

NANOZEOLITES FROM THERMOELECTRIC WASTE FOR APPLICATION TO WASTEWATER TREATMENT: REVIEW¹

NANOZEÓLITAS A PARTIR DE RESÍDUOS DE TERMOELÉTRICA PARA APLICAÇÃO NO TRATAMENTO DE ÁGUA: UMA REVISÃO

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ABSTRACT

Along the years, nanozeolites have been proved extremely efficient in wastewater treatment. These nanostructured materials have a series of applications such as nanoadsorption, nanocatalysts to heterogenous photocatalyst, membrane separation processes and Fenton-like, showing a good removal of inorganic and organic pollutants. However, conventional synthesis of nanozeolite have some limitations, such as the formation of secondary pollutants by using organic supports or prolonged reaction times. In this context, the present work aims to provide an overview about the application of nanozeolites from coal fly ash in wastewater treatment. For this purpose, Science Direct platform was used, with the descriptors “Zeolite” AND “fly ash” AND “organic pollutants” AND “wastewater”, according to Boolean logic, and limiting to the last 5 years (from January 2015 to September 2020). Then, 22 articles were found being 70% of these studies corresponding to the nanoadsorption technology, 15% to Fenton-like treatment, 10% membrane separation processes, and only 5% to application in heterogenous photocatalysis, like supported catalysts. Therefore, it is possible to identify a wide application of these materials for application in wastewater treatment, being a potential alternative to green nanotechnology with sustainable development.

Keywords: Nanotechnology, Membrane Separation Processes, Heterogenous Photocatalysis, Nanoadsorption, Fenton-like Process.

RESUMO

Ao longo dos anos, as nanozeólitas tem-se mostrado extremamente eficientes no tratamento de água residuária. Estes materiais nanoestruturados possuem uma série de aplicação em nanadsorção, nanocatalisadores para fotocatalise heterogênea, processos de separação por membranas e processos do tipo Fenton, apresentando uma boa remoção de poluentes orgânicos e inorgânicos. Entretanto, a síntese convencional de nanozeolite apresentam algumas limitações como a formação de poluentes secundários pelo uso de suportes orgânicos ou os tempos de reação prolongados. Em vista disso, o presente trabalho tem por objetivo fornecer uma revisão sobre os principais trabalhos científicos envolvendo nanozeólitas a partir de cinzas de termoelétrica para aplicação no tratamento de água residuária. Para isso, fez-se uso da plataforma Science Direct, utilizando os descritores “Zeolite” AND “fly ash” AND “organic pollutants” AND “wastewater”, segundo a lógica Booleana, limitando nos últimos 5 anos. Assim, foram encontrados 22 artigos, sendo 70% deles relacionados à tecnologia de nanoadsorção, 15% a processos do tipo Fenton, 10% referentes a processos de separação por

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membranas e apenas 5% envolvendo fotocatalise heterogênea, com o emprego de catalisadores suportados. Por conseguinte, é possível identificar uma grande aplicação desses materiais para aplicação no tratamento de águas residuárias, sendo uma alternativa potencial da nanotecnologia verde com o desenvolvimento sustentável.

Keywords: *Nanotecnologia, Processos de Separação Por Membranas, Fotocatálise Heterogênea, Nanoadsorção, Processo do tipo Fenton.*

INTRODUCTION

WASTEWATER POLLUTION

Nowadays, it has been noticed a water quality deterioration, mainly caused to the climate change, population growth and industrial expansion (KUNDURU *et al.*, 2017). Wastewater contaminants can be organic or inorganic (ALJERF, 2018). Inorganic pollutants can be chromium, cadmium, lead, zinc, nickel, copper and mercury (ALI, 2012). In relation to the organic contaminants some examples are dyes, pharmaceuticals, hydrocarbons and aromatics compounds, including volatile compounds like benzene, toluene, ethylbenzene and xylene (CHENG *et al.*, 2016; DERIKVANDI; NEZAMZADEH-EJHIEH, 2017; SUNDARARAMAN *et al.*, 2018; SIVALINGAM; SEN, 2019; HASHEMI; ESLAMI; KARIMZADEH, 2019).

Paralely, conventional techniques of wastewater treatment are characterized by presenting low efficiency to removal these contaminants since they have high molecular weight (recalcitrant pollutants) and the complex and variable composition of the wastewaters (ABBAS *et al.*, 2015). Thus, nanotechnology seems promising in solving emergent problems faced in wastewater treatment, specifically the technical challenges related to the removal of contaminants such as toxic elements and persistent organic pollutants (KUNDURU *et al.*, 2017).

Nanotechnology is the control and manipulation of matter at atomic and molecular scale and its useful for developing novel technologies (SANDERS, 2018). Moreover, it is an interdisciplinary area, which possess a background on medicine, biology, chemistry and physics (BATTARD, 2012). A material at nanoscale is thought to have at least one of its dimensions between 1 and 100 nanometers (THERON; WALKER; CLOETE, 2008). Nowadays, the use of nanotechnology has been emerging fast due to the unique properties of the nanomaterials and their diversity nanomaterials can be used as catalysts in Photo-Fenton and Fenton-like processes as well as in heterogeneous photocatalysis (such as catalytic support or as nanocatalysts) (WESTERHOFF *et al.*, 2016), nano-adsorbents and components of membranes-based separation processes (ANIS *et al.*, 2020). Also, these nanomaterials can fix some drawbacks found in conventional water and wastewater treatments such as operational ones.

NANOADSORPTION

Adsorption is commonly defined as the ability that certain solids have in concentrate chemical species onto its surface (TIEN, 2019). In addition, it can be thought as a mass transfer phenomenon where a solute (or adsorbate) is transferred from the fluid phase to the surface of a solid adsorbent (GEANKOPLIS; HER-SEL; LEPEK, 2018). Thus, nanoadsorption is the use of nanomaterials as adsorbents (THERON; WALKER; CLOETE, 2008). A nanoadsorbent shows unique properties such as high specific surface area in relation to the pore volume, fast interaction with the solutes, good thermal and physicochemical stabilities and high adsorption capacity (MINTOVA; JABER; VALTCHEV, 2015). Like adsorption process, the nanoadsorption seems to be limited to equilibrium (WORCH, 2012). Once the equilibrium of the system adsorbate-adsorbent is reached, the adsorption cease, and the adsorbent capacity can be evaluated.

Equilibrium of nanoadsorption

Models such as Freundlich and Langmuir models can describe the equilibrium of nanoadsorption. They are empirical models used to suggest mechanism of adsorption and are so useful in describe profiles relating equilibrium concentration with adsorbent capacities (NASCIMENTO *et al.*, 2015). Moreover, these models are so applicable in water and wastewater processes, where the contaminants of major public concern are generally the solutes of interest. The following Equations represents the nonlinear (1) and linear (2) form of Langmuir model isotherms.

$$q_e = \frac{q_{max} \cdot C_e}{1 + K_L \cdot C_e} \quad (1)$$

$$\frac{1}{q_e} = \frac{1}{K_L \cdot q_{max}} \left(\frac{1}{C_e} \right) + \frac{1}{q_{max}} \quad (2)$$

Where,

q_{max} = maximum adsorbent capacity ($\text{mg} \cdot \text{g}^{-1}$);

K_L = Langmuir constant ($\text{L} \cdot \text{mg}^{-1}$);

q_e = adsorbent capacity ($\text{mg} \cdot \text{g}^{-1}$);

C_e = equilibrium concentration of adsorbate ($\text{mg} \cdot \text{L}^{-1}$)

This model assumes monolayer formation due to homogenous coverage of active sites of the solid nanoadsorbent by the adsorbate molecules. It also assumes that all sites are equal in energy, which accounts for no competition among the adsorbate molecules by the active sites (TIEN, 2019). Moreover, separation factor (R_L) can be used for predicting if the nanoadsorption will be favorable or unfavorable.

$$R_L = \frac{1}{1 + K_L \cdot C_0} \quad (3)$$

A favorable nanoadsorption is characterized by values of R_L between 0 and 1. Otherwise, the nanoadsorption is referred to as unfavorable.

Freundlich model is presented in the Equations 4 and 5 and assumes surface heterogeneity of the nanoadsorbent. In this model, there is multilayer formation onto surface of the solid material due to the coverage of active sites by the adsorbate molecules (STAVRINO; AGGELLOPOULO; TSAKIROGLOU, 2018). The parameter $1/n$ represents the intensity of nanoadsorption or strength of the adsorbate-adsorbent interaction, and K_F is empiric value (NASCIMENTO *et al.*, 2014). The values between 1 and 10 for n indicate a favorable nanoadsorption (WORCH, 2012).

$$q_e = K_F (C_e)^{1/n} \quad (4)$$

$$\log(q_e) = \log(K_F) + \frac{1}{n} \cdot \log(C_e) \quad (5)$$

Where,

q_{\max} = maximum adsorbent capacity (mg.g^{-1});

K_L = Langmuir constant (L.mg^{-1});

q_e = adsorbent capacity (mg.g^{-1});

C_e = equilibrium concentration of adsorbate (mg.L^{-1})

Kinect of nanoadsorption

Kinects of nanoadsorption is useful for describe the concentration profile of the adsorbate along the time. In addition, it can be used to keep track the amount of solute adsorbed onto nanoadsorbent along the time of overall process. For this purpose, two main kinetic models are overused - the pseudo-first (PFO) and the pseudo-second order (PSO) model (LEE *et al.*, 2018). The first model assumes a unique relationship of adsorbate molecule and active site of the nano-adsorbent, while the second model shows on a 2:1 relationship of adsorbate molecules and actives sites (LUEKING *et al.*, 2016; SONG *et al.*, 2016). It has become a common practice to adopt or assume the PFO as physisorption-related mechanism and the PSO as chemisorption-related mechanism (CHANG *et al.*, 2020). The Equations both in the derivative (6 and 7) and integrated (8 and 9) forms are presented below.

$$\frac{dq_t}{dt} = k_1(q_e - q_t) \quad (6)$$

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2 \quad (7)$$

$$q_t = q_1(1 - e^{-k_1 \cdot t}) \quad (8)$$

$$q_t = \frac{t}{\left(\frac{1}{k_2 \cdot q_2^2}\right) + \left(\frac{t}{q_2}\right)} \quad (9)$$

Where,

t = time elapsed (min)

q_t = amount adsorbed at time t (mg.g⁻¹);

C_t = adsorbate concentration at time t (mg L⁻¹)

q_e = amount adsorbed at equilibrium (mg g⁻¹);

k_1 = pseudo-first order rate constant (min⁻¹);

k_2 = pseudo-second order rate constant (L mg⁻¹.min⁻¹);

HETEROGENOUS PHOTOCATALYSIS

Conventional photocatalysts

Heterogenous photocatalysis is an Advanced Oxidative Process (AOPs) applied to water and wastewater treatment, with high efficiency of organic compounds onto mineralization process as well as of pathogens inactivation (TEOH; SCOTT; AMAL, 2012; XU *et al.*, 2017; GURUNG *et al.*, 2019; BRIONES *et al.*, 2020). Thus, general mechanism of heterogenous photocatalysis consists in the generation of hydroxyl radicals (HO[•]) onto catalytic surface (semiconductor material) under visible or ultraviolet radiation. These species can mineralize almost completely the organic matter, by nonselective redox reactions induced at surface of the photocatalyst (PATEL; YADAV; PATEL, 2013). For this purpose, various materials are used as photocatalysts such as titanium dioxide (TiO₂), niobium pentoxide (Nb₂O₅), magnesium oxide (MgO), zinc oxide (ZnO) and iron oxides (Fe₂O₃) (PATIL; NAIK; SHRIVASTAVA, 2011; FOTEINIS *et al.*, 2018; ONG; NG; MOHAMMAD, 2018; MURARO *et al.*, 2020). It is important to highlight that the TiO₂ is one of the most useful nanophotocatalyst used commercially due to availability, low-cost of acquisition, and safety (QU; ALVAREZ; LI, 2013; NGUYEN *et al.*, 2020).

Catalytic support

Nanocatalysts can be divided into monoliths, porous, supported, unsupported and molecular sieves (MU *et al.*, 2013; CUENYA; BEHAFARID, 2015; KAUSHIK; MOORES, 2017; ZHAO; JIN, 2018; CROSS *et al.*, 2019). The supported nanocatalysts are based on a dispersion of nanoparticles onto a less active substance, called support, which is so useful in heterogenous photocatalysis.

(HE *et al.*, 2018; BAENA-MONCADA *et al.*, 2019). The use and development of these nanocatalysts emerge as a solution of technical problem commonly faced when semiconductors are used individually (ZABIHI-MOBARAKEH; NEZAMZADEH-EJHIEH, 2015). For example, the use of TiO_2 has a main drawback the tendency of agglomeration of nanoparticles as well as the difficulty for separating the catalyst after the treatment (NEZAMZADEH-EJHIEH; BAHRAMI, 2014). Moreover, the presence of the support reduces these problems and can enhance the photocatalytic activity of the nanophotocatalyst (LI *et al.*, 2014). Additionally, the catalytic support might help in the adsorption of chemical species during the process (JAFARI *et al.*, 2016). It is worth to mention that the main advantage of these supported nanocatalysts is based on the reuse of the material after the treatment (TEIXEIRA *et al.*, 2016; DIMITRIJEVIC *et al.*, 2019; MURARO *et al.*, 2020). The reduction of only 2% of photocatalytic activity are reported in some scientific papers (GHASEMI *et al.*, 2016).

PHOTO-FENTON AND FENTON-LIKE PROCESS

Conventional Photo-Fenton process

Photo-Fenton process is an advanced water and wastewater system, which makes use of iron-based catalyst and hydrogen peroxide as an oxidant agent (CEN; NAN, 2018). This technology has been proved extremely efficient in the oxidation of persistent contaminant commonly found in wastewaters such as pesticides, dyes, organic pollutants, oils, and detergents (HASSANSHAH; KARIMI-JASHNI, 2018; KARCI *et al.*, 2018; BRINDHA *et al.*, 2018). Then, Photo-Fenton process is based on the generation of free radicals ($\cdot\text{OH}$ and $\cdot\text{O}_2^-$) due to redox reactions that take place on the metal catalyst surface (CARAM *et al.*, 2018). These highly reactive oxygen species react in a nonselective manner with different pollutants found in water and wastewater. The oxidation of Fe^{2+} to Fe^{3+} is which regenerate the photocatalyst (PAIVA *et al.*, 2018).

Fenton-like process

Analogously to Fenton and Photo-Fenton Process, Fenton-like process is based on the free radicals generation, which react nonselective with persistent wastewater pollutants (PAIVA *et al.*, 2018). The main difference between the conventional Fenton processes and Fenton-like is in the type of catalyst used (ZHOU *et al.*, 2018; SHI *et al.*, 2020). In the Fenton-like, supported catalysts like iron oxide onto zeolite or graphene oxide (GO) are generally encountered (ZHAO *et al.*, 2018). Moreover, catalytic support can fix some operational problems of Fenton reaction as well as overcome some commons drawback of the treatment (QIAN *et al.*, 2018). The iron-based catalyst can be found either on microscale or on nanoscale, most often as nanozerovalent iron, associated or not to a support (QIU *et al.*, 2020).

MEMBRANE SEPARATION PROCESSES

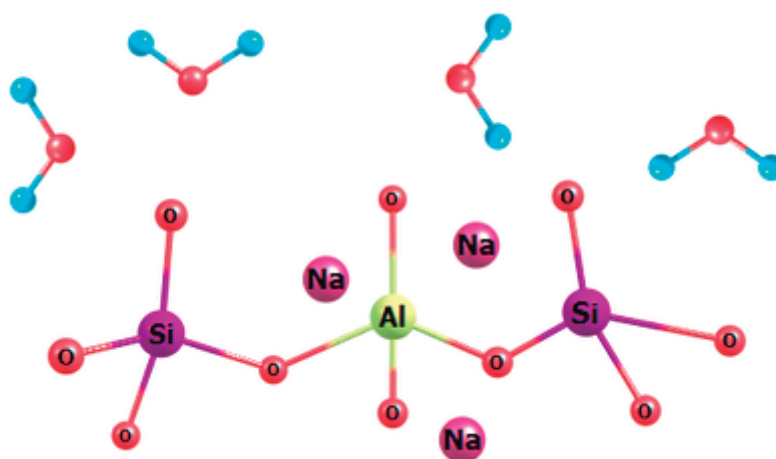
Membrane separation processes are commonly encountered at industrial scale, both applied to rejection of dyes, mono and divalent ion as well as to reduce the hardness of wastewater (MASTROPIETRO *et al.*, 2016; ANIS *et al.*, 2020). The membrane is composed of microporous material that can incorporate or not some fillers such as nanostructured zeolites, graphene and graphene oxide (GO), iron nanoparticles and so on (CHEN *et al.*, 2020; AMBRE *et al.*, 2019; LIU *et al.*, 2020). It is very promising for removal of contaminants from water at municipal level and it is divided into microfiltration, ultrafiltration, nanofiltration, and reverse osmosis (DAS *et al.*, 2014). The main synthetic methods of the membranes are either by precipitation method or phase inversion. Some works in the literature reveals high rejections of dyes, up to 95-99% (ORMANCI-ACAR *et al.*, 2020).

NANOSTRUCTURED ZEOLITES

Occurrence of nanozeolites and some applications

Nanostructured zeolites are microporous structures composed of aluminosilicate frameworks disposed in tetrahedral shape (JHA; SINGH, 2012). Then, their structures consist of SiO_4 and AlO_4^- units; with Si and Al atoms are centers of tetrahedral and the O atoms the vertex (KOOHSARYAN; ANBIA, 2016), according to the Figure 1

Figure 1 - Molecular geometry of a hydrated nanozeolite.



Source: Author.

Due to microcrystalline framework, nanostructured zeolites show unique proprieties that differ them from the commercial ones (MINTOVA; JABER; VALTCHEV, 2015; AMINI *et al.*, 2019), such as high selectivity to a series of pollutants, high specific surface area, uniform pore distribution

and high (hydro)thermal stability (ENNAERT *et al.*, 2016). Moreover, it is important to highlight that the pores uniformity is what differ the nanozeolites from any other porous material (GEANKOPLIS; HER-SEL; LEPEK, 2018). Thus, these nanomaterials have application on petroleum refining (JI; YANG; YAN, 2019), water and wastewater treatment (ANIS *et al.*, 2020), green chemistry (AGARWAL; PARK; PARK, 2019), energy storage (ZHANG *et al.*, 2019), gaseous and liquid-phase adsorption (PHAM; LEE; KIM, 2016; RASOULI *et al.*, 2012), ion exchange (LATEEF *et al.*, 2016) and catalytic processes (WEN *et al.*, 2019). Therefore, nanozeolites are composed of aluminosilicates (GEANKOPLIS; HER-SEL; LEPEK, 2018). However, titanium, boron, germane and phosphosilicate-based nanozeolites can be found in the literature (MI *et al.*, 2017; BIESEKI *et al.*, 2018; YU *et al.*, 2020).

Synthesis of nanozeolites

Nanozeolites can be found either in natural or synthetic form. The synthesis is mainly by hydrothermal method, although solvothermal and ionothermal methods can be used to synthesize the nanozeolites (STOCK; BISWAS, 2011). The main difference between these three methods is the type of the solvent used (aqueous, organic and ionic liquids) (LIMA *et al.*, 2019). Moreover, nanozeolite synthesis is extremely dependent of the pressure, temperature, pH, Si/Al ratio and time of reaction. (LIMA *et al.*, 2019). The synthesized materials can be classified such as low-silica, intermediate-silica and high-silica nanozeolites (JHA; SINGH, 2012), according to the Table 1.

Table 1 - Classification of nanozeolites in terms of Si/Al ratio.

Nanozeolite type	Si/Al ratio	Examples (structure)
Low-silica nanozeolite	Less than 2	Analcime (ANA), Cancrinite (CAN), Natrolite (NAT), Na-X (FAU), Philipsite (PHI), Sodalite (SOD)
Intermediate-silica nanozeolite	Between 2 and 5	Chabazite (CHA), Faujasite (FAU), Mordenite (MOR), Na-Y (FAU)
High-silica nanozeolite	Greater than 5	Zeolite-B (BEA), ZSM-5 (MFI)

Source: Author.

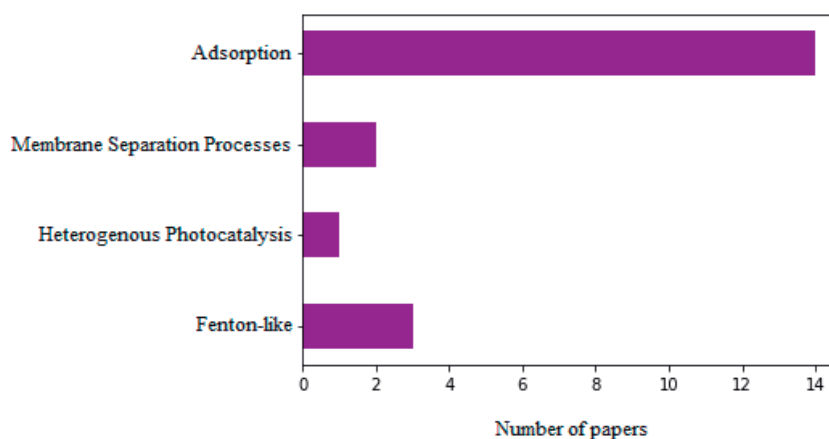
MATERIAL AND METHODS

This paper was developed based on the literature review about nanostructured zeolites from coal fly ash with application to wastewater treatment. For this purpose, the Science Direct platform was used. “Zeolite” AND “fly ash” AND “organic pollutants” AND “wastewater” were used as the descriptors of the research, according to Boolean logic. For this work was selected papers published in the last 5 years - from January 2015 to September 2020. Thus, 22 articles were found being 2 of them excluded for some reasons (they are either related to biomedical applications like drug delivery or referred to soil remediation).

RESULTS AND DISCUSSION

Figure 2 shows the results about the main published papers about nanozeolites from coal fly ash and applications, limiting the last 5 years (January 2015 to September 2020).

Figure 2 - Main published papers with nanozeolites from coal fly ash and applications (January 2015-September 2020).



Source: author.

According to Figure 2, it is noticeable that 70% of the studies are corresponding to the nano-adsorption technology, 15% referred to Fenton-like treatment, 10% membrane separation processes and 5% about heterogenous photocatalysis, which makes use of supported catalysts. As it can be seen, the number of researches on adsorption technology is remarkable compared to the others. It occurs mainly due to the versatility of nanomaterials used as nanoadsorbents, the relative low-cost of the synthesis of them (it can be made from waste), and the ease of operation. In addition, nanoadsorption technology seems to be very efficient in the removal of persistent organic pollutants of wastewater. Table 2 shows the discussion about the use of nanozeolite from coal fly.

Table 2 - Main published works using nanozeolite from coal fly ash between January 2016 and September 2020.

Research	Comment	Reference
Adsorption of CO ₂ and combustion products onto zeolite P made from coal fly ash	High CO ₂ uptake. Adsorption capacity at industrial scale equals 45 kg pollutant per ton of nanoadsorbent	MONASTERIO-GUILLOT <i>et al.</i> , 2020
Degradation of p-nitrophenol and p-nitroaniline by nano-CuO/Fly Ash Zeolite P by Fenton-like process	Fast adsorption of metals	VISA, 2016
Degradation of p-nitrophenol and p-nitroaniline by nano-CuO/Fly Ash Zeolite P by Fenton-like process	96% and 84% degradation of p-nitrophenol and p-nitroaniline was achieved with 6% copper supported onto zeolite	SUBBULEKSHMI; SUBRAMANIAN, 2017
Degradation of p-nitrophenol and p-nitroaniline by nano-CuO/Fly Ash Zeolite P by Fenton-like process	96% and 84% degradation of p-nitrophenol and p-nitroaniline was achieved with 6% copper supported onto zeolite	XIE <i>et al.</i> , 2017
Adsorption de COT onto Zeolite Y	90% of COT removal from aqueous solution, using 0.4 g of NZ-Y	HASHEMI; ESLAMI; KARIMZADEH, 2019

Removal of Cr ⁶⁺ from aqueous solution by membrane separation processes	Membrane with geopolymer-zeolite in its composition able to achieve 85.45% metal rejection	HE <i>et al.</i> , 2020
Adsorption of Phenol onto Faujasite (Na-Y nanozeolite)	Good removal of phenol at high concentration after 25 min.	MOHAMMED <i>et al.</i> , 2019
Degradation of Methylene Blue by heterogeneous photocatalysis	70% degradation after 80 min, using TiO ₂ supported on magnetic zeolite.	ALBERTI <i>et al.</i> , 2019
Adsorption of BTX (benzene, toluene, and xylene) onto NaP-1 nanozeolite	85-90% Benzene removal. Suitable adsorbent for removal for xylene and toluene from wastewater	SZALA <i>et al.</i> , 2015
Adsorption of 2-chlorophenol onto nanozeolite-copper	85% removal of the organic contaminant. Excellent specific surface area of the prepared nanoadsorbent (890 m ² g ⁻¹)	HUONG; LEE; KIM, 2016
Adsorption of NH ₃ -N and Methylene Blue onto nanozeolite (coal gangue) and activated carbon	Good removal for removal of dyes at high concentration in wastewater. Physisorption for NH ₃ -H adsorption and Chemisorption for dye adsorption	WANG <i>et al.</i> , 2018
Adsorption of Cr-NH ₃ and dyes onto clinoptilolite (CL-nanozeolite)	96.7% Cr-NH ₃ removal and 90.6% dyes removal after 60 min, using 60.4 mg of nanoadsorbent. High adsorption capacity (175.5 mg g ⁻¹)	ALJERF, 2018
Adsorption of cadmium from mining wastewater. Adsorption onto Na-A and Na-X nanozeolite from coal fly ash	60-90% removal of Cd ²⁺ from aqueous solution. High adsorption capacity (736.38 and 684.46 mg g ⁻¹), with chemisorption mechanism	SANTASNACHOK; KURNIAWAN; HINODE, 2015
Cr ⁶⁺ and Acid Blue dye from wastewater	96% dye removal and 92% Cr ⁶⁺ from aqueous solution. Physisorption for dye adsorption process and chemisorption for metal adsorption process	HAILU <i>et al.</i> , 2017
Congo red degradation by Fenton-like process	92-95% dye degradation using Fe supported on zeolite from coal gangue after 40 min. Pseudo-first order model of degradation	SUNDARARAMAN <i>et al.</i> , 2018
Adsorption of phenol onto nanozeolite-X/AC nanoadsorbent	Good adsorption capacity for phenol (37.92 mg g ⁻¹). Equilibrium has achieved after 100 min	CHENG <i>et al.</i> , 2016
Adsorption of Methylene Blue onto natural chabazite	80-96% removal of dye after 3 h and using 65 mg of nanoadsorbent	AYSAN <i>et al.</i> , 2016
Cd rejection by membrane separation process	Good Rejection after 153 min both using zeolite powder (54 mg) and zeolite fiber (150 mg) incorporated in the membrane	LI <i>et al.</i> , 2019
Degradation of phenol by Fenton-like process	100% phenol after 30 min using catalyst made from coal gangue waste (nanozeolite P and iron). 63% TOC removal and 92% TOC removal after 60 min	ARAKI <i>et al.</i> , 2019

Source: author.

NANOZEOLITES AND NANOADSORPTION

According to Table 2 and Figure 2, it is noticeable a remarkable use of nanostructured zeolites applied as nanoadsorbent. From January 2015 to September 2020, there were several works with application to adsorption of toxic or persistent pollutant from wastewater. As can be seen from Table 2, the main contaminants used as adsorbate are either heavy metal (cadmium, chromium, zinc, and lead)

or dyes. The target dyes include Methylene Blue, Congo Red and Acid Blue dye. Organic pollutants of phenol class (phenol, bisphenol A, chlorophenol) as well as nitrogen-based organic compounds are present in the related works. Moreover, nanostructured zeolite showed high adsorption capacity for dyes and organic compound yielding to excellent removal (up to 80-90%) of those pollutants from aqueous solution. In these studies, adsorption isotherms and kinetic model were applied to experimental data. To summing up, the adsorption of dyes onto nanozeolites seems to be most by physisorption, with PFO fitted to data. On the other hand, the adsorption of heavy metals onto nanozeolites were found to be mainly due to chemisorption, with the data in good agreement with Langmuir and PSO model. With respect to phenolic compounds, the predominance of either physisorption or chemisorption seems to be extremely dependent on the type of nanozeolite synthesized. It is important to mention that adsorbent capacity for the pollutants were in the range of 12.83 to even 754 mg g⁻¹ in the selected studies. Indeed, the high surface area of nanostructured zeolites play the major role in their good adsorbent capacity (QIAN; LI, 2015; SANTASNACHOK; KURNIAWAN; HINODE, 2015; HUONG; LEE; KIM, 2016; ALBERTI *et al.*, 2019) .

NANOZEOLITES AND HETEROGENOUS PHOTOCATALYSIS

From this investigation, only one work related to heterogenous photocatalysis was found. The advanced oxidative process has made use of a conventional TiO₂ nanoparticles supported onto magnetic zeolite synthesized from iron and aluminum industrial waste. The nanophotocatalyst showed outstanding properties such as excellent thermal and chemical stabilities as well as the excellent regeneration capacity. At specific experimental conditions (catalyst concentration equals 0.5 g L⁻¹, initial concentration of 0.05 g L⁻¹ of Methylene Blue, acidic pH, temperature of 298 K), 70% of dye degradation was achieved after 80 min of the treatment.

NANOZEOLITES AND FENTON-LIKE PROCESSES

Three works were found for Fenton-like processes using nanostructured zeolites from coal fly ash and coal gangue. In all of them, the nanostructured zeolites are used as support, which results in enhanced degradation of persistent or toxic water and wastewater contaminants. Using Nanozeolite P with 6% nano-CuO incorporated into it yields to 84% and 96% removal of p-nitroaniline and p-nitrophenol after 180 min at experimental conditions - H₂O₂ 30% v/v dose equals 10 mL L⁻¹, 500 mg L⁻¹ of catalyst CuO-NZP, acidic pH and 298 K. Congo Red dye (4 mmol L⁻¹) was found to be degraded in the order of 92 to 95% in wastewater by the presence of 0.861 mmol L⁻¹ H₂O₂, 30 °C, pH 3 and ferrous

concentration equals 0.2 mmol L^{-1} , after 40 minutes. Similarly, phenol removal from aqueous solution was reported when Na-A and Na-X nanozeolites (from coal fly ash) was used in the Fenton-like process. By this work, it was possible to conclude that activated carbon and pristine coal fly ash can be characterized as alternatives supports applied in Fenton-like processes. Therefore, in the last work a supported catalyst showed excellent activity and chemical/thermal stability, yielding to almost 100% degradation of phenol from zinc mining wastewater. At experimental conditions (200 μL of H_2O_2 , 30% v/v, 30 mL of phenol 50 mg L^{-1} , $60 \text{ }^\circ\text{C}$, and 5 mg Cu/ZSM-5), 63% and 92% TOC reduction were achieved after just 30 and 60 minutes. The nanozeolite were synthesized from coal gangue waste.

NANOZEOLITES TO MEMBRANE SEPARATION PROCESSES

Few membrane processes work-related were found. In this view, all of them deals with rejection of inorganic contaminants such as cadmium (Cd^{2+}) and chromium (Cr^{6+}). In one of them, a geopolymer associated to type Y nanozeolite is used for removing Cd^{2+} from aqueous solution at experimental conditions - flow rate of 0.5 mL min^{-1} , 100 kPa absolute pressure and slurry concentration equals $100\text{-}200 \text{ mg L}^{-1}$ of Cd^{2+} . For this purpose, 150 mg zeolite fibers and 54 mg zeolite powder were tested. Both membranes showed fast kinetic for rejection of the heavy metal (from 93 to 153 min), resulting in high rejections. In other study, a geopolymer associated to nanozeolite Li-ABW is used for removal of Cr^{6+} at pH 7, 10 kPa, and 1000 mg L^{-1} of feed concentration, resulting in 84.45% of metal rejection.

CONCLUSION

From the present study it was possible to look at the main applications of nanostructured zeolites synthesized from thermoelectric waste (fly ashes). As can be seen, the main precursor of this nanozeolites is the coal gangue residue, an aluminum-rich material. Also, it contains significant amount of silica in its composition, which make it suitable to the synthesis of nanostructured zeolites with application to wastewater treatment. Thus, the occurrence of these nanomaterials applied to removal of various contaminants (either organic or inorganic) from wastewater has been increase fast. It is justified by the unique properties of obtained nanomaterial such as high specific surface area, fast reactivity, and interaction with water pollutants. In addition, the excellent physicochemical and thermal stability serve as a compliment of the properties of interest in wastewater treatment. At the same time, it is worth to point out that the recovery capacity of the nanoadsorbents (especially the magnetic ones), supported photocatalysts, and membranes is remarkable.

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