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APPLICATION OF NANOCOMPOSITES FOR DETECTION AND REMOVAL GLYPHOSATE OF WASTEWATER: A REVIEW[1]

APLICAÇÃO DE NANOCOMPÓSITOS PARA DETECÇÃO E REMOÇÃO DE GLIFOSATO DE EFLUENTES: UMA REVISÃO NARRATIVA

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ABSTRACT

The global trade in pesticides is expanding in response to the demands of the agroindustry. Glyphosate serves as the primary active ingredient in numerous herbicides, typically applied through spraying. However, the glyphosate not taken up by the plants accumulates in the soil and wastewater, posing potential risks to the environment and human health by contributing to the development of various diseases. Nanocomposites are a class of materials that exhibit enhanced properties by the synergistic effects arising from the combination of their constituents. This emerging technology holds significant promise in the field of detecting and eliminating pesticides and other chemical compounds from wastewater. A bibliographic review was carried out in the Periódicos Capes database with the descriptors "nanocomposites" and "glyphosate" with the Boolean operator "and". Articles available in full for consultation and original articles in English were included. Articles without full access and literature review articles were excluded. The timeline was selected automatically due to the articles found from 2017 to 2023. 42 articles were selected. Thus, we sought to evaluate the properties and efficiency of the nanocomposites used in the literature.

Keywords: Pesticides; nanotechnology; contamination.

RESUMO

O comércio global de pesticidas está se expandindo em resposta às demandas da agroindústria. O glifosato serve como o principal ingrediente ativo em vários herbicidas, normalmente aplicados por pulverização. No entanto, o glifosato não absorvido pelas plantas se acumula no solo e nas águas residuais, representando riscos potenciais ao meio ambiente e à saúde humana, contribuindo para o desenvolvimento de várias doenças. Os nanocompósitos são uma classe de materiais que exibem propriedades aprimoradas pelos efeitos sinérgicos decorrentes da combinação de seus constituintes. Esta tecnologia emergente é significativamente promissora no campo da detecção e eliminação de pesticidas e outros compostos químicos de águas residuais. Foi realizada uma revisão bibliográfica na base de dados Periódicos Capes com os descritores "nanocompósitos" e "glifosato" com o operador booleano "e". Foram incluídos artigos disponíveis na íntegra para consulta e artigos originais em inglês. Artigos sem acesso total e artigos de revisão de literatura foram excluídos. A linha do tempo foi selecionada automaticamente devido aos artigos encontrados de 2017 a 2023. Foram selecionados 42 artigos. Assim, buscamos avaliar as propriedades e eficiência dos nanocompósitos utilizados na literatura.

Palavras-chave: *Agrotóxicos; nanotecnologia; contaminação.*

1 Research Work

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INTRODUCTION

The global trade in pesticides continues to expand annually, with changes and innovations driven by market demands (Boccioni *et al.,* 2023). Glyphosate, an organophosphate compound, is a key ingredient in over 750 agrochemicals and serves as the primary active component in numerous herbicides (Barcellos, 2023). Glyphosate is officially authorized for sale in 130 countries, and it is estimated that around 919,000 tons of this pesticide are used each year (Valavanidis, 2018).

The initial findings about glyphosate emerged in the 1950s. It wasn't until 1970, however, that Dr. Phil Hamm made a crucial discovery regarding glyphosate's ability to bind with metal, thereby inhibiting a crucial metabolic pathway in plants (Dill *et al.,* 2010). Just months after Dr. Hamm's discovery, the product was already undergoing field tests. Subsequently, in 1971, the first glyphosate-based agrochemical was introduced to the market (Richmond, 2018).

Glyphosate is commonly sold in the form of a salt, which is obtained by formulating it with counter-ions such as potassium, ammonium, or isopropylamine. It is then diluted in water, mixed with surfactants and other additives, and applied by spraying onto the leaves of the target plant (Barcellos, 2023; Dill *et al.,* 2010; Richmond, 2018). Numerous studies have been conducted to investigate the use of glyphosate. While it is considered to be mildly toxic, there is evidence of its impact on the environment, primarily stemming from the resistance developed by certain weed species. Prolonged exposure to the pesticide or ingestion of contaminated food has also been found to affect humans (Junior *et al.,* 2002).

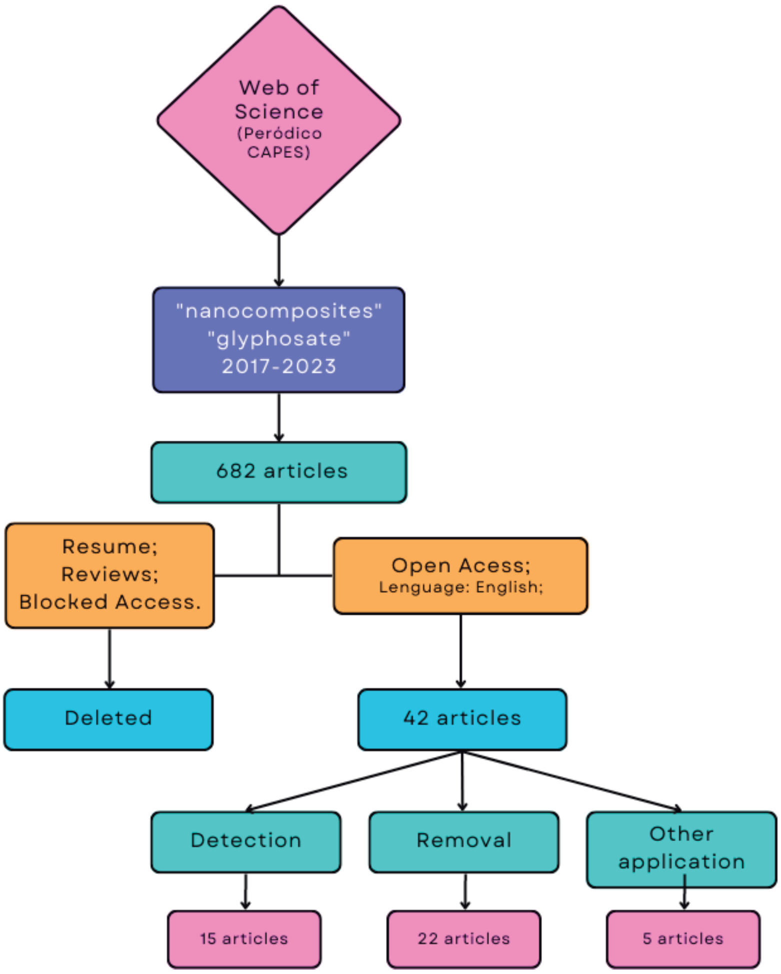
Glyphosate is commonly used on a variety of crops such as rice, coffee, sugarcane, corn, pasture, soy, sorghum, and wheat (Cabral, 2018). As a result of its application in agriculture, some of the residues from this pesticide, which are not taken in by the plant, persist in the soil, accumulate in water sources, and contribute to cross-contamination (CEE, 2019). The management and sustainable use of water resources necessitate wastewater treatment to ensure human and environmental safety in light of the presence of pesticides (Li *et al.,* 2021). Over the years, various technologies have been studied for the removal of complex compounds, particularly challenging-to-remove pesticides (Melo *et al.,* 2022).

Nanotechnology is an emerging and versatile field that encompasses various areas of research. One such area involves the utilization of nanocomposites for the effective removal of complex components from wastewater. Nanocomposites boast a high surface area, making them highly efficient for this purpose, and they also exhibit superior properties compared to conventional composites (Cao *et al.,* 2019). Thus, the objective of this work was to review the existing literature to seek further clarification about the nanocomposites used and the main existing nanotechnologies for the removal of glyphosate from wastewater.

METHODOLOGY

A literature review was conducted to explore the use of nanocomposites for the removal of glyphosate using the photocatalyst method. The bibliography was compiled by searching articles in the "Periódico Capes" database (Web of Science) using the keywords "nanocomposites" and "glyphosate." Only articles available in full and written in English were considered. The focus of the search was on selecting articles that discuss the photocatalyzed removal of glyphosate. Articles without full access, abstracts, review articles, and publications before 2017 were excluded. The search for articles was carried out in May 2023.

Figure 1 - Scheme of the methodology used.



Source: Built by the author.

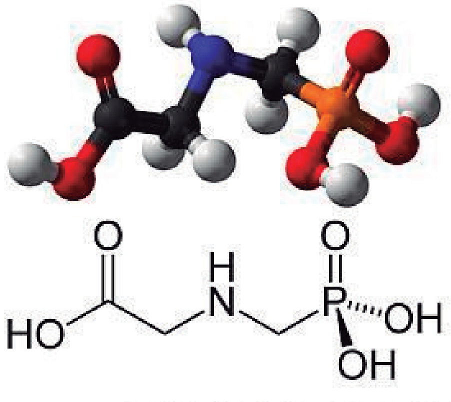
In the end, 42 articles were selected and separated into detection, 15 articles, removal, 22 articles, and for other applications, 5 articles.

The articles categorized as removal were further subdivided into the main methods used in research. Among the primary techniques highlighted were adsorption, catalytic, magnetic, membranes, oxidative, and photocatalytic reduction.

RESULTS AND DISCUSSIONS

Glyphosate, which is also referred to as N-(phosphonomethyl)glycine as per the International Union of Pure and Applied Chemistry (IUPAC), is an organophosphate compound with a molecular formula of $C_3H_8NO_5P$ and a molecular mass of 169.1 g mol^{-1} (IBAMA, 2019). This molecule exhibits high polarity, resulting in its high solubility in water and low solubility in common organic solvents such as ethanol and acetone (Junior, 2002; Cao *et al.,* 2019).

Figure 2 - Glyphosate chemical structure.



Source: Adapted from [13].

Glyphosate first came under scrutiny in the 1950s, and soon after, it was introduced as a herbicidal agrochemical (Dill *et al.,* 2010). Presently, glyphosate is a constituent of over 750 agrochemicals and serves as the primary active ingredient in numerous herbicides (Barcellos, 2023).

The herbicide containing glyphosate is sprayed on the leaves of weeds, disrupting the plant's nutrient absorption (CEE, 2019; Cabral, 2018). Any remaining glyphosate not absorbed by the plant's leaves ends up in the soil. Despite the plant's roots being unable to take in the glyphosate from the soil (Boccioni *et al.,* 2023; Barcellos, 2023), studies have detected glyphosate concentrations in both soil and groundwater (Valavanidis, 2018; Li *et al.,* 2021).

Many studies have examined the toxicological effects of glyphosate. While the World Health Organization (WHO) considers its acute toxicity to be low at 5600 mg/kg (Junior *et al.,* 2018; Jayasumana *et al.,* 2014), the accumulation of this herbicide in soil and water systems raises concerns about its potential impact on the environment and human health.

According to recent research, exposure to glyphosate, either through direct contact or indirect ingestion, has been linked to a range of health issues including gastrointestinal disorders, obesity, diabetes, heart disease, depression, infertility, microcephaly, cancer development, Alzheimer's, Parkinson's, and disruptions in hormonal balance (Cao *et al.,* 2019; Cabral, 2018).

As reported by BBC News Brasil in 2019, the European Union, Austria, and Germany have chosen to ban the use of glyphosate. Meanwhile, other countries like the Netherlands, Mexico, and France have taken steps to eliminate glyphosate usage within 5 years. In Brazil, glyphosate use is still permitted after reassessments by the National Health Surveillance Agency - Anvisa, but with specific restrictions (ANVISA, 2020).

Nanocomposites are materials that incorporate one or more materials, offering great promise due to their multifunctionality and the potential to enhance their properties through unique combinations (Zhu *et al.,* 2001) Additionally, nanocomposites are distinguished by the presence of at least one material at the nanometer scale, which is less than 100 nm. This results in a high surface area, making the material potentially useful for removing chemical pollutants from the environment (Ries *et al.,* 2023).

In researching nanocomposites, a comprehensive search was conducted to identify and potentially eliminate glyphosate from contaminated areas within the timeframe of 2017 to 2023. Table 1 presents the primary references concerning the detection of glyphosate.

Table 1 - References related to glyphosate detection.

Reference	Nanocomposite	Target
(Balaji *et al.,*2020)	BiOCl-BiOBr@Pt/Au semiconductor-plasmonic	Detection of pesticides.
(Do *et al.,* 2020)	Chitosan (CS), CS/ZnO, CS/GO	Detection of glyphosate.
(He *et al.,* 2022)	Co_3O_4/ZnO/Au	Detection of glyphosate.
(Luo *et al.,* 2022)	ratiometric fluorescent and smartphone-integrated colorimetric by carbon dots encapsulated porphyrin metal-organic frameworks	Detection of glyphosate.
(Ma *et al.,* 2023)	Gold Nanoclusters and Silica-Coated Carbon Dots-Assisted Ratiometric	Detection of glyphosate.
(Maji *et al.,* 2023)	CuCo2O4, Protonated-g-C3N4, 3D-Graphene Oxide Sheets	Detection of glyphosate.
(Qiang *et al.,*2022)	UiO-67 with porous carbon derived from Ce-MOF	Detection of glyphosate.
(Ren *et al.,* 2022)	Molecularly imprinted polymer and graphene oxide nanocomposite.	Detection of glyphosate in corn.
(Butmee *et al.,* 2021)	Screen-printed carbon electrode with silver nanoparticles, reduced graphene oxide.	Detection of glyphosate.
(Santanna *et al.,* 2022)	Activated biochar (AB4) and reduced graphene oxide (rGO).	Detection of pesticides and glyphosate.
(Thakkar *et al.,* 2022)	Graphene oxide, amino- and guanidine functionalized graphene oxide, exfoliated graphene, and commercial graphene nanoplatelets.	Detection of pesticides and glyphosate.
(Valle *et al.,* 2021)	Ag-doped ZnO/AgO	Detection of glyphosate.
(Wang *et al.,* 2022)	Ti3C2Tx/Cu-BTC	Detection of glyphosate.
(Zarejousheghani *et al.,* 2021)	(3-acrylamidopropyl) trimethylammonium chloride, [2-(acryloyloxy)ethyl] trimethylammonium chloride, and diallyl dimethylammonium chloride	Detection of glyphosate.
(Zhang *et al.,* 2020)	CuAl-LDH/Gr	Detection of glyphosate in water.

Source: Built by the author

The detection of glyphosate revealed a significant number of references featuring nanocomposites with graphene, often functionalized with metals such as copper, cobalt, and silver, as well as nanocomposites of metals like bromine, silver, gold, zinc oxide, titanium, and copper.

Additionally, nanocomposites are utilized for the removal of glyphosate, and the selected references can be found in Table 2.

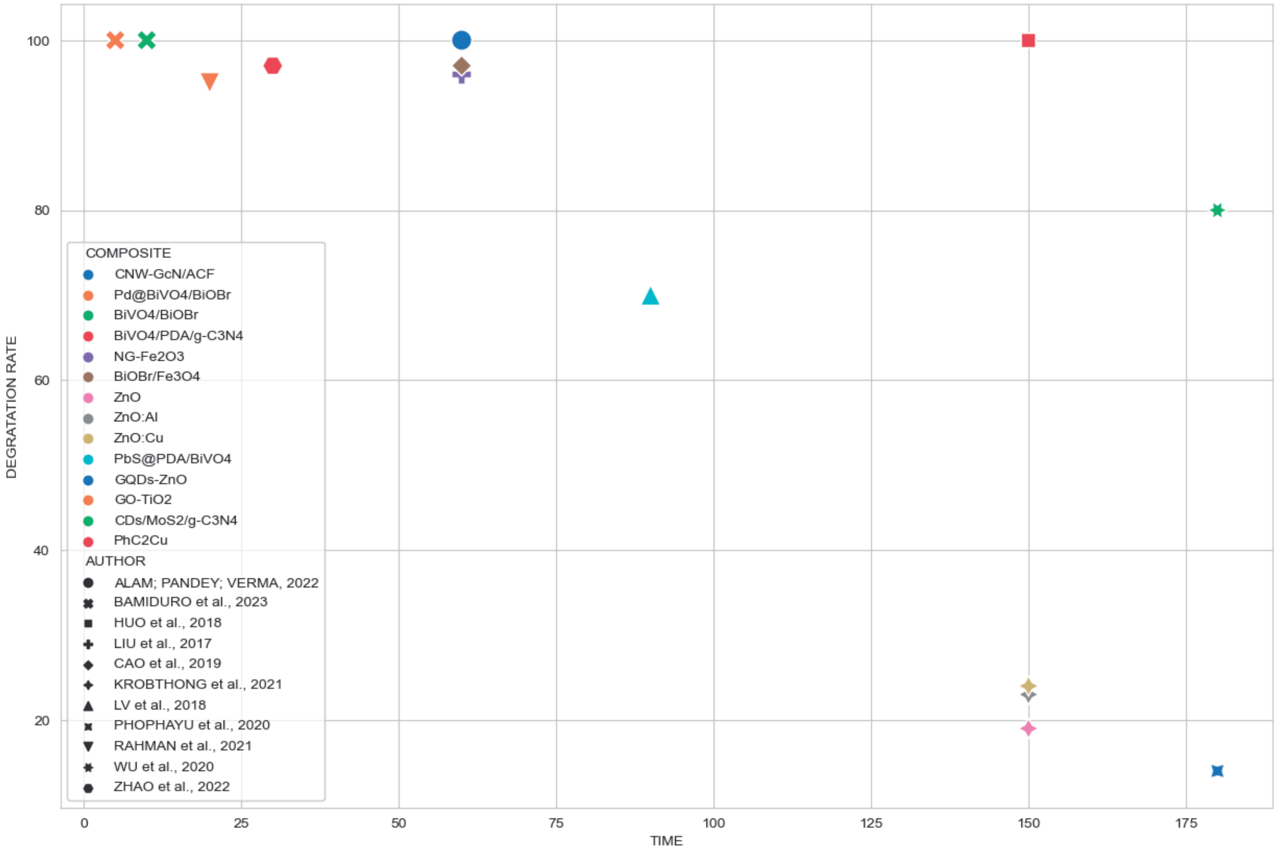
Table 2 - References related to glyphosate removal.

Reference	Nanocomposite	Target
(Zhou *et al.*, 2020)	Ag/ZrGP	Catalytic reduction for removing pesticides.
(Hosseini *et al.*, 2019)	Graphene oxide/TiO2	Remove glyphosate, trifluralin, and butachlor herbicides by polysulfone membranes.
(Briceno *et al.*, 2022)	CoFe2O4-chitosan-graphene	Glyphosate removal.
(Huang *et al.*, 2022)	LDH@Bt	Glyphosate and heavy metal removal.
(Milojevic-Ravik *et al.*, 2018)	Polyaniline/FeZSM-5	Oxidative degradation of herbicide glyphosate.
(Zue *et al.*, 2021)	$Fe_3Ce_1O_x$	Glyphosate removal.
(Li *et al.*, 2021)	Graphene doped with -Ti/-V/-Cr/-B/-Ca/-N/-Cu/-O/-Pd/-Pt	Removal of glyphosate by adsorption.
(Marin *et al.*, 2019)	Graphene oxide with $MnFe_2O_4$	Removal of glyphosate by adsorption.
(Santos *et al.*, 2017)	Graphene oxide with α and γ-Fe2O3.	Removal of glyphosate by adsorption.
(Yan *et al.*, 2022)	HFO@NDA88 and HFO	Removal of pesticides by adsorption.
(Yang *et al.*, 2017)	UiO-67/GO	Removal of organophosphorus pesticides by adsorption.
(Alam *et al.*, 2022)	CoNiWO4-gCN	Photocatalytic oxidation of glyphosate.
(LV *et al.*, 2020)	PbS@PDA/BiVO4	Photocatalytic degradation of glyphosate.
(Phopayu *et al.*, 2020)	Quantum dots-zinc oxide nanocomposites	Photocatalytic degradation of organic dyes and commercial herbicide.
(Tahman *et al.*, 2021)	Cellulosic fabric based on graphene/ TiO2 nanocoating.	Photocatalytic degradation of glyphosate.
(Wu *et al.*, 2020)	Cyclodextrins grafted MoS2/g-C3N4	Photocatalytic degradation of glyphosate and Cr (VI).
(Zhao *et al.*, 2022)	PhC2Cu nanowires	Photocatalytic degradation of glyphosate and methyl violet.
(Liu *et al.*, 2017)	Pyrrolic-N-doped graphene oxide/ Fe2O3 mesocrystal.	Photocatalytic degradation of glyphosate.
(Krobthong *et al.*,2022)	ZnO, ZnO/Al, and ZnO/Cu	Photocatalytic degradation of glyphosate.
(Cao *et al.*, 2019)	BiOBr/Fe3O4	Photocatalytic degradation of glyphosate in water.
(Huo *et al.*, 2018)	BiVO4/Polydopamine/g-C3N4	Photocatalytic degradation of glyphosate.
(Bamiduro *et al.*, 2023)	Pd@BiVO4/BiOBr	Photocatalytic degradation of glyphosate.

Source: Built by the author.

Several methods have been employed for removing glyphosate, with adsorption and photodegradation being the most effective. Other removal processes include catalytic reduction (Zhou *et al.,* 2020), magnetic removal (Briceno *et al.,* 2022; Huang *et al.,* 2022), and membrane filtration (Hosseini *et al.,*2019). A variety of references have utilized nanocomposites such as iron-cobalt (Briceno *et al.,* 2022), iron-cerium (Zue *et al.,* 2021), iron-graphene oxide (Marin *et al.,* 2019; Santos *et al.,* 2017; Liu *et al.,* 2017), and iron-bromine-bismuth (Cao *et al.,* 2019), as well as graphene doped with metallic elements like chromium, bromine, titanium, silver, and copper (Li *et al.,*2021). Additionally, nanocomposites containing silver (Zhou *et al.,* 2020), bismuth (Cao *et al.,* 2019; Huo *et al.,* 2018), titanium (Hosseini *et al.,* 2019; Li *et al.,* 2021; Tahman *et al.,* 2021), and zinc (Phopayu *et al.,* 2020) have been employed. For a visual analysis of glyphosate degradation, refer to Figure 3 below.

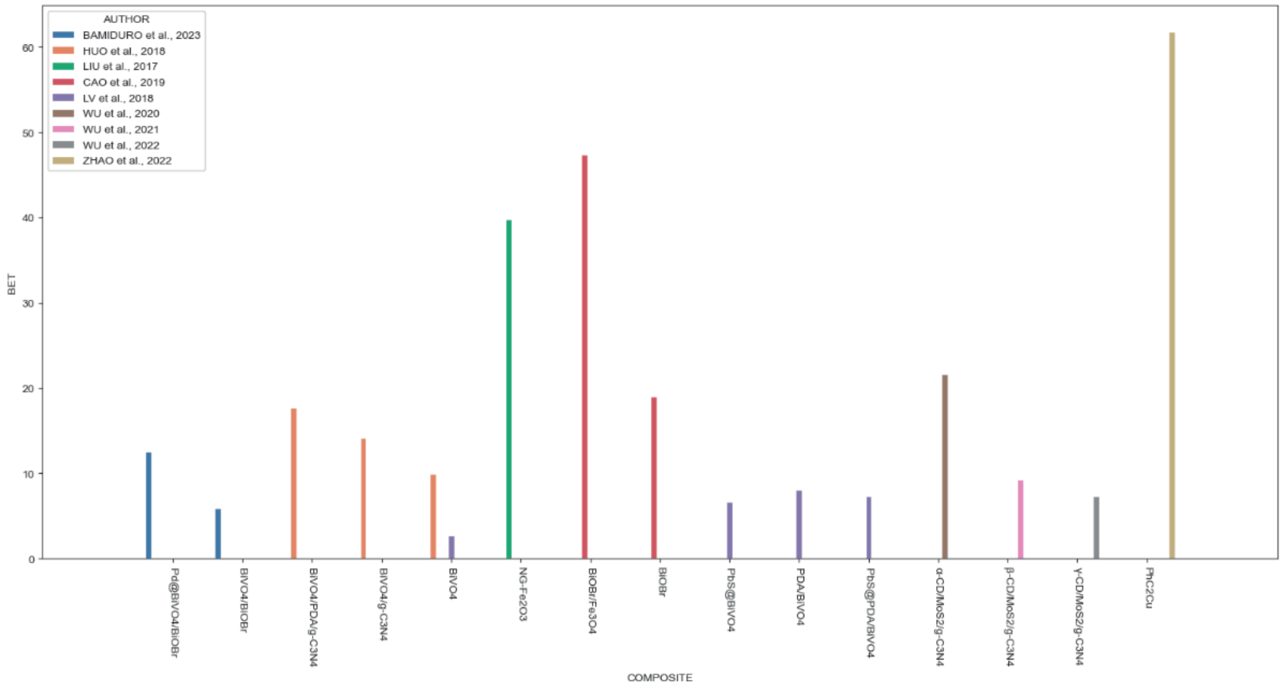
Figure 3 - Maximum Glyphosate degradation and time of reaction of different composites.



Source: Built by the author.

As per the data, it is noted that a majority of the nanocomposites achieved maximum glyphosate degradation within 60 minutes, while others achieved satisfactory degradation (over 50%) within 175 minutes. Additionally, the surface area of the nanocomposites was analyzed, as depicted in Figure 4.

Figure 4 - BET area of different composites.



Source: Built by the author.

The references also examined the surface area (BET) of photodegradation, highlighting the efficiency of nanocomposites with high surface area for degrading glyphosate. Additionally, the references delved into other applications of nanocomposites with glyphosate, which are detailed in Table 3.

Table 3 - References related to other applications of nanocomposite with glyphosate.

Reference	Nanocomposite	Target
(Gao *et al.,*2020)	TiO2/Biochar with Light-Switchable Wettability.	Adhesive to improve herbicide adhesion.
(Chen *et al.,* 2018)	Biochar, attapulgite (ATP), glyphosate (Gly), azobenzene (AZO), and amino silicon oil (ASO)	Controlled release of the herbicide
(Chi *et al.,* 2023)	Attapulgite (ATP), NH4HCO3, amino silicon oil (ASO), poly(vinyl alcohol) (PVA), and glyphosate (Gly)	Controlled release of the herbicide
(Chi *et al.,* 2021)	Palygorskite (Pal), ferroferric oxide (Fe3O4), glyphosate (Gly), and amino silicon oil (ASO).	Controlled release of the herbicide
(Zhang *et al.,* 2020)	Attapulgite (ATP), glyphosate (Gly), and calcium alginate (CA)	Controlled release of the herbicide

Source: Built by the author

Several studies have examined the use of nanocomposites for the controlled release of herbicides and pesticides, including glyphosate, in combination with adhesive to enhance adhesion (Gao *et al.,* 2018), amino silicon oil (Chen *et al.,* 2018; Chi *et al.,* 2021), ferroferric oxide (Chi *et al.,* 2021), and calcium alginate (Zhang *et al.,* 2020).

CONCLUSION

The review highlights the significant role of nanocomposites in effectively detecting and removing glyphosate from wastewater. It underscores the diverse range of nanocomposites being utilized for this purpose, emphasizing the need to develop environmentally friendly options that exhibit high efficacy in the detection and removal of glyphosate from water systems.

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