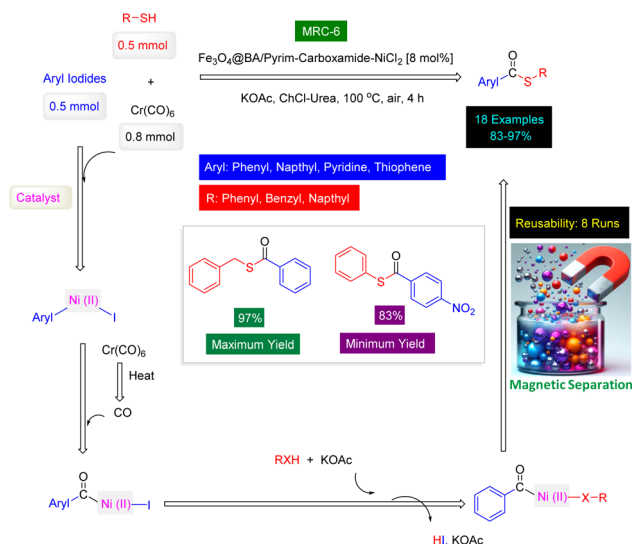
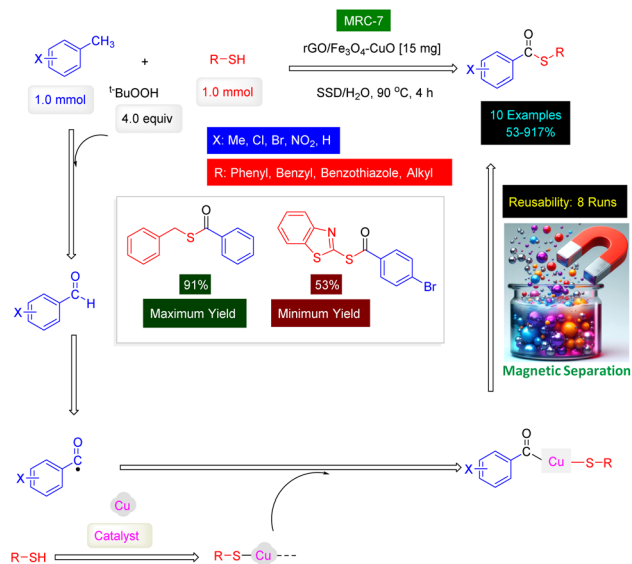


Fig. 7 Structure of magnetic catalysts in MRCs synthesis of thioesters.



Scheme 6 $\text{Fe}_3\text{O}_4@\text{BA}/\text{Pyrim-carboxamide-NiCl}_2$ (MRC-6) catalyzed preparation of thioesters from thiols.

reaction of methylene derivatives with aryl or alkyl thiols, utilizing *t*-BuOOH and employing $\text{rGO}/\text{Fe}_3\text{O}_4\text{-CuO}$ nanocomposite as an effective and recyclable catalyst.¹⁴³ The catalyst $\text{rGO}/\text{Fe}_3\text{O}_4\text{-CuO}$ [MRC-7] (Fig. 6) was synthesized through a multi-step procedure that involved coating graphene with Fe_3O_4 nanoparticles and then attaching copper oxide onto its surface. The structure and morphology of the $\text{rGO}/\text{Fe}_3\text{O}_4\text{-CuO}$ catalyst were thoroughly confirmed *via* XRD and TEM analyses. To optimize the reaction, the influence of the catalyst quantity and solvent was investigated. As illustrated in Scheme 7, ten thioester derivatives were produced with relatively favorable yields in an aqueous medium under mild reaction conditions. The reaction between toluene derivatives and *t*-BuOOH in the



Scheme 7 $\text{rGO}/\text{Fe}_3\text{O}_4\text{-CuO}$ (MRC-7) catalyzed preparation of thioesters from thiols.

presence of the rGO/Fe₃O₄-CuO catalyst generated a carbonyl radical. During the reaction with aryl and alkyl thiols in the presence of the rGO/Fe₃O₄-CuO catalyst, a sulfide-copper intermediate was formed; this intermediate then entered the reaction cycle, leading to the synthesis of the desired thioester products with good yields. Recycling tests indicated that the rGO/Fe₃O₄-CuO catalyst [MRC-7] could be separated *via* magnetic decantation and reused six times without a decline in its catalytic performance.

2.6. MCRs synthesis of thiazoles

Thiazole ring compounds represent a vital class of heterocyclic compounds that are increasingly recognized in medicinal chemistry for their remarkable range of biological activities, making them important candidates for drug development and therapeutic applications.¹⁴⁹ The thiazole ring, a five-membered structure containing both sulfur and nitrogen atoms, is a key feature in many pharmacologically active molecules.¹⁵⁰ These compounds exhibit a broad spectrum of therapeutic properties, including antimicrobial, antifungal, antiprotozoal, and anti-tumor activities.¹⁵¹ For instance, thiazole derivatives have been found to be effective in treating bacterial infections, with some acting as potent inhibitors of bacterial growth.¹⁵² Additionally, thiazole rings are present in several antiretroviral drugs, which are crucial in the management of HIV/AIDS.¹⁵³ In the realm of cancer treatment, certain thiazole compounds have demonstrated antineoplastic properties, making them valuable in the development of new anticancer drugs.¹⁵⁴ Moreover, the thiazole ring is a component of vitamin B1 (thiamine), which is essential for carbohydrate metabolism and normal neural function.¹⁵⁵ The versatility of thiazole compounds in drug design is further highlighted by their inclusion in drugs with anti-inflammatory and neuroprotective effects, showcasing their potential in treating a variety of diseases and conditions.^{156,157} Several bioactive examples of thiazole molecules are shown in Fig. 8.¹⁵⁸

Magnetic catalysts are transforming the landscape of thiazole derivative synthesis, offering a sustainable and efficient approach to synthesis.¹⁵³ These catalysts typically consist of magnetic nanoparticles that can be easily separated from the reaction mixture using an external magnet, thus simplifying the purification process and reducing waste.⁷² For instance, iron

oxide magnetic nanoparticles have been employed as a reusable catalyst for the synthesis of various thiazole derivatives, enhancing the green chemistry aspect of these reactions.⁷² The use of such catalysts aligns with the principles of green chemistry, aiming to minimize environmental impact while maintaining high efficiency and product yield.

The innovative research team of Bodke has pioneered an effective and streamlined methodology for the one-pot three-component synthesis of isatin-thiazole derivatives. By employing a catalytic amount of biogenic Fe₂O₃ nanoparticles [MRC-8], this approach not only enhances efficiency but also underscores the remarkable potential of using biogenic materials in modern chemical synthesis. This development represents a significant advancement in the field, promising to simplify complex reactions and improve overall yields.¹⁵⁷ A variety of spectroscopic techniques, such as SEM, FT-IR, TGA, XRD, TEM, and VSM analyses, were employed to confirm the structure of Fe₂O₃ nanoparticles. A one-pot, three-component reaction combining different isatins, thiosemicarbazide, and various phenacyl bromides was promoted by Fe₂O₃ nanoparticles under reflux conditions in ethanol, leading to the successful synthesis of isatin-thiazole products with high yields (Scheme 8). TEM images of the Fe₂O₃ nanoparticles showed that the particles are spherical and fall within the nanometer size range, with some degree of aggregation noted. Tests for reusability demonstrated that the Fe₂O₃ catalyst [MRC-8] maintained its high catalytic activity even after being used 5 times.

In a research study, Hangirgekar *et al.* described a comprehensive and effective method for a one-pot three-component reaction involving tosylates, aryl aldehydes, and thiosemicarbazide to create a collection of hydrazinyl thiazoles, utilizing Fe₂O₃ nanoparticles [MRC-8] as a nanomagnetic catalyst.¹⁵² This study examined the effects of varying Fe₂O₃ nanoparticle quantities and solvent types to optimize reaction conditions. Characterization techniques including XRD, SEM, VSM, HR-TEM, EDX, and FT-IR were utilized to analyze the structural properties of the Fe₂O₃ nanoparticles. We also performed a one-pot three-component reaction using tosylates, aryl aldehydes, and thiosemicarbazide, with Fe₂O₃ nanoparticles as

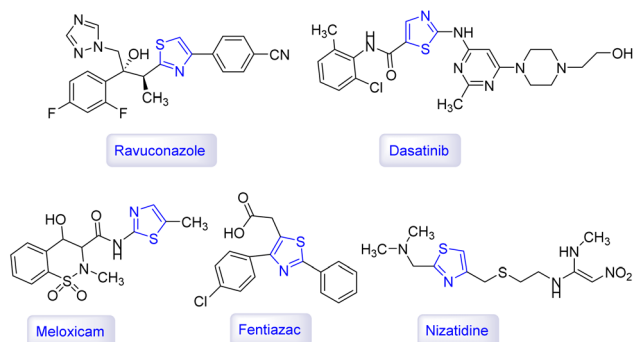
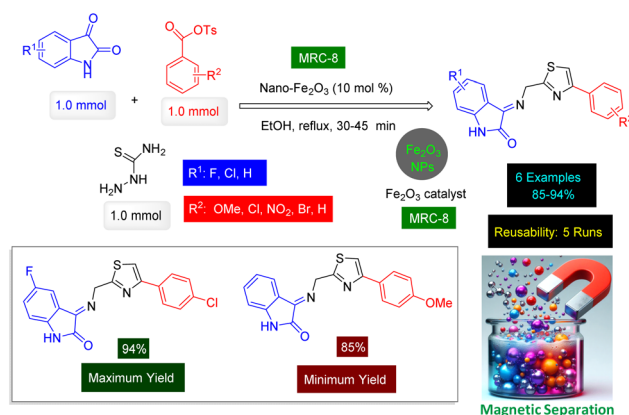
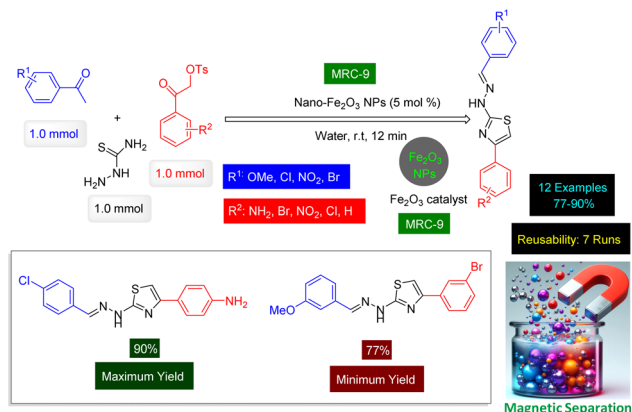


Fig. 8 Several examples of bioactive compounds based on the thiazole structure.



Scheme 8 Fe₂O₃ nanoparticles (MRC-8) catalyzed preparation of isatin-thiazole derivatives from thiosemicarbazide.

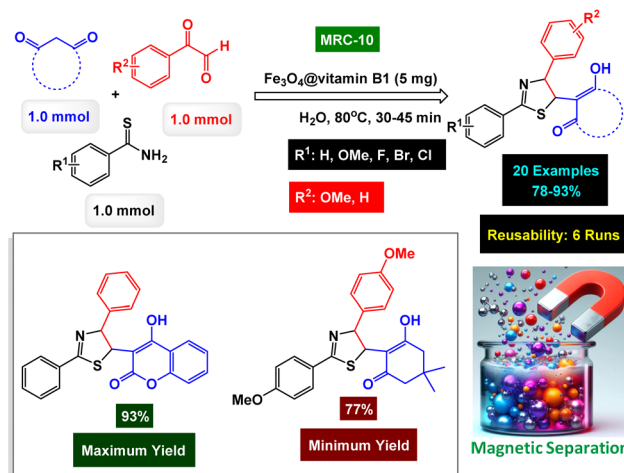




Scheme 9 Fe_2O_3 nanoparticles (MRC-9) catalyzed preparation of hydrazinyl thiazole derivatives from thiosemicarbazide.

the catalyst in water (see Scheme 3). Reusability tests confirmed that the Fe_3O_4 @dopamine-PO-CuBr₂ catalyst [MRC-9] has high catalytic activity despite being reused 7 times (Scheme 9).

A novel and environmentally friendly approach have been developed for the synthesis of biologically active trisubstituted 1,3-thiazole derivatives. This methodology utilizes a one-pot, three-component reaction involving aryl glyoxal monohydrate, cyclic 1,3-dicarbonyl compounds, and thioamides. Central to this innovative process is the use of a unique catalyst: vitamin B1 that is immobilized on the surface of magnetic Fe_3O_4 nanoparticles, referred to as $[\text{Fe}_3\text{O}_4\text{@vitamin B1}]$. This method not only enhances the efficiency of the reaction but also allows for the convenient recovery of the catalyst using a magnetic field, making it a significant advancement in synthetic chemistry as reported by researchers Shaterian and Molaei.¹⁵⁶ The formation of the Fe_3O_4 @vitamin B1 nanocatalyst [MRC-10] (Fig. 9) was confirmed through EDX and FT-IR techniques. As shown in Scheme 10, one-pot three-component reactions involving aryl glyoxal monohydrate, cyclic 1,3-dicarbonyls, and



Scheme 10 Fe_3O_4 @vitamin B1 nanomaterial (MRC-10) catalyzed preparation of trisubstituted 1,3-thiazole derivatives from thioamides.

thioamides were conducted in an aqueous medium under mild conditions, resulting in the successful synthesis of trisubstituted 1,3-thiazole products with moderate to high yields. Reusability tests confirmed that the Fe_3O_4 @vitamin B1 catalyst [MRC-10] has high catalytic activity despite being reused 6 times.

In a research paper authored by Saleh Shafik and Jasim Elaibi, a novel method for synthesizing thiazole derivatives has been introduced, characterized by its efficiency, cost-effectiveness, and simplicity.¹⁵⁹ As shown in Scheme 11, this approach employs a one-pot three-component reaction involving ethyl chloroacetate, thiosemicarbazide, and various acetophenone derivatives, all facilitated by the use of Fe_3O_4 @ CeO_2 nanocatalyst [MRC-11]. The research details the preparation of Fe_3O_4 @ CeO_2 magnetic nanoparticles through a hydrothermal method, resulting in nanoparticles with an average size of approximately 25 nm. These nanoparticles were thoroughly characterized using FT-IR and XRD techniques. The FT-IR analysis confirmed the presence of specific functional groups associated with each oxide, while the XRD results

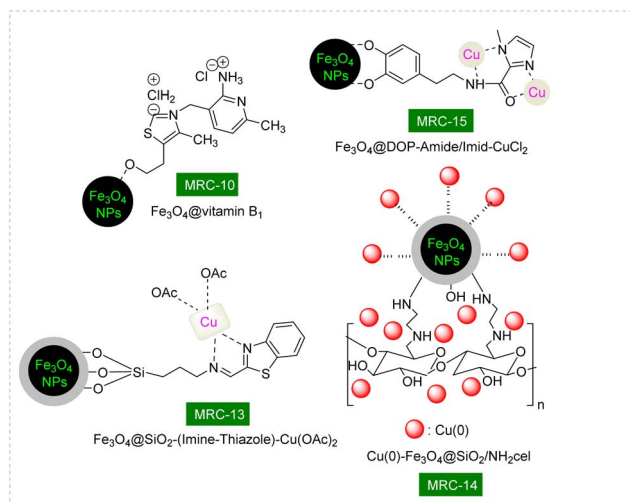
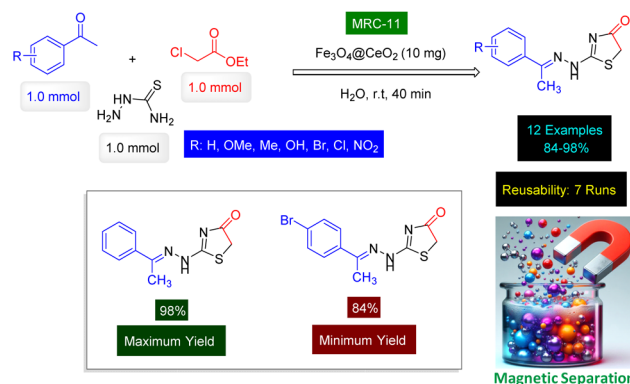


Fig. 9 Structure of magnetic catalysts in MCRs synthesis of thiazoles and benzothiazoles.



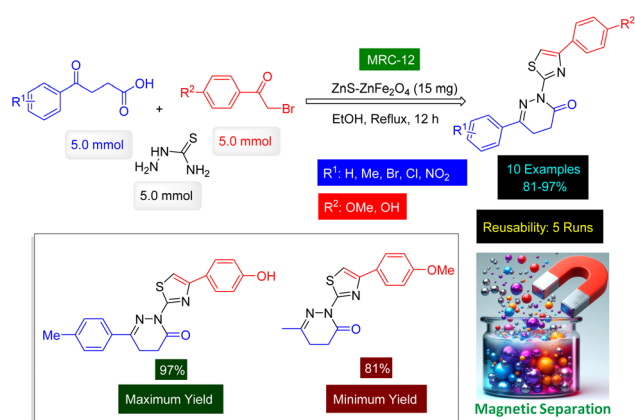
Scheme 11 Fe_3O_4 @ CeO_2 nanomaterial (MRC-11) catalyzed preparation of thiazole derivatives from thiosemicarbazide.

validated the successful synthesis of the nanocatalyst by identifying distinct peaks corresponding to the oxides involved. One of the standout features of this synthetic method is its high efficiency, yielding thiazole products in a remarkably short time frame of just 40 minutes. Moreover, the process is notable for its ease of execution, mild reaction conditions, and outstanding performance, making it an appealing option for researchers in the field. The integration of mechanization further enhances the practicality of this approach, positioning it as a valuable contribution to the synthesis of thiazole derivatives. Reusability tests confirmed that the $\text{Fe}_3\text{O}_4@\text{CeO}_2$ catalyst [MRC-11] has high catalytic activity despite being reused 7 times.

In an important study conducted by Mokhtari and Naseri, a sophisticated and highly effective method was unveiled for synthesizing 2-(thiazol-2-yl)-4,5-dihydropyridazine-3-(2H)-one derivatives through a novel multicomponent cyclization process.¹⁶⁰ This innovative synthetic pathway combines three key reactants: ketoacid, thiosemicarbazide, and phenacyl bromide, which together form the foundation of the desired compounds. The reaction is significantly enhanced by the use of a specialized catalyst, $\text{ZnS-ZnFe}_2\text{O}_4$ [MRC-12], known for its heterogeneous and recoverable properties, all while utilizing ethanol as a solvent under controlled reflux conditions (Scheme 12). This research is characterized by several noteworthy features that demonstrate its potential: efficient direct synthesis, intriguing structural complexity, rapid reaction times, mild and green reaction conditions, impressive efficiency. Overall, Mokhtari and Naseri's findings present a comprehensive and promising strategy for producing complex organic molecules, setting the stage for future explorations in therapeutic applications and contributing significantly to advancements in the field of organic chemistry. Reusability tests confirmed that the $\text{ZnS-ZnFe}_2\text{O}_4$ catalyst [MRC-12] has high catalytic activity despite being reused 5 times.

2.7. MCRs synthesis of benzothiazoles

Benzothiazole compounds represent an important category of heterocyclic compounds that have attracted interest due to their broad biological and pharmaceutical characteristics.¹⁶¹ These



Scheme 12 $\text{ZnS-ZnFe}_2\text{O}_4$ nanomaterial (MRC-12) catalyzed preparation of pyridazinone-thiazole derivatives from thiosemicarbazide.

compounds exhibit a wide range of biological activities, including antimicrobial, anticancer, antifungal, anthelmintic, anti-diabetic, and amyloid imaging agents, making them crucial in drug development and therapeutic applications.^{162,163} The synthesis of benzothiazole derivatives is of great scientific interest as they are found in bioorganic and medicinal chemistry with applications in drug discovery.¹⁶⁴ They are also used industrially as antioxidants and vulcanization accelerators.¹⁶⁵ The diverse biological activities of benzothiazole compounds, such as their use in radioactive amyloid imaging agents and as anticancer agents, underscore their importance in the synthesis of new pharmacological products with potential benefits in treating various diseases.¹⁶⁶ The structure of several bioactive benzothiazole molecules is shown in Fig. 10.^{167,168}

Magnetic catalysts offer several advantages in the synthesis of benzothiazole compounds. They are easy to synthesize and provide high catalytic activity, which can significantly reduce the reaction time.¹⁶⁹ Additionally, their magnetic properties allow for easy separation from the reaction mixture, enhancing the convenience of the process.¹⁷⁰ The reusability of magnetic catalysts also contributes to a more sustainable and cost-effective synthesis, as they can be recovered and reused multiple times without significant loss of activity.¹⁷⁰ Furthermore, the large surface area of these catalysts facilitates better interaction with reactants, leading to higher yields and more efficient reactions.

Patra and his team meticulously described the innovative construction of copper acetate ($\text{Cu}(\text{OAc})_2$) that was effectively immobilized onto the surface of specially modified magnetic nanoparticles featuring an imine-thiazole ligand. The resulting composite, identified as $[\text{Fe}_3\text{O}_4@\text{SiO}_2(\text{imine-thiazole})-\text{Cu}(\text{OAc})_2]$, functions as a magnetically recoverable catalyst for copper. This catalyst facilitates the synthesis of 2-substituted benzothiazoles *via* a streamlined one-pot three-component reaction. This reaction involves 2-iodoanilines, benzyl chlorides, and a sulfur source, all carried out in the presence of K_2CO_3 as the base, while utilizing dimethyl sulfoxide (DMSO) under controlled thermal conditions.¹⁷¹ TGA and VSM investigations revealed that the $\text{Fe}_3\text{O}_4@\text{SiO}_2(\text{imine-thiazole})-\text{Cu}(\text{OAc})_2$ nanomaterial [MRC-12] (Fig. 8) exhibits remarkable stability and magnetic properties. The attempted synthesis involving a template reaction with 2-iodoaniline, benzyl chloride, and sulfur (S8) was unsuccessful when either the catalyst

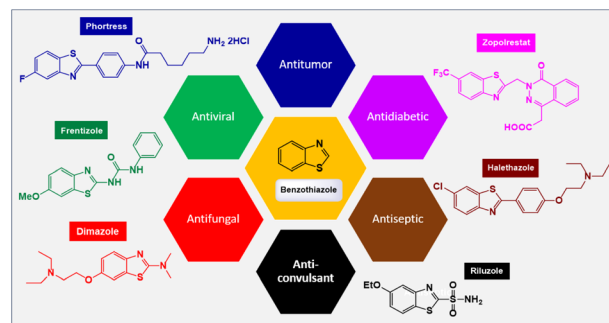
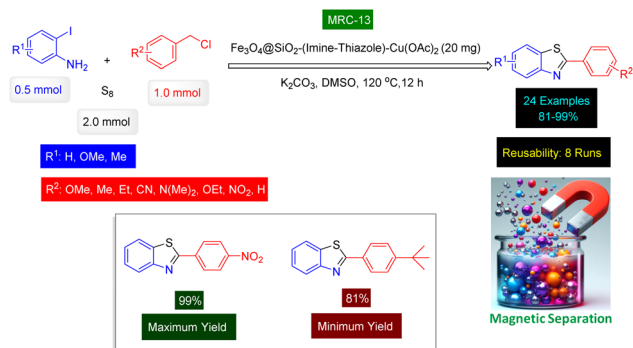
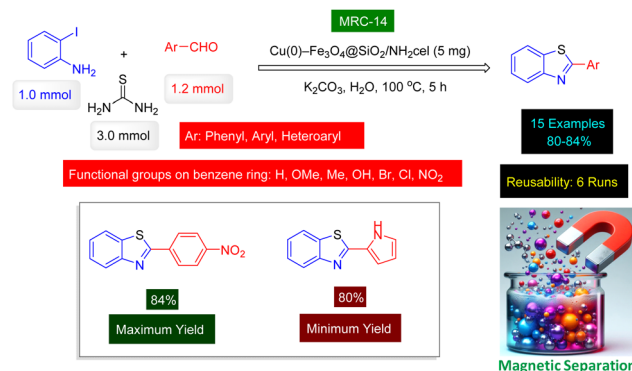


Fig. 10 Several bioactive examples of benzothiazole molecules.





Scheme 13 $\text{Fe}_3\text{O}_4\text{@SiO}_2\text{-(imine-thiazole)-Cu(OAc)}_2$ nanomaterial (MRC-13) catalyzed preparation of 2-substituted benzothiazole derivatives from S_8 .



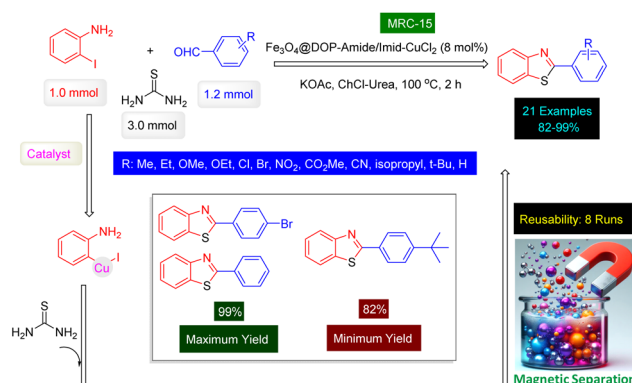
Scheme 14 $\text{Cu(0)-Fe}_3\text{O}_4\text{@SiO}_2\text{/NH}_2\text{cel}$ nanomaterial (MRC-14) catalyzed preparation of 2-aryl benzothiazole derivatives from thiourea.

or base was omitted, indicating the essential role of these components in the reaction. Building upon the findings detailed in this methodology, a diverse array of 2-substituted benzothiazole products was successfully generated, with a total of 24 distinct examples demonstrating yields ranging from high to excellent, all achieved under mild reaction conditions (as illustrated in Scheme 13). Reusability tests confirmed that the $\text{Fe}_3\text{O}_4\text{@SiO}_2\text{-(imine-thiazole)-Cu(OAc)}_2$ catalyst [MRC-13] has high catalytic activity despite being reused 8 times.

Paul *et al.* successfully synthesized a promising category of 2-substituted benzothiazoles with high yields through an innovative one-pot, three-component reaction. This reaction involved the combination of 2-iodoaniline, various aldehydes, and thiourea, all carried out in refluxing water. The remarkable aspect of their approach was the use of copper nanoparticles, which were supported on an ethylene diamine functionalized inorganic/organic composite known as $[\text{Cu(0)-Fe}_3\text{O}_4\text{@SiO}_2\text{/NH}_2\text{cel}]$. This composite served as a nanomagnetic recoverable catalyst, providing both efficiency and ease of recovery at the completion of the reaction. The process is illustrated in Scheme 14, showcasing the effectiveness of this catalytic system in facilitating the desired chemical transformations.¹⁶¹ The study focused on investigating the type of catalyst to determine the optimal conditions for the reaction. Additionally, the quantity of the $\text{Cu(0)-Fe}_3\text{O}_4\text{@SiO}_2\text{/NH}_2\text{cel}$ nanocatalyst [MRC-14] (Fig. 8) was carefully assessed to achieve the best results. TEM and SEM images revealed that the average size of the Cu(0) nanoparticles was approximately 9 nm. This measurement closely aligned with the values obtained through XRD analysis, indicating a strong consistency between the different characterization methods used. Reusability tests confirmed that the $\text{Cu(0)-Fe}_3\text{O}_4\text{@SiO}_2\text{/NH}_2\text{cel}$ catalyst [MRC-14] has high catalytic activity despite being reused 6 times.

In a remarkable synthetic endeavor, Zhang and his research team discovered an innovative catalytic system utilizing a $\text{Fe}_3\text{O}_4\text{@DOP-amide/Imid-CuCl}_2$ nanocomposite.¹⁷² This system operates in the presence of potassium acetate (KOAc) within a CHCl_3 -urea solvent environment, making it an eco-friendly and highly effective method for producing 2-aryl benzothiazoles. The process involves a one-pot, three-component reaction,

integrating 2-iodoaniline, aromatic aldehydes, and thiourea as the sulfur source. This catalytic approach has proven to be versatile, successfully yielding a wide array of 2-aryl benzothiazoles with high to excellent efficiency. When evaluated against other reported catalysts and magnetic nanocatalysts, the $\text{Fe}_3\text{O}_4\text{@DOP-amide/Imid-CuCl}_2$ nanocatalyst [MRC-15] (Fig. 8) stands out due to several key characteristics. Notably, it features the clever design of a magnetic ligand achieved through an ammonolysis reaction. Additionally, it demonstrates impressive magnetic properties, which facilitate easy separation and recovery. The nanocatalyst showcases remarkable catalytic activity in the synthesis of various benzothiazole derivatives, while also emphasizing its reusability and high stability throughout multiple catalytic cycles. This combination of attributes underscores the potential of this nanocomposite in advancing sustainable and efficient chemical synthesis. Reusability tests confirmed that the $\text{Fe}_3\text{O}_4\text{@DOP-amide/Imid-CuCl}_2$ catalyst [MRC-15] has high catalytic activity despite being reused 5 times. The techniques of SEM, VSM, BET, and ICP-OES



Scheme 15 $\text{Fe}_3\text{O}_4\text{@DOP-amide/Imid-CuCl}_2$ nanomaterial (MRC-15) catalyzed preparation of 2-aryl benzothiazole derivatives from thiourea.



demonstrated that the $\text{Fe}_3\text{O}_4@\text{DOP-amide}/\text{Imid-CuCl}_2$ catalyst, after being reused 8 times, exhibited significant stability, as its magnetic characteristics, structure, and surface properties were comparable to those of the fresh catalyst (Scheme 15).

2.8. MCRs synthesis of sulfonamides

Sulfonamides are a class of synthetic antimicrobial agents that have played a significant role in treating bacterial infections due to their ability to inhibit the synthesis of folic acid in bacteria, which is essential for their growth and replication.^{173,174} They are structural analogs of *para*-aminobenzoic acid (PABA) and competitively inhibit the enzyme dihydropteroate synthase in the folate synthesis pathway.¹⁷⁵ They are known for their ability to inhibit enzymes like carbonic anhydrase and dihydropteroate synthetase, which makes them effective in treating various disease states including diuresis, hypoglycemia, thyroiditis, inflammation, and glaucoma.^{176–178} This disruption of folate production is crucial as it leads to bacterial cell death. In medicine, sulfonamides are used to treat a variety of bacterial infections, including urinary tract infections, respiratory infections, and certain types of diarrhea.¹⁷⁹ The structure of several bioactive examples of sulfonamide molecules are shown in Fig. 11.^{180–182} Chemically, the sulfonamide group ($-\text{SO}_2\text{NH}_2$) imparts certain properties to these compounds, such as solubility and the ability to interact with biological targets, which can be modified by substituting different groups onto the molecule to enhance its therapeutic index.^{183,184} The preparation of sulfonamides has been revolutionized by the use of magnetic catalysts, which offer a sustainable and efficient approach to synthesizing these compounds.¹⁸⁵ These catalysts facilitate one-pot, multi-component reactions under eco-friendly conditions, and their magnetic properties allow for easy separation and reuse, thus reducing waste and improving reaction efficiency.¹⁸⁶ The use of such catalysts is a testament to the advancements in green chemistry, providing a more environmentally friendly and cost-effective method for producing sulfonamides, which continue to be vital in the pharmaceutical industry.¹⁸⁶

In 2023, a paper was published in which Chang's research group used copper(II) bromide immobilized on Fe_3O_4 nanoparticles modified by dopamine and styrene epoxide as a magnetic reusable catalyst [$\text{Fe}_3\text{O}_4@\text{dopamine-PO-CuBr}_2$] to

prepare *N*-aryl sulfonamide derivatives.¹⁸⁷ The authors first covered the surface of Fe_3O_4 nanoparticles with dopamine, then by the aminolysis of styrene epoxide, an attractive magnetic ligand was made to stabilize the CuBr_2 (Fig. 12) [MRC-16]. VSM analysis showed that the $\text{Fe}_3\text{O}_4@\text{dopamine-PO-CuBr}_2$ catalyst has high magnetic properties, and TGA analysis confirmed the thermal stability of the $\text{Fe}_3\text{O}_4@\text{dopamine-PO-CuBr}_2$ catalyst. After conducting optimization experiments on the selected model reaction, the three-component reactions of phenyl boronic acids with aniline derivatives in the presence of DABSO as the SO_2 source was successfully catalyzed by $\text{Fe}_3\text{O}_4@\text{dopamine-PO-CuBr}_2$ nanocomposite in the presence of KOAc in DMSO under thermal conditions for 8 h. As seen in Scheme 16, a category of the *N*-aryl sulfonamide products (21 examples) was synthesized with high efficiencies (76–97%) under the standardized conditions. Reusability tests confirmed that the $\text{Fe}_3\text{O}_4@\text{dopamine-PO-CuBr}_2$ catalyst [MRC-16] has high catalytic activity despite being reused 8 times.

The innovative research team headed by Shams has successfully devised a highly effective and environmentally sustainable technique for synthesizing a range of sulfonamide derivatives, utilizing MNPs-benzo[d]imidazole-Cu [MRC 17] (Fig. 11) as a magnetically separable catalyst.¹⁸⁸ The MNPs-benzo[d]imidazole-Cu nanocatalyst was synthesized through a four-step process, which included the silica protection of Fe_3O_4 nanoparticles, functionalization with (3-aminopropyl) trimethoxysilane (APTMS), modification with (1-*H*-benzo[d]imidazol-2-yl)methanamine, and coordination with copper(II) chloride. Characterization by XRD analysis confirmed the stability and alignment of the catalyst's structure with previously reported magnetic nanoparticles. XPS and FT-IR spectroscopy further validated the bond formation between the ligand and the copper complex. The resulting nanocomposite was utilized for the synthesis of sulfonamide derivatives through a one-pot, three-component reaction involving aryl-diazonium tetrafluoroborate, DABCO(SO_2)₂, and *N*-

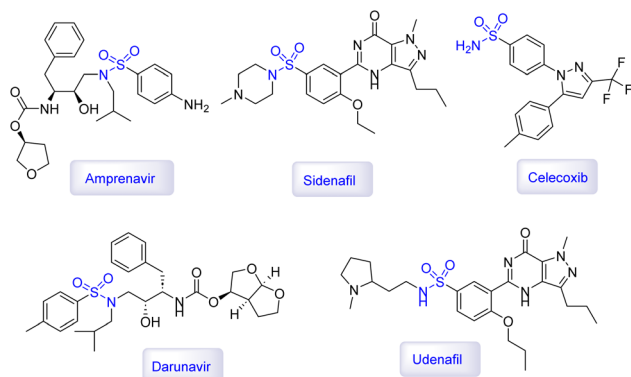


Fig. 11 Several bioactive examples of sulfonamide molecules.

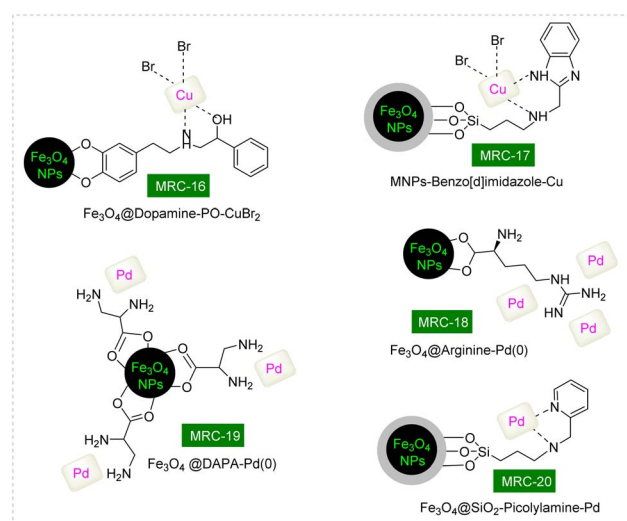
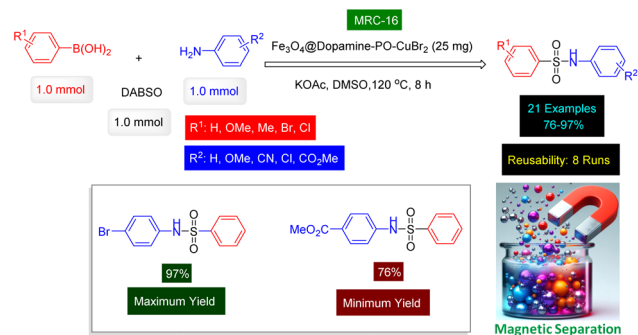


Fig. 12 Structure of magnetic catalysts in MCRs synthesis of sulfonamides.

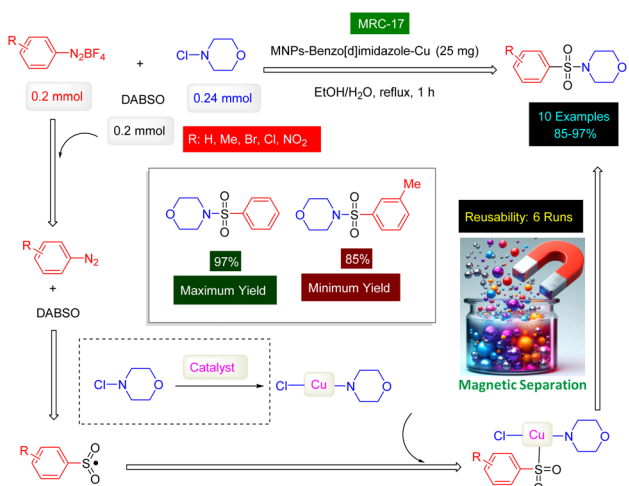




Scheme 16 Fe_3O_4 @dopamine-PO-CuBr₂ (MRC-16) catalyzed preparation of *N*-aryl sulfonamides from DABSO.

chloroamines in an aqueous medium under mild conditions, as illustrated in Scheme 17. The synthesis of sulfonamide products presents a remarkable opportunity, significantly enhanced by the choice of catalyst and solvent. In carefully controlled conditions, our team successfully produced a diverse range of ten sulfonamides, achieving astonishing yields between 88% and 97%. Our innovative MNPs-benzo[d]imidazole-Cu catalyst demonstrated impressive reusability; when tested with 4-(phenylsulfonyl)morpholine as the model substrate, it retained its catalytic efficiency across six consecutive cycles, showcasing its exceptional durability and effectiveness. Moreover, we have proposed a comprehensive mechanistic pathway that elucidates the formation of sulfonamides through the strategic reactions of aryldiazonium tetrafluoroborate, DABCO(SO₂)₂, and *N*-chloroamines-facilitated seamlessly by the cutting-edge MNPs-benzo[d]imidazole-Cu nanocomposite (MRC-17). This is clearly outlined in Scheme 17, emphasizing the sophistication and potential of our approach.

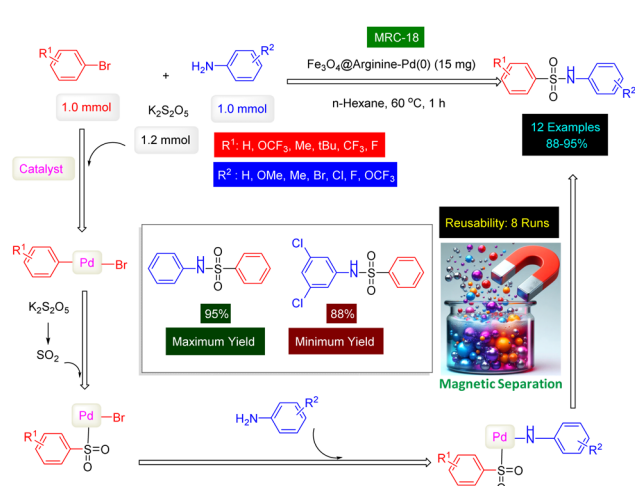
Naderi's research team has made a significant advancement by presenting an innovative and flexible method for synthesizing sulfonamides. This approach utilizes a palladium complex anchored to the surface of magnetic nanoparticles,



Scheme 17 MNPs-benzo[d]imidazole-Cu (MRC-17) catalyzed preparation of sulfonamides from DABSO.

which are enhanced with arginine, providing an exceptionally efficient and magnetically recoverable catalyst.¹⁰⁰ The magnetic Fe_3O_4 nanoparticles were expertly coated with arginine, resulting in the innovative Fe_3O_4 @arginine nano ligand. This was followed by the strategic attachment of the Pd(0) complex on its surface, culminating in the advanced Fe_3O_4 @arginine-Pd(0) nanocatalyst [MRC-18] (Fig. 11), designed to enhance catalytic efficiency. The bonding of C=N groups with the palladium complex was verified through FT-IR spectroscopy. In this study, thermogravimetric analysis (TGA) was employed to confirm the successful attachment of functional groups on the surface of the magnetic nanoparticles, demonstrating the effectiveness of the immobilization process. The research further explored the potential of the Fe_3O_4 @arginine-Pd(0) nanocomposite, referred to as MRC-18, as a catalyst for one-pot three-component reactions involving bromo benzenes, anilines, and potassium persulfate (K₂S₂O₈). Conducted in *n*-hexane under mild conditions, these reactions resulted in the efficient synthesis of various sulfonamide derivatives, showcasing an impressive yield range of 88% to 95% across ten different examples within a remarkably short reaction time of less than one hour (as depicted in Scheme 18). To assess the practical applicability of the Fe_3O_4 @arginine-Pd(0) catalyst (MRC-18), the researchers conducted tests to evaluate its reusability. The synthesis of *N*-phenylbenzene sulfonamide was selected as a model reaction for this purpose. Remarkably, the recovered catalyst retained its catalytic efficacy across eight consecutive cycles, illustrating its robustness and efficiency. Furthermore, the research group put forth a plausible mechanistic pathway to elucidate how sulfonamides are formed through the one-pot three-component reactions catalyzed by MRC-18. This proposed mechanism is carefully illustrated in Scheme 18, providing a comprehensive understanding of the reaction dynamics involved.

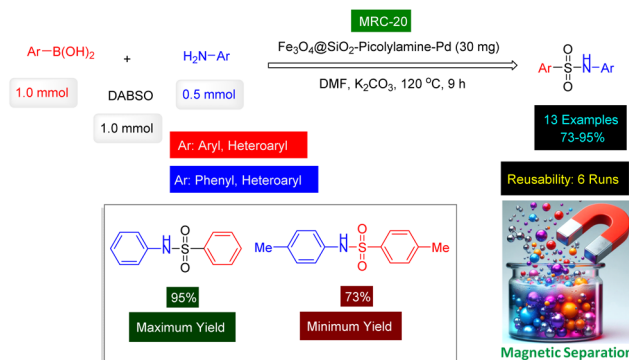
Jin has developed an innovative magnetic palladium (Pd) catalyst, designated as Fe_3O_4 @DAPA-Pd(0), which facilitates the synthesis of a wide variety of *N*-aryl and alkyl sulfonamides. This catalyst is created by immobilizing a Pd(0) complex onto iron



Scheme 18 Fe_3O_4 @arginine-Pd(0) (MRC-18) catalyzed preparation of *N*-aryl sulfonamides from K₂S₂O₈.

oxide (Fe_3O_4) nanoparticles, which have been coated with 2,3-diaminopropionic acid, enhancing the stability and effectiveness of the catalyst in various reactions. In his research, Jin thoroughly examined the catalytic performance of this system in a one-pot, three-component reaction process. This process involves a combination of aryl or heteroaryl iodides, amines, and sulfur dioxide (SO_2), all while employing potassium bicarbonate (KHCO_3) as a base within an ionic liquid solvent. This approach not only streamlines the synthetic pathway but also showcases the practical applications of the catalyst in producing valuable sulfonamide compounds efficiently.¹⁸⁹ The TEM and XRD analyses provided strong evidence confirming that the particles are situated within the nanometer range, measuring approximately 10 to 25 nanometers. Additionally, the VSM analysis revealed that the Fe_3O_4 @DAPA-Pd(0) catalyst, referred to as **MRC-19** (Fig. 11), demonstrates exceptional magnetic properties both before it is recovered and after. As depicted in Scheme 19, a diverse array of aryl and heteroaryl halides was meticulously reacted with a selection of aryl, heteroaryl, and alkyl amines. This careful approach resulted in the successful synthesis of sulfonamide products, achieving high yields in the process. The recyclability of the Fe_3O_4 @DAPA-Pd(0) catalyst, designated as [**MRC-19**], was thoroughly investigated in the context of synthesizing sulfonamides. The results demonstrated that the recovered catalyst maintained its catalytic efficiency effectively, allowing it to be reused for up to eight consecutive cycles without any noticeable decline in performance. To illustrate the process, a proposed synthetic pathway for the formation of sulfonamides is presented. This pathway involves the reaction of aryl or heteroaryl iodides with amines and sulfur dioxide (SO_2), all facilitated by the Fe_3O_4 @DAPA-Pd(0) nanomaterial (**MRC-19**) in the presence of potassium bicarbonate (KHCO_3). This mechanism is visually represented in Scheme 19, highlighting the intricate interactions that enable the successful synthesis of sulfonamides.

In a different methodological framework, Saleh-Shafik and Saif meticulously documented the development of a palladium

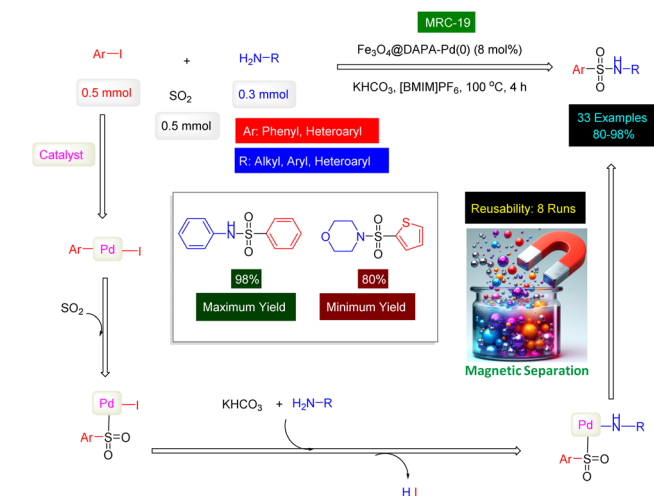


Scheme 20 Fe_3O_4 @ SiO_2 -picolylamine-Pd (**MRC-20**) catalyzed preparation of sulfonamides from DABSO.

(Pd) complex that is immobilized on nanoparticles consisting of a core of iron oxide (Fe_3O_4) coated with silica (SiO_2). These nanoparticles were further modified with picolylamine, resulting in the innovative formation of a magnetic catalyst, designated as [Fe_3O_4 @ SiO_2 -picolylamine-Pd] (referred to as **MRC-20**) (Fig. 11). This newly synthesized magnetic catalyst demonstrates significant potential for the efficient production of a specific class of sulfonamides, showcasing its application in catalytic processes.¹⁹⁰ The Fe_3O_4 @ SiO_2 -picolylamine-Pd catalyst (**MRC-20**) was developed using a straightforward method with readily available materials. The construction of the Fe_3O_4 @ SiO_2 -picolylamine-Pd catalyst [**MRC-20**] (Fig. 11) was verified through FT-IR, TGA, EDX, BET, and ICP-OES analyses. The Fe_3O_4 @ SiO_2 -picolylamine-Pd catalyst (**MRC-20**) effectively catalyzed one-pot three-component reactions involving anilines, phenylboronic acids, and DABSO in the presence of K_2CO_3 in DMF under thermal conditions, resulting in high yields of the corresponding sulfonamide products (Scheme 20). The reutilization of the Fe_3O_4 @ SiO_2 -picolylamine-Pd catalyst (**MRC-20**) was investigated using *N*-phenylbenzene sulfonamide as the model substrate, and the recovered catalyst maintained its catalytic efficiency over six consecutive cycles.

2.9. MCRs synthesis of diaryl sulfones

Sulfones are a class of organic compounds that have found extensive applications across various fields due to their unique chemical properties.¹⁹¹ In the medical and pharmaceutical industries, sulfones are utilized for their antimicrobial properties, particularly in the treatment of leprosy and tuberculosis.¹⁹² The introduction of sulfonic acid groups into biomaterials through sulfonation enhances their properties, making them suitable for use in drug delivery systems, tissue engineering, and regenerative medicine.¹⁹³ This chemical modification can improve biomaterial interactions with cells, promoting cell adhesion, proliferation, and differentiation. In the biological realm, sulfones serve as intermediates in the synthesis of complex molecules, including those used in medicinal chemistry.¹⁹⁴ Sulfones are a crucial class of compounds in the medical field, with dapson being a prominent example. Dapsone has been used as an antibiotic to treat various bacterial infections,



Scheme 19 Fe_3O_4 @DAPA-Pd(0) (**MRC-19**) catalyzed preparation of sulfonamides from SO_2 .



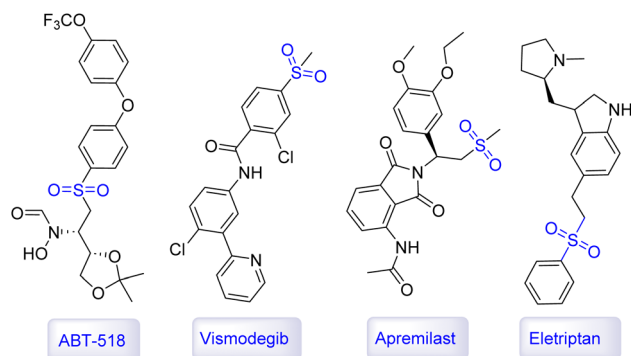


Fig. 13 Several bioactive examples of sulfone molecules.

including leprosy, dermatitis herpetiformis, and tuberculosis. It also played a role in the treatment of pneumocystis pneumonia (PCP), particularly in patients with compromised immune systems.¹⁹⁵ Another significant application of sulfones in medicine is in the treatment of inflammatory diseases. For instance, sulfasalazine is used in managing rheumatoid arthritis and ulcerative colitis, due to its anti-inflammatory properties.¹⁹⁶ These examples highlight the importance of sulfones in developing treatments that improve the quality of life for patients with infectious and chronic inflammatory diseases.¹⁹⁷ The structure several bioactive examples of sulfone molecules is shown in Fig. 13.^{198,199}

Their role in chemical synthesis is also significant; they act as intermediates in various reactions, such as the Ramberg-Bäcklund reaction, which is useful in the construction of biologically active molecules or functional materials.²⁰⁰ Industrially, sulfones are employed in the creation of polymers and agrochemicals, where their ability to function as electron-withdrawing substituents or leaving groups is particularly valuable.²⁰¹ The versatility of sulfones is further exemplified by their use in the synthesis of sulfonimides, which are important intermediates for accessing other organosulfur compounds. Overall, the applications of sulfones are diverse and continue to expand as research advances in these areas.

Magnetic catalysts have emerged as a significant tool in the synthesis of sulfones, offering a sustainable and efficient alternative to traditional methods. These catalysts are particularly advantageous due to their ease of separation using an external magnetic field, which simplifies the purification process and minimizes waste. In the context of sulfone synthesis, magnetic catalysts can facilitate various reactions, including the oxidation of sulfides, the coupling of sulfinates, and C-H bond activation. These advancements reflect the ongoing efforts to develop more sustainable and environmentally friendly approaches in organic synthesis, aligning with the broader goals of green chemistry. Overall, the integration of magnetic catalysts in sulfone synthesis aligns with the principles of green chemistry, aiming to minimize waste and reduce the environmental footprint of chemical processes.

In an innovative research endeavor led by Mustafa and his research team, a novel nanocomposite, $\text{Fe}_3\text{O}_4@\text{DABA-PA-CuBr}_2$, was successfully developed using a straightforward and cost-

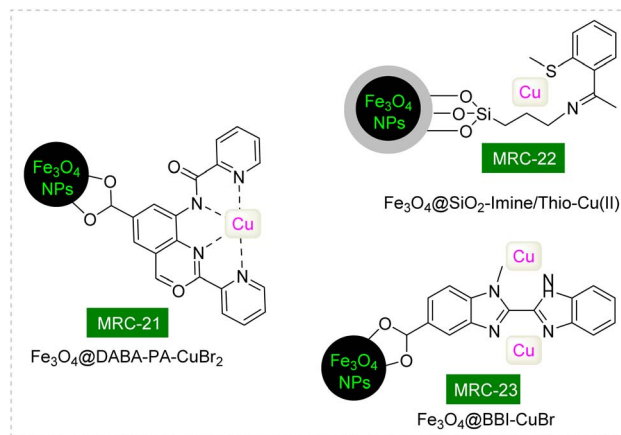
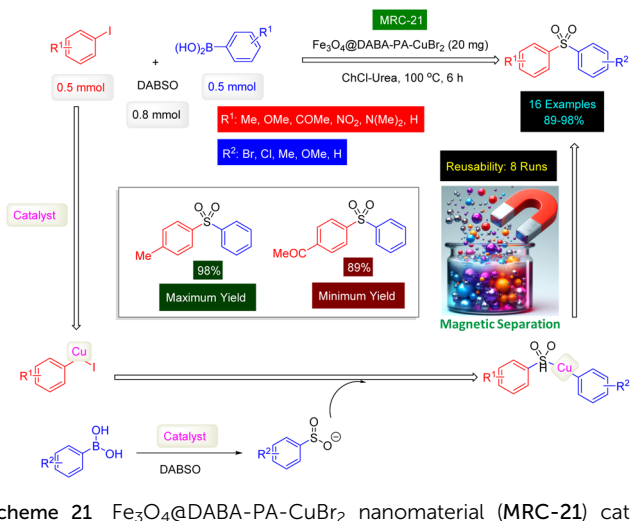


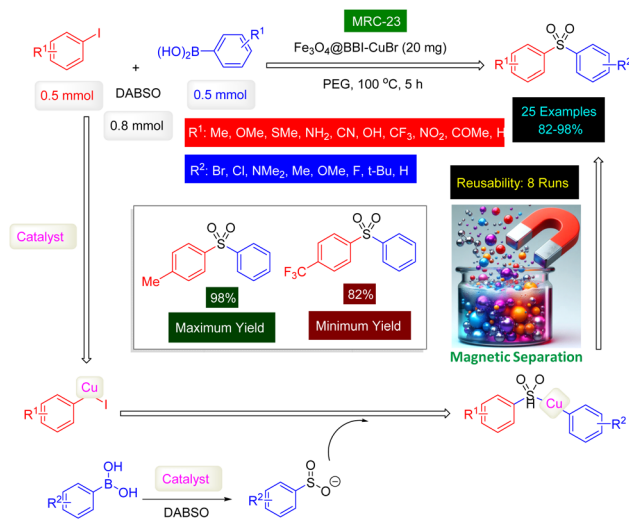
Fig. 14 Structure of magnetic catalysts in MCRs synthesis of diaryl sulfones.

effective approach.²⁰² This innovative method effectively immobilizes copper(II) bromide onto the surface of magnetic nanoparticles, which have been expertly modified with 3,4-diaminobenzoic acid and picolinic acid. This strategic enhancement significantly boosts the binding capabilities and functional properties of these nanoparticles, making them a powerful solution for a variety of applications. The study highlights the use of deep eutectic solvents (DESSs), which are recognized as green and environmentally friendly alternatives, serving as both catalysts and solvents in various chemical reactions. The application of the $\text{Fe}_3\text{O}_4@\text{DABA-PA-CuBr}_2$ nanocatalyst [MRC-21] (Fig. 14) in a ChCl-urea solvent system at a reaction temperature of 100 °C demonstrated an appealing and sustainable catalytic solution. This system facilitated the efficient one-pot three-component coupling reaction of aryl iodides, aryl boronic acids, and DABSO-acting as a source of sulfur dioxide-leading to the synthesis of a diverse array of diaryl sulfones. The results underscore the potential of this catalytic system in advancing green chemistry methodologies. Reusability tests confirmed that the $\text{Fe}_3\text{O}_4@\text{DABA-PA-CuBr}_2$ catalyst [MRC-21] has high catalytic activity despite being reused 8 times. The analysis conducted using VSM, TEM, and ICP-OES techniques on the reused $\text{Fe}_3\text{O}_4@\text{DABA-PA-CuBr}_2$ catalyst after eight catalytic cycles indicated that the fundamental properties of the catalyst remained remarkably stable. Specifically, the structural integrity, magnetic characteristics, morphology, and particle dimensions demonstrated minimal alterations, suggesting that the catalyst retains its effectiveness and stability over repeated uses (Scheme 21).

Fakri Mustafa and his team have made a groundbreaking advancement in catalysis with the creation of a magnetically recoverable copper catalyst. Discover an innovative catalyst designed through the precise immobilization of $\text{Cu}(\text{OAc})_2$ onto the surface of silica-coated magnetic Fe_3O_4 nanoparticles ($\text{Fe}_3\text{O}_4@\text{SiO}_2$). This advanced approach combines cutting-edge materials science with exceptional catalytic properties, making it a game-changer in the field.²⁰³ These nanoparticles are expertly functionalized with amine and thiol groups, enhancing

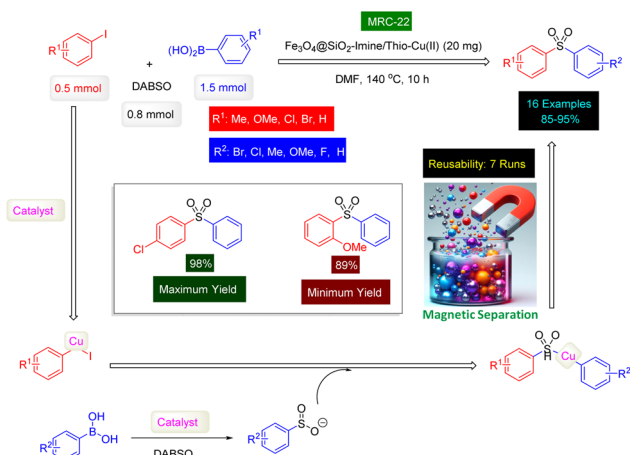


Scheme 21 $\text{Fe}_3\text{O}_4\text{@DABA-PA-CuBr}_2$ nanomaterial (MRC-21) catalyzed preparation of diaryl sulfone derivatives from DABSO.



Scheme 23 $\text{Fe}_3\text{O}_4\text{@BBI-CuBr}$ nanomaterial (MRC-23) catalyzed preparation of diaryl sulfone derivatives from DABSO.

their ability to support the copper species effectively. The resulting $\text{Fe}_3\text{O}_4\text{@SiO}_2\text{-imine/Thio-Cu(II)}$ nanocomposite [MRC-22] (Fig. 13) has been thoroughly characterized using a suite of advanced techniques such as FT-IR spectroscopy, SEM, EDX, TEM, XRD, VSM, ASS, and ICP-OES. The findings reveal that this nanocomposite exhibits remarkable catalytic activity in the synthesis of biologically active diaryl sulfones, showcasing its potential in real-world applications. This method offers numerous benefits: the catalyst can be easily separated using an external magnetic field, resulting in efficient processing. Moreover, it yields excellent results in terms of product yield and reaction time, while utilizing a non-toxic metal catalyst. These features make the $\text{Fe}_3\text{O}_4\text{@SiO}_2\text{-imine/Thio-Cu(II)}$ nanocomposite an attractive and practical option for synthesizing diaryl sulfones, promising to enhance the efficiency and sustainability of chemical processes in various applications. Reusability tests confirmed that the $\text{Fe}_3\text{O}_4\text{@SiO}_2\text{-imine/Thio-Cu(II)}$ catalyst [MRC-1] has high catalytic activity despite being reused 8 times (Scheme 22).



Scheme 22 $\text{Fe}_3\text{O}_4\text{@SiO}_2\text{-imine/Thio-Cu(II)}$ nanomaterial (MRC-22) catalyzed preparation of diaryl sulfone derivatives from DABSO.

To facilitate the preparation of diaryl sulfones, a novel copper(I) complex immobilized on magnetic Fe_3O_4 nanoparticles, referred to as $\text{Fe}_3\text{O}_4\text{@BBI-CuBr}$, was developed by Mustafa and his research team.²⁰⁴ This innovative nanomagnetic catalyst serves as both a green and highly efficient option for the sulfonylative-Suzuki-Miyaura cross-coupling reaction, which combines aryl iodides with arylboronic acids in the presence of DABSO as a source of sulfur dioxide. The characterization of the synthesized $\text{Fe}_3\text{O}_4\text{@BBI-CuBr}$ nanocatalyst [MRC-23] (Scheme 23) involved a range of advanced spectroscopic techniques, confirming its composition and structural integrity. Employing this methodology, the team successfully synthesized a variety of diaryl sulfones, achieving notable efficiencies, ranging from good to excellent yields. The reactions were conducted under environmentally friendly and mild conditions, enhancing the sustainability of the process. One of the standout features of the $\text{Fe}_3\text{O}_4\text{@BBI-CuBr}$ catalyst is its magnetic property, which allows for its easy separation from the reaction mixture using a magnet. This convenient separation method also enables the catalyst to be reused multiple times, making the process more economical and efficient. Additionally, when heterocyclic compounds were utilized as substrates, the resulting products were obtained with acceptable yields, demonstrating the versatility of this catalytic system in different reaction contexts. Reusability tests confirmed that the $\text{Fe}_3\text{O}_4\text{@BBI-CuBr}$ catalyst [MRC-23] has high catalytic activity despite being reused 7 times. The results obtained from the XRD, TEM, and ICP-OES techniques reveal that the reused catalyst demonstrates remarkable stability and retains its catalytic activity even after being utilized in seven consecutive cycles (Scheme 23).

3. Summary and outlook

This study has extensively explored the use of magnetically recoverable catalysts in the multicomponent synthesis of



organosulfur compounds, presenting a significant advancement in the field of catalysis. The employment of these catalysts has demonstrated an enhancement in the efficiency and selectivity of the reactions and a substantial reduction in environmental impact through the ease of catalyst recovery and reuse. Our findings underscore the effective integration of magnetic recovery technology with robust catalytic performance, providing a sustainable approach to complex synthesis processes. The magnetic properties of these catalysts allow for easy separation and recycling, significantly reducing catalyst loss and improving the overall efficiency and sustainability of the process. Additionally, magnetic catalysts often exhibit high selectivity and catalytic activity, improving reaction rates and product yields. Their versatility in various reaction conditions, combined with the ability to facilitate green chemistry practices such as solvent-free reactions, further enhances their appeal. The implications of this research for the catalysis community are profound. By leveraging the unique properties of magnetically recoverable catalysts, researchers can streamline the synthesis of organosulfur compounds, which are critical in various industrial applications, including pharmaceuticals, agrochemicals, and materials science. Recovering and reusing catalysts without significant losses in activity or selectivity presents a cost-effective and environmentally friendly alternative to traditional methods, which often involve tedious purification steps and generate considerable waste. The ability to manipulate magnetic catalysts with external magnetic fields also opens new possibilities for reaction control and optimization. While challenges remain in terms of scalability and the development of more robust magnetic materials, the progress made in recent years highlights the potential of these catalysts to revolutionize the synthesis of organosulfur compounds. Continued research and innovation in this field will likely lead to even more efficient, cost-effective, and environmentally friendly synthetic strategies, positioning magnetic catalysts as a key tool in the future of organosulfur chemistry.

3.1. Advantageous of the use of magnetic catalysts

✓ **Ease of recovery and reusability:** magnetic catalysts can be easily separated from reaction mixtures using an external magnetic field. This simplifies the recovery process and allows the catalysts to be reused multiple times without significant loss of activity.

✓ **Enhanced catalytic performance:** magnetic catalysts often exhibit high surface area and unique electronic properties, which can enhance their catalytic activity and selectivity in various chemical reactions.

✓ **Versatility:** magnetic catalysts can be used in a wide range of reactions, including oxidation, reduction, and condensation reactions. They are particularly useful in complex organic syntheses and industrial processes.

3.2. Importance in green chemistry

✓ **Reduction of waste:** the ability to recover and reuse magnetic catalysts reduces the amount of waste generated in chemical

processes. This aligns with the green chemistry principle of minimizing waste production.

✓ **Energy efficiency:** magnetic catalysts can often operate under milder conditions (lower temperatures and pressures), which reduces energy consumption and makes the processes more sustainable.

✓ **Reduced use of hazardous substances:** the use of magnetic catalysts can eliminate the need for hazardous solvents and reagents, further reducing the environmental impact of chemical processes.

3.3. Modern catalyst science

✓ **Nanotechnology integration:** the development of magnetic nanocatalysts involves advanced nanotechnology techniques, which allow for precise control over the size, shape, and composition of the catalysts. This leads to improved performance and new catalytic properties.

✓ **Innovative reactor designs:** magnetic catalysts enable the design of novel reactors, such as magnetic fluidized-bed reactors, which can enhance reaction efficiency and scalability.

✓ **Mechanistic insights:** research into magnetic catalysts provides valuable insights into reaction mechanisms, helping scientists to design more efficient and selective catalysts for various applications.

3.4. Future research directions

Looking ahead, the field of magnetically recoverable catalysts holds vast potential for further exploration and innovation. Future research could focus on several promising areas:

✓ **Catalyst optimization:** there is ongoing scope to enhance the design and functionality of magnetically recoverable catalysts. Research could focus on optimizing the composition, structure, and surface properties to increase catalytic activity, stability, and selectivity for a broader range of reactions.

✓ **Expansion of substrate scope:** exploring the use of magnetically recoverable catalysts in reactions involving a wider variety of substrates would be invaluable. It would expand the applicability of these catalysts in synthesizing a more diverse array of chemical products.

✓ **Mechanistic studies:** detailed mechanistic studies could provide deeper insights into the interactions at the molecular level between the catalyst and reactants. Understanding these interactions could lead to the rational design of catalysts tailored for specific reactions.

✓ **Integration with flow chemistry:** combining magnetically recoverable catalysts with continuous flow processes could revolutionize industrial-scale chemical synthesis. This integration promises to enhance the efficiency of chemical processes, reducing both time and resource expenditure.

✓ **Environmental impact assessment:** while these catalysts are designed for reusability and reduced waste, systematic studies assessing their long-term environmental impact are crucial. Research could focus on lifecycle analysis and the potential ecological footprint of the synthesis processes utilizing these catalysts.



By continuing to investigate these areas, the field of magnetically recoverable catalysts will not only refine its own techniques and applications but also significantly contribute to the broader discipline of sustainable chemical synthesis. The profound utility and versatile applications of these catalysts invite a collaborative, interdisciplinary approach to research, promising exciting developments in the synthesis of complex molecules and the advancement of green chemistry principles. The use of magnetic catalysts in the multicomponent synthesis of organosulfur compounds offers a compelling perspective on the future of green and efficient chemical processes. In conclusion, while there are hurdles to address, the potential of magnetic catalysts in the synthesis of organosulfur compounds is vast. With continued advancements in materials science, catalyst design, and reaction engineering, magnetic catalysts could become a cornerstone of sustainable and efficient synthetic methodologies, making significant contributions to both green chemistry and industrial applications.

Data availability

No primary research results, software or code have been included and no new data were generated or analyzed as part of this review.

Author contributions

Mostofa Kazemi was responsible for the supervision, conceptualization, and design of the study. Ramin Javahershenas played a pivotal role in overseeing the research project, providing strategic direction, guidance, and insight throughout the course of the study. Mostofa Kazemi was responsible for defining the key objectives, designing the study methodology, and maintaining the project timeline. Fadhil Faez Sead and Vicky Jain were responsible for the investigation and searching of articles. Anjan Kumar, Rekha M. M., and Mayank Kundlas played an instrumental role in the drafting of the manuscript, while Sofia Gupta and Mukesh Kumari articulated the findings from the research and presented the data in a clear, prepared figures. Mostofa Kazemi and Ramin Javahershenas conceived the idea, analyzed the data, and wrote the original draft. They also reviewed, edited, and proofread the draft. All authors contributed to the discussion and editing of the manuscript. The final version of the manuscript was read, reviewed, and approved by all authors.

Conflicts of interest

The authors declare no conflict of interest.

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