



Aloe vera waste biomass-based adsorbents for the removal of aquatic pollutants: A review



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ABSTRACT

Aloe vera has been cultivated for many centuries for its beneficial properties, finding application in a wide range of medical and health products. Nowadays, the research has also focused on an alternative use of *Aloe vera* which is related to environmental applications such as clean water technology/wastewater treatment process. In recent years, biosorption has been shown to be a cost-effective and efficient alternative method for removing various pollutants from wastewater and water. This work provides a comprehensive review on using *Aloe vera* waste biomass-based sorbents, as well as modified counterparts, for the removal of heavy metals, dyes and other pollutants from aqueous media. The discussed biosorbents have been grouped in five categories based on the treatment of the *Aloe vera* leaves. Adsorption mechanisms, in addition to the significant factors influencing sorption capability like physical and chemical properties of the adsorbent, initial concentration, initial pH and temperature of the solution, dosage and contact time, have been discussed in detail. Furthermore, the applied equilibrium and kinetic models have been also summarized. The history, taxonomy, botany, and applications of *Aloe vera* are also presented in brief.

1. Introduction

Aloe vera has been recognized as one of the most prominent medicinal plants with various applications (Baruah et al., 2016). Because *Aloe vera* leaves contain numerous bioactive compounds, its usage has been associated with several health benefits since ancient times. Some of the reported properties include wound healing, anticancer, antioxidant, immunomodulatory and laxative (Reynolds and Dweck, 1999), amongst others. Nevertheless, it must be pointed out that the biological activity of these compounds is attributed to their synergist effect and not to a sole action of an individual substance (Avijgan et al., 2014; Hamman, 2008). Besides medicine and pharmaceuticals, *Aloe vera* is being valorized by the food industry, cosmetology and nanotechnology (Kumar and Yadav, 2009; Soltanizadeh and Ghiasi-Esfahani, 2015;

Yapar, 2017). Recently, *Aloe vera* and its by-products have attracted attention for employment in environmental applications.

Water is considered as an important and scarce commodity in all countries around the world. This substance is the source of life and is of the basic survival needs for human (Enoh and Christopher, 2015). A global issue is the quality of water which is deteriorating day by day mainly due to the anthropogenic activities, population growth, unplanned urbanization, rapid industrialization and unskilled utilization of natural water (De Gisi et al., 2016; Enoh and Christopher, 2015).

Intense industrial and agricultural activities result in the contamination of wastewater with various pollutants such as dyes, pesticides, toxic heavy metals, organic compounds, phenols, dyes, and other persistent organic pollutants (Anastopoulos and Kyzas, 2015a). These types of pollutants enter the food chain and are taxed with causing toxic

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effects, cancer, and diseases, thus affecting the aquatic biota and human health (Anastopoulos and Kyzas, 2015a). Moreover, wastewaters derived from municipalities and from various industries, which are highly loaded with various pollutants, have become of serious global environmental issues in the last decades (Anastopoulos et al., 2013). Therefore the removal of emerging contaminants of concern is now as ever important in the production of safe drinking water and the environmentally responsible release of wastewater (Grassi et al., 2012).

Compared to other purification technologies such as chemical precipitation, carbon adsorption, ion exchange, evaporation, and membrane filtration (Rajasulochana and Preethy, 2016), biosorption appears as a promising process because of its advantages in terms of low-cost operation, easiness of handling, avoidance of generating secondary pollutants (e.g. toxic sludge), and high efficiency over a wide range of pollutants (Anastopoulos and Kyzas, 2014; Javanbakht et al., 2014; Rao and Prabhakar, 2011).

There are many biosorbents exhibiting high adsorption efficacy for aquatic pollutants such as algae (Anastopoulos and Kyzas, 2015b; Bilal et al., 2018; Jayakumar and Govindaradjane, 2017), fungi (Dhankhar and Hooda, 2011), bacteria (Hansda and Kumar, 2015; Vijayaraghavan and Yun, 2008), plants waste such as maize, barley husk, jute, cotton stalks, rice husk, food crop straw (Saba et al., 2016), composts (Anastopoulos and Kyzas, 2015a; Anastopoulos et al., 2013), agricultural peels (Anastopoulos and Kyzas, 2014; Bhatnagar et al., 2015; Dadwal and Mishra, 2017; Salmani et al., 2017), olive oil industry waste (Anastopoulos et al., 2015; Bhatnagar et al., 2014), coffee residues (Anastopoulos et al., 2017b), barks (Şen et al., 2015), banana waste (Ahmad and Danish, 2018) wood waste (Saeed et al., 2005; Shin et al., 2007), and sugar industry waste (Anastopoulos et al., 2017a).

For the first time, the present review article summarizes and discusses the synthesis of *Aloe vera* waste biomass-based sorbents (raw, treated, ash, activated carbons) and their performance in the removal of different aquatic pollutants. Isotherm, kinetic, and equilibrium modelling have been discussed in details. Moreover, parameters which affect the biosorption process, such as the solution's pH, contact time, temperature and biosorbent's dose, have been also evaluated.

2. History, taxonomy and botany

The etymology of *Aloe vera* comes from a combination of Arabic and Latin words. “Alloeh” means “shining bitter substance” in Arabic and “vera” means “true” in Latin (Basmatker et al., 2011). The first recorded pharmaceutical use of *Aloe vera* has been depicted in a Mesopotamian clay tablet (1750 B.C.E.). Egyptians used it for skin infections (550 B.C.E) and, in 74 C.E., Pedanius Dioscorides, a Greek physician in the Roman army, reported that *Aloe vera* could heal wounds, hemorrhoids, infections and be used as a treatment for hair loss, in his book *De Materia Medica* (Shelton, 1991). Intensive cultivation started in the 1920s and in the 1960s Dr. Bill C. Coates extracted *Aloe vera* gel without losing the healing properties. Nowadays, Mexico is the leading country among the *Aloe vera* producers worldwide (Pal et al., 2013).

The taxonomy of *Aloe vera* is presented in Fig. 1. Its botanical family includes also other plants with established chemical properties. Based on the International Rules of Botanical Nomenclature, the official scientific name is *Aloe barbadensis* Mill. with *Aloe vera* (L.) Burm. F. as a synonym (Sánchez-Machado et al., 2017).

The *Aloe vera* plant is a perennial xerophyte, with thick, fleshy, sharp leaves that form a rosette at the stem. The green, triangle leaves (12–16 per plant) are up to 0.5 m long and can weight up to 1.5 kg when mature. They contain gel, with 98% water and 0.66% total solids, that arises from the parenchyma cells (Baruah et al., 2016; Maan et al., 2018; Shelton, 1991). Red, yellow, purple or pale striped flowers bloom from October to January and fruits develop during spring. The plant usually does not suffer from diseases, except the presence of black fungal spots or a soft bacterial rotting that may occur occasionally. It lasts long drought periods and cannot survive at low temperatures. *Aloe*

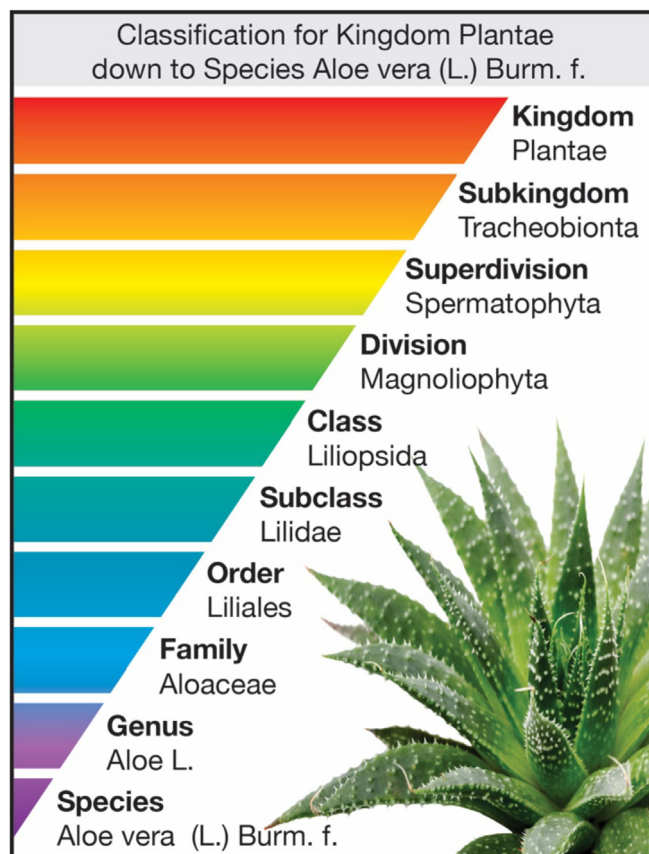


Fig. 1. The taxonomy of *Aloe vera*.

vera is native to Northern Africa and can also be found in the Mediterranean region, India, South America and South Africa along the Nile (Ahlawat and Khatkar, 2011; Maan et al., 2018).

3. *Aloe vera* applications

The content of *Aloe vera* is characterized by its abundance in compounds with biological activity, namely mono-, di- and polysaccharides, phenolic compounds, minerals, water- and lipid soluble vitamins, organic acids and lipids (Minjares-Fuentes and Femenia, 2017; Sánchez-Machado et al., 2017). Their beneficial effects and applications are summarized below with an emphasis on recent advances.

3.1. Food industry

According to Food and Drug Administration (FDA) *Aloe vera* can be safely consumed as “dietary supplement” in the U.S. In Europe, it is listed among “flavoring compounds” in accordance with European Commission Annex I of Regulation No. 1831/2003 (Javed and Atta-Ur, 2014).

The antimicrobial properties of *Aloe vera* have been valorized by the food industry for the production of herbal edible coatings. In addition to their activity against microorganisms, such coatings enhance shelf life of products since they prevent moisture loss (Hassan et al., 2017). *Aloe vera* gel coatings have been used for post-harvest protection of peaches, papaya, sapota, melon and other fruits (Hazrati et al., 2017; Kuwar et al., 2015; Padmaja et al., 2015; Riaie et al., 2017). Recently, Chin et al. investigated the potential of combining fish gelatin and various concentrations of *Aloe* gel, for the production of composite films (Chin et al., 2017). The composite films showed improved physical and functional properties and concentration-dependent antioxidative activity which makes active packaging with these films feasible.

The water retention property of *Aloe vera* improved the final quality

of low meat beef burgers, that usually lack consumers' acceptance due to cooking losses and size reduction (Soltanizadeh and Ghiasi-Esfahani, 2015).

Functional foods and beverages' market are growing due to the consumers' turning towards these products in order to address perceived nutritional shortfalls. Ready to serve drinks, milk, ice-cream, confectionery, cultured buttermilk etc. are examples of products that *Aloe vera* has been added, to provide its health benefits (Mishra and Sangma, 2017; Mudgil et al., 2016). Cuvas-Limon et al. conducted a review of a novel approach of symbiotic functional foods containing *Aloe Vera* (Cuvas-Limón et al., 2016). *Aloe vera* acts as the prebiotic source and could be combined with probiotic cultures to boost the nutritional value of the products.

3.2. Medicinal uses

Despite there is a vast literature regarding the topic, published over the years, and because it is not the main purpose of the present review, only a brief description of the medicinal uses of *Aloe vera* are given below. The uses are categorized according to organs or systems to which they are applied.

3.2.1. Skin

From wound healing to skin moisturizing, *Aloe vera's* effects on skin have been extremely studied. Various components of *Aloe vera* have biological activities related to skin conditions, namely acemannan, mannose-6-phosphate, anthraquinones, gibberellin, aloin or aloin A or barbaloin, saponin, etc. The aforementioned substances assist with wound healing and aging of skin by promoting collagenization, stimulation of blood flow, destruction of dead cells, etc. (Baruah et al., 2016; Javed and Atta-Ur, 2014; Sánchez-Machado et al., 2017) (Baruah et al., 2016).

3.2.2. Oral cavity

The antimicrobial activity of *Aloe vera* has been studied against oral pathogenic bacteria. Jain et al. tested the inhibitory effect of different *Aloe vera* gel concentrations on microorganisms that were isolated from periapical abscess and periodontal abscess of 20 patients (Jain et al., 2016). *Aloe vera* gel had comparable effects with antibiotics and, therefore, it is proposed as an effective anti-bacterial agent. Other conditions that *Aloe vera* showed beneficial results include lichen planus, aphthous stomatitis, oral submucous fibrosis, pulpotomy of primary teeth, prevention of dry sockets, obturation of primary teeth, disinfection of irrigation units, bleeding and painful gums, disinfection of gutta-percha cones, burning mouth syndrome and in radiated head and neck cancer patients (Arijani and Khoswanto, 2008; Digra et al., 2014; Renu et al., 2011; Saritha et al., 2010).

3.2.3. Gastrointestinal system

Consumption of *Aloe vera* has been linked with the treatment or improvement of various gastrointestinal diseases/conditions such as constipation, colitis, irritable bowel syndrome, peptic ulcer and colon cancer (Asadi-Shahmirzadi et al., 2012; Mahmoud and Hassanein, 2012; Suboj et al., 2012).

3.3. Cosmetology

The moisturizing and skin soothing effect of *Aloe vera* gel could not be undervalued by the cosmetic industry. Products such as soaps and cleansers, sunscreens, face antiaging creams, lotions, and coating for tissue papers contain *Aloe vera* from 1% up to 98% along with other essential oils and ingredients (Javed and Atta-Ur, 2014).

3.4. Veterinary medicine

Aloe vera has been introduced as a functional food ingredient in pet diets. Clinical studies on cats and dogs showed that the use of *Aloe vera* among other bioactive compounds, in the frame of a well-balanced diet,

provided various health benefits. More specifically, the consumption of *Aloe vera* enhanced the eyesight health by reducing corneal keratinization and tear/mucus production, improved oxidative imbalance and ameliorated biochemical parameters which are inextricably linked to the well-being of pets (Di Cerbo et al., 2017). The use of fermented *Aloe vera* has been proposed as an alternative feeding solution for pigs (Mi et al., 2018).

3.5. Phytoremediation applications

The potential phytoremediation ability of *Aloe vera* has been investigated recently. Phytoremediation is a biological method of removing toxic heavy metals from polluted soils, using plants as adsorbents. Due to its eco-friendly character, low cost and easiness to use, this technology has gained attention since the scientific community and competent authorities face the problem of pollution as a great challenge that needs effective solutions. Gavi et al. tested the ability of *Aloe vera* to improve the quality of domestic waste water, in a region of Pune India, regarding B.O.D, C.O.D, pH, D.O, and conductivity (Gavi et al., 2016). Results showed that *Aloe vera* reduced the contaminants in the treated wastewater at almost 30%.

In another study, the concentration of zinc, cadmium and lead in the plant parts was analyzed and compared to the corresponding levels of these elements in the soil at three different regions in Iran (Panahpour et al., 2013). According to the results, zinc concentration seems to have an inverse analogy to wet weight of gel, cadmium is stored preferably in the gel as well. Likewise, *Aloe vera* has been used, as an indigenous plant of the Niger Delta region, for phyto-extraction of zinc, iron and lead from soil samples obtained from the area of Nigeria that have been contaminated with the aforementioned metals (Tanee and Amadi, 2016). The root system presented higher accumulation ability of the elements, compared to the plant shoots. The metal translocation factor value was below 1 and therefore *Aloe vera* cannot be characterized as a hyper accumulator, but results indicate a phytoremediation potential. On the contrary, Shokri et al. with their experimental work on the adsorption of trivalent and tetravalent chromium, copper, lead, cadmium and nickel showed that transition factor values of *Aloe vera* for all elements were higher than 1 (Shokri et al., 2016).

4. *Aloe vera* waste-adsorbents

4.1. Categories of the reported *Aloe vera* derived materials

The reported *Aloe vera* derived materials were not obtained following a specific treatment protocol. Different drying procedures, chemical or thermal treatment have been followed. Moreover, the decoration/functionalization of *Aloe vera* with magnetic nanoparticles has been also presented. To this end, and for the sake of comparison, the materials in this study were organized in 5 general categories (Fig. 2). All the studied materials were obtained as powder with specific range of particle size in every case. Only in one study, the use of the as-obtained juice from the interior jelly part of the leaves after the removal of skin was reported.

The first category includes the materials that were not treated in any way (NT), while the second one includes the received biomass/powder after air drying at temperatures less than 110 °C (AD). In the third class, the materials obtained after the chemical treatment with strong acids (CT) of some of the above-mentioned air dried biosorbents (AD category) were collected. The fourth category includes all the obtained materials after thermal treatment (TT), in air or at inert atmosphere. The materials of the TT series are referred to at the original papers as well as herein as ash or carbons, depending if during the thermal treatment oxygen was present or not. Finally, the carbonized counterparts that were functionalized/decorated with magnetic nanoparticles or chemically activated are collected in a separate category (CF). The latter class of the materials is ultimately more interesting, since the magnetic property with the addition of small amount of inorganic phase can be a useful asset in real-life applications for

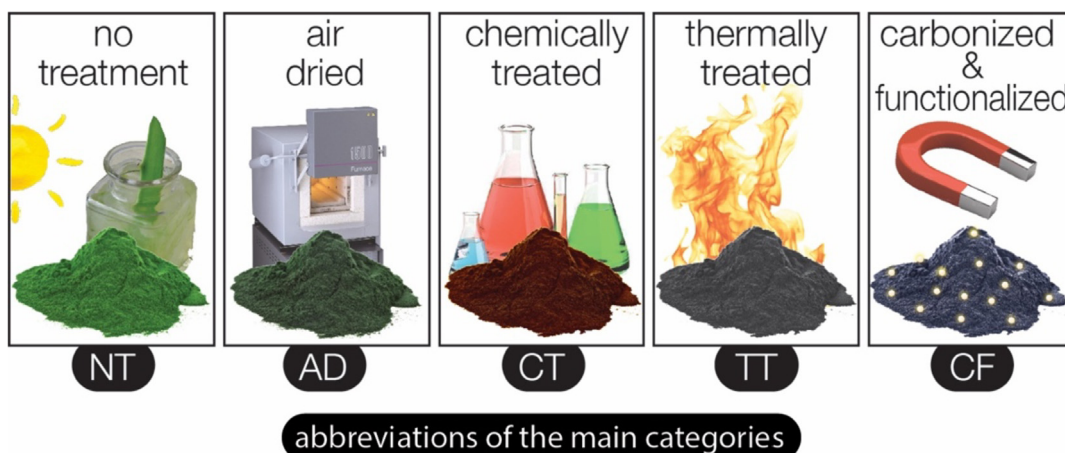


Fig. 2. A schematic representation of the 5 categories of the *Aloe vera* derived adsorbents.

their separation after use (Saroyan et al., 2017).

4.2. Synthesis of the materials

In order to conduct a meaningful comparison of materials that were

obtained with similar synthetic procedures but from different raw biomass or waste, the detailed synthetic procedure of all the herein discussed materials are included in this section. In Fig. 3, all the *Aloe vera* derived samples were collected and categorized based on the above-mentioned five categories. Since the size of the used adsorbent

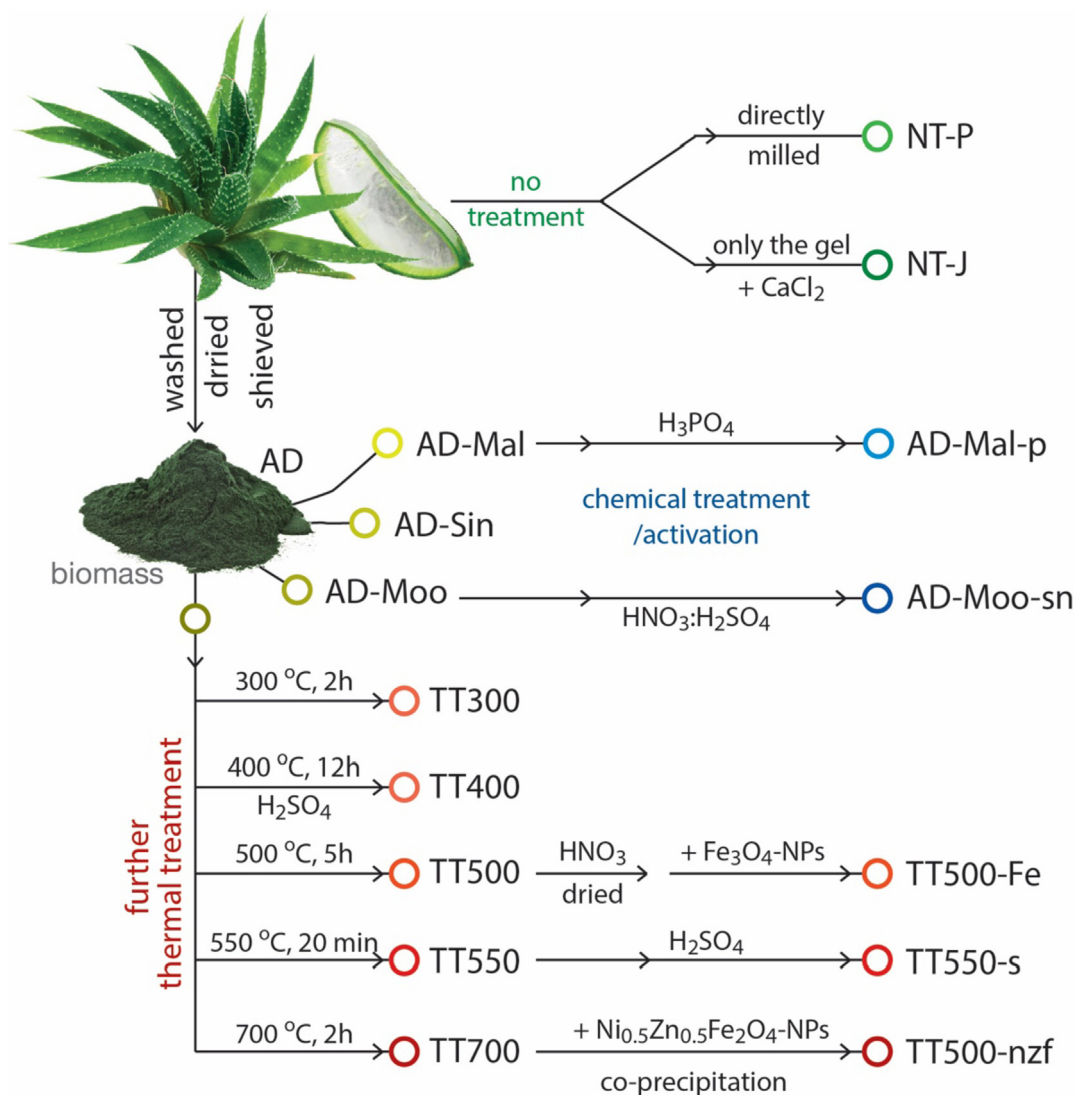
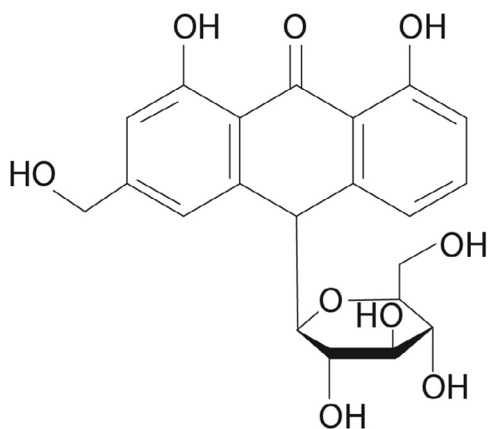


Fig. 3. Categories of all the *Aloe vera* waste-biomass derived materials reported in this review.



Scheme 1. The molecule of Aloin (known also as barbaloin).

has been found to play a crucial role on the adsorption performance (Giannakoudakis et al., 2016), the size of the used particles will be also presented, upon availability at the original papers.

4.2.1. No treated materials (NT)

The materials in this category were used before any prior drying. However, avoiding this strategy is recommended, since presence of aloin in high level (up to 6.6% w/w of the dry *Aloe vera* leaves) is most likely, in such material. Aloin (Scheme 1), known also as barbaloin, is an anthraquinone glycosyl which was banned from the Food and Drug Administration of USA in 2002, since it is linked as potential carcinogenic agent. Two samples from this category were evaluated as adsorbents:

NT-P: This sample was prepared by milling and sieving the as-collected biomass/waste, without any further physical or chemical treatment. The used particles were between 75 and 300 μm (Pragathiswaran et al., 2013).

NT-J: The leaves were washed under tap water to remove undesired impurities. The thick epidermis or skin was carefully separated from gel part or parenchyma of the leaves. The latter part was smashed and the formed transparent liquid/juice was preserved in glass bottles. The juice was used with CaCl_2 mixture for the experiments (Shyam and Kalwania, 2014).

4.2.2. Air dried biosorbents (AD)

This category includes biomass that was obtained from *Aloe vera* leaves/waste after washing with deionized water to remove the adhering moieties and undesired impurities (such as dust), followed by the removal of the gel and color. The received materials were dried at ambient conditions, but at different temperature in each report. In all cases, the temperature was less than 60 °C, in order to remove the water and to preserve the organic components. Different kinds of grinders and sizes of sieves were also used. In Table 1, all the reported materials and details about the AD treatment process, are summarized. The air-dried samples of this series of the powder are abbreviated with AD, following by -Xxx from the first 3 letters of the first author surname. It is worth

Table 2

The reported *Aloe vera* biomass-based materials obtained after thermal treatment.

Abbreviation	Thermal treatment procedure ^a	Air presence during the thermal treatment	Ref.
TT300	105 °C (10 h)/300 °C (2 h)	Yes	(Malakootian et al., 2014)
TT400	400 °C (12 h) + H_2SO_4 1:1 (w/v)	Yes	(Arivoli et al., 2008)
TT500	110 °C (15 h)/500 °C (5 h)	Yes	(Abedi et al., 2016)
TT550	150 °C (24 h)/550 °C (20 min)	No	(Basiri et al., 2015; Khaniabadi et al., 2015, 2016a, 2016b, 2017)
TT700	80 °C (24 h)/700 °C (2 h)	No	(Beigzadeh and Moeinpour, 2016)

^a The conditions prior to the slash refer to the initial drying procedure, while the latter part to the temperature and the duration of the final thermal treatment.

Table 1

The reported materials obtained after air drying at mild conditions.

Abbreviation ^a	Temperature and duration ^b	Powder size (μm)	Country of origin	Citation
AD-Mal (2)	30 days R.T. shadow/50–60 °C, 3 h	53–74	India	(Malik et al., 2015a, 2015b)
AD-Moo (2)	5 days in sun/50 °C, 2 h	75–125	Iraq	(Moosa et al., 2016a; b)
AD-Sin	–	0.2–0.3	India	(Singh et al., 2017)

^a Numbers in parentheses refer to the number of papers in which the material has been mentioned.

^b The use of slash is to separate different treatment steps. R.T.: refers to Room Temperature and Pressure. “–”: details not presented.

pointing out that AD-Moo revealed a surface area of 13.8 m^2/g . For the other materials of the air-dried series, no porosity details were reported.

4.2.3. Chemically treated dried powder (CT)

The effect of chemical treatment with strong acids has been studied in the case of AD-Mal and AD-Moo. The former one was treated with phosphoric acid (1 M for 12 h) and it is referred to as AD-Mal-p. AD-Moo was also activated by nitric and sulfonic acid mixtures of ratios [1:2], [1:1], and [2:1]. The material obtained by the treatment with the latter $\text{HNO}_3\text{:H}_2\text{SO}_4$ ratio revealed the highest surface area (24.6 m^2/g) and it is referred herein to as AD-Moo-sn. In both cases, the acid activated materials were extensively washed with distilled water until stable pH and then dried between 80 and 90 °C for 6–12 h.

4.2.4. Thermally-treated *Aloe Vera* biomass (TT)

This category contains the derived materials from *Aloe vera* biomass that were washed and dried under mild conditions, and afterwards were thermally treated at different temperatures, ranging from 300 to 700 °C. The herein reported materials are abbreviated as TTxxx, where ‘xxx’ stands for the temperature in which the thermal treatment took place. TT300, TT400, and TT500 were synthesized under the presence of oxygen, while TT550 and TT700 in an oxygen deficient atmosphere. Depending on the temperature and the presence or absence of air (oxygen) during the synthesis, the materials are referred to at the original reports either as ash or carbon. Unfortunately, no characterizations of the structural features have been provided in most of the cases in order to obtain a better insight into the real nature of these materials. Porosity analysis was presented only in the case of TT700 decorated with inorganic nanoparticles. The detailed synthetic routes for all the TTxxx series of materials, as well as the further modification/activation, are collected in the following part and can be seen additionally summarized in Table 2. Some representative scanning electron microscopy images (in the cases that they were available) can be seen in Fig. 4.

TT300 (Malakootian et al., 2014): Initially, the gel of *Aloe vera* leaves was extracted. The remaining part of the leaves was washed with distilled water for several times to remove all the dirt particles, and afterwards was dried at 105 °C for 10 h. The studied ash was obtained after thermal treatment at 300 °C for 2 h.

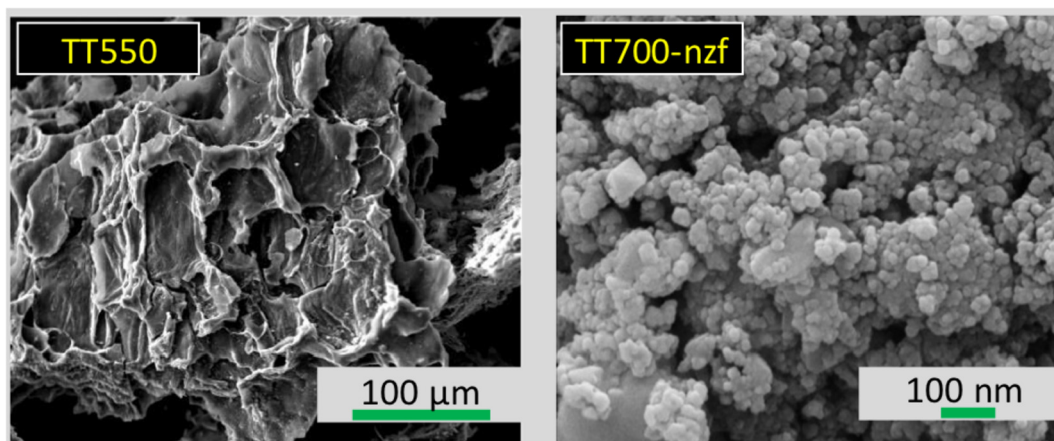


Fig. 4. SEM images of *Aloe vera* leaves carbonized at 550 °C (TT550) and carbonized at 700 °C followed by decoration with $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ nanoparticles (TT700-nzf). Adapted with permission from references [Khaniabadi et al., 2016b; Beigzadeh and Moeinpour, 2016]. (Copyright 2016 Elsevier and 2016 Elsevier).

TT400 (Arivoli et al., 2008): Air-Dried leaves of *Aloe barbadensis* Mill. were thermally treated for 12 h in a furnace at 400 °C with concentrated sulfuric acid in a ratio of 1:1 (w/v). The obtained material was washed with distilled water (until constant pH of the slurry was achieved) and was dried at 100 °C for 4 h. This material is referred to as a carbon in the original study, although it is not clearly mentioned if the thermal treatment had taken place in the absence of air. Moreover, no porosity data were reported.

TT500 (Abedi et al., 2016): The *Aloe vera* leaves were washed with deionized water and dried in a conventional oven for 15 h at 110 °C, while the latex of the leaves were drained previously. The obtained material was thermally treated for 5 h at 500 °C in a muffle furnace.

TT550 (Basiri et al., 2015; Khaniabadi et al., 2015, 2016a, 2016b, 2017): After the separation of the gel part from the *Aloe vera* leaves, the remaining part was washed with DI-water and dried for 24 h at 150 °C. The dried material was crushed, and the obtained particles of size 30–60 μm were carbonized for 20 min at 550 °C in an airless atmosphere.

TT700 (Beigzadeh and Moeinpour, 2016): *Aloe vera* shells were washed and dried at 80 °C for 24 h in an air oven, and then were ground and sieved (2–3 mm). The obtained sieved powder was carbonized at 700 °C (with a heating rate of 10 °C/min for 2 h) in an oxygen deficient atmosphere.

4.2.5. Carbonized and functionalized materials (CF)

Carbonized counterparts of some of the above mentioned thermally treated materials were further functionalized/decorated with magnetic nanoparticles or chemically activated. The decoration/functionalization of carbon or textiles with inorganic phases has been reported as an effective strategy in order to obtain materials with improved removal capabilities (Deliyanni and Bandosz, 2011; Gallios et al., 2017; Kyzas et al., 2014; Wallace et al., 2017).

TT500 was decorated with iron oxide nanoparticles in order to introduce to the final material magnetic properties. At the first stage, the as-prepared TT500 was activated with HNO_3 , washed with hydrochloric acid and DI-water in order to remove the precipitated inside the pores salts and/or traces of acid, and dried for 24 h at 80 °C. The dried material was added to a diluted HNO_3 solution (0.001 M), and the mixture was left for 3 h at room temperature and then was oven-dried at 60 °C. At the final step, Fe_3O_4 nanoparticles (Fe-NPs) were introduced to a suspension of the above dried material and were mixed for 1 h. The final suspension was oven-dried at 80 °C. This material is referred to as TT500-Fe.

A chemically activated form of TT550 has been also reported (TT550-s). TT550 was transferred into a sulfuric acid solution of 0.1 N concentration for 12 h. Afterwards the suspension was filtered and

washed with deionized water. The received activated carbon was dried at 105 °C for 12 h in an electrical oven.

TT700 was decorated with $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ nanoparticles followed a co-precipitation procedure of the nitrate salts of Ni, Zn, and Fe with the simultaneous presence of TT700. The obtained composite is referred to as TT700-nzf. The authors of this work presented SEM images of TT700-nzf (Fig. 4). It can be observed that the surface of the carbonaceous phase is homogeneously covered with nanoparticles of size around 25 nm. The surface area of TT700-nzf showed a comprehensive value compared to other carbons obtained by natural wastes of 814 m²/g, while the total pore volume 0.726 cm³/g.

4.3. Characterizations

The characterization of produced materials would be interesting, but unfortunately, the reported studies have been focused almost exclusively on their pollutant removal capabilities, rather than to extensive characterizations. FTIR, XRD, and SEM analysis were presented in a limited number of reports in order to show the differences before and after the adsorption. The study of the surface chemistry of the mildly dried samples is mostly important. Specifically, the FTIR analysis of AD-Moo showed the presence of numerous surface functional groups. The most intense bands and their assignment to the type of the vibration modes are collected in Table 3. These vibrations correspond to the presence of the following groups: 1) H-bonded phenolic, 2) acetyl, 3) carboxyl, and 4) amines. The existence of polysaccharide sugars, such as galactose and glucans was also reported.

The FTIR analysis of the thermally treated sample TT400 (400 °C, with the presence of sulfuric acid) showed four intense bands at 3400, 2900, 1555, 1180 cm⁻¹, attributed to sulfur containing surface moieties. TT700 (prepared by carbonization at 700 °C) of the washed and dried raw shells showed the presence of nitrogen containing groups. The three intense bands at 3452, 1555, and 981 cm⁻¹ were attributed

Table 3

The bands and their assignment for AD-Moo (Moosa et al., 2016a, b).

Frequency (cm ⁻¹)	Vibration mode
3000–3600	O–H (s)/N–H (s)
2924	C–H (s)/N–H (s)
2356	C–N (s)/O=C=O (s)
1732	C=O (s)
1643	O–C–O (s)/C=C (s)/N–h (b)
1431	C–H (b)
1269	C–N (s)/C–O–C (s)
1076	C–O (s)/C–N (s)

(s): stretching vibrations, (b): bending vibrations.

Table 4List of the best isotherms and kinetic models for describing the adsorption of heavy metals on *Aloe vera* waste-based biosorbents.

Adsorbent ^a	Heavy metals	Optimum pH/ time	Isotherm model ^b	Kinetic model ^c	Maximum monolayer adsorption capacity (mg/g)	Reference
Ni _{0.5} Zn _{0.5} Fe ₂ O ₄ magnetic nanoparticles supported by <i>Aloe vera</i> shell carbon (TT700-nzf)	Ag ¹⁺	5/30min	L,F	–	243.90	(Beigzadeh and Moeinpour, 2016)
Ni _{0.5} Zn _{0.5} Fe ₂ O ₄ magnetic nanoparticles supported by <i>Aloe vera</i> shell carbon (TT700-nzf)	Pb ²⁺	5/15 min	F	–	47.2	(Namavari and Moeinpour, 2016)
<i>Aloe vera</i> leaves ash (TT500)	Pb ²⁺	6/7 min	L	Ps2	500.00	(Abedi et al., 2016)
Magnetically modified <i>Aloe vera</i> leaves ash (TT500-Fe)	Pb ²⁺	6/10 min	L	Ps2	333.30	(Abedi et al., 2016)
<i>Aloe barbadensis</i> leaves (AD-Mal)	Pb ²⁺	4/30 min	L,F	Ps1	86.4 ^d	(Malik et al., 2015b)
Modified with H ₃ PO ₄ <i>Aloe vera</i> leaves (AD-Mal-p)	Pb ²⁺	4.5/25 min	L	Ps1	96.2 ^d	(Malik et al., 2015b)
<i>Aloe vera</i> leaves (AD-Moo)	Pb ²⁺	5/360 min	F	–	4.68	(Moosa et al., 2016a)
Acid activated <i>Aloe vera</i> leaves (AD-Moo-sn)	Pb ²⁺	5/360 min	F	–	6.21	(Moosa et al., 2016a)
<i>Aloe Vera</i> leaves (AD-Sin)	Cu ²⁺	5/120 min	F	Ps2	2.27	(Singh et al., 2017)
<i>Aloe vera</i> leaves ash (TT500)	Cu ²⁺	6/7 min	L	Ps2	117.60	(Abedi et al., 2016)
Magnetically modified <i>Aloe vera</i> leaves ash (TT500-Fe)	Cu ²⁺	6/10 min	L	Ps2	345.00	(Abedi et al., 2016)
<i>Aloe vera</i> leaves ash (TT500)	Cr ³⁺	6/7 min	L	Ps2	142.80	(Abedi et al., 2016)
Magnetically modified <i>Aloe vera</i> leaves ash (TT500-Fe)	Cr ³⁺	6/10 min	L	Ps2	333.30	(Abedi et al., 2016)
<i>Aloe vera</i> leaves ash (TT500)	Zn ²⁺	6/7 min	L	Ps2	200.00	(Abedi et al., 2016)
Magnetically modified <i>Aloe vera</i> leaves ash (TT500-Fe)	Zn ²⁺	6/10 min	L	Ps2	71.40	(Abedi et al., 2016)
<i>Aloe vera</i> leaf powder (AD-Moo)	Zn ²⁺	5/360 min	F	–	5.08	(Moosa et al., 2016b)
Acid activated <i>Aloe vera</i> powder (AD-Moo-sn)	Zn ²⁺	5/360 min	F	–	5.41	(Moosa et al., 2016b)

^a The abbreviations of the materials used in this work are given inside the parentheses.

^b L: Langmuir; F: Freundlich.

^c Ps1: Pseudo-first-order kinetic model; Ps2: Pseudo-second-order kinetic model.

^d Initial Pb²⁺ concentration = 0.3 g/L, contact time = 30 min, adsorbent dose 0.3 g/50 mL.

to the stretching vibrations of N–H, N–O, and C–N bonds, respectively (Beigzadeh and Moeinpour, 2016).

5. *Aloe vera* waste-adsorbents for aquatic pollutants removal

In this part, all the reported adsorption results regarding the water treatment by *Aloe vera*-derived materials are presented. For the sake of discussion, the results were categorized based on the type of pollutant. Three basic categories, such as metals (single metal solution, multi metals solution), dyes, and miscellaneous (fluoride, aniline and 4-chloro-phenols) are discussed in details. In Tables 4–6 are tabulated isotherms, kinetic models, maximum adsorption capacities, as well as the optimum pH and contact time conditions. Unfortunately, the study

of isotherm models and the kinetics were very limited in almost all the herein presented articles. The fitting of Langmuir or Freundlich isotherms were presented in an epidemically extent in order to present which model fits better. Similarly, the kinetic studies were limited on the appliance of the pseudo-first and second kinetic models.

5.1. Single metal removal

The removal capabilities of the *Aloe vera* derived materials have been focused on the following metals: Ag⁺, Pb²⁺, Cu²⁺, Cr³⁺, and Zn²⁺, using batch experiments.

The adsorption of Ag⁺ by Ni_{0.5}Zn_{0.5}Fe₂O₄ magnetic nanoparticles supported by *Aloe vera* shell carbon (TT700-nzf) was examined by

Table 5List of the best isotherms and kinetic models for describing the adsorption of dyes on *Aloe vera* waste-based biosorbents.

Adsorbent ^a	Dyes	Optimum pH/ time	Isotherm model ^b	Kinetic model ^c	Maximum monolayer adsorption capacity (mg/g)	Reference
Activated carbon from <i>Aloe vera</i> waste (TT550-s)	Methylene blue	12/40 min	F	Ps2	129.87	(Khaniabadi et al., 2016a)
Activated carbon from <i>Aloe vera</i> leaves shell (TT550)	Congo red	2/20 min	F	Ps2	1850	(Khaniabadi et al., 2017)
Activated carbon from <i>Aloe vera</i> waste (TT400)	Congo red	2/50 min	L,F	–	35.71	(Arivoli et al., 2008)
Activated carbon from <i>Aloe vera</i> waste (TT400)	Malachite green	10/50 min	L,F	–	34.58	(Arivoli et al., 2008)
Activated carbon from <i>Aloe vera</i> waste (TT400)	Rhodamine B	10/50 min	L,F	–	10.31	(Arivoli et al., 2008)
Activated carbon from <i>Aloe vera</i> waste (TT400)	Rosel bengal	2/50 min	L,F	–	7.69	(Arivoli et al., 2008)
<i>Aloe vera</i> leaves ash (TT300)	Reactive red 198	3/20 min	F	–	47.27 ^d	(Malakootian et al., 2014)
<i>Aloe vera</i> leaves ash (TT300)	Reactive blue 19	3/20 min	F	–	54.32 ^d	(Malakootian et al., 2014)
Activated carbon prepared from the <i>Aloe Vera</i> leaves and modified with sulfuric acid (TT550-s)	Methyl orange	3/60 min	F	Ps2	196.07	(Khaniabadi et al., 2016b)

^a The abbreviations of the materials used in this work are given inside the parentheses.

^b L: Langmuir; F: Freundlich.

^c Ps1: Pseudo-first-order kinetic model; Ps2: Pseudo-second-order kinetic model.

^d Obtained from batch study (initial dye concentration 60 mg/L, pH = 3, contact time = 20 min, adsorbent dose 0.4 g/L).

Table 6List of the best isotherms and kinetic models for describing the adsorption of miscellaneous pollutants on *Aloe vera* waste-based biosorbents.

Adsorbent	Pollutant	Optimum pH/ time	Isotherm model	Kinetic model	Maximum monolayer adsorption capacity (mg/g)	Reference
Activated carbon from <i>Aloe vera</i> waste (TT550)	4-chlorophenol	2/40 min	F	Ps2	47.6	(Khaniabadi et al., 2015)
<i>Aloe vera</i> waste	Fluoride	7/60 min	L, F	Ps1	40	(Rayappan et al., 2014)
The gel of <i>Aloe vera</i> leaves and calcium chloride (NT-J)	Fluoride	2.45/40 min	L, F	–	0.12	(Shyam and Kalwania, 2014)
<i>Aloe vera</i>	Fluoride	7/ < 60 min	L	Ps1	4.41	(Murugan and Subramanian, 2002)
<i>Aloe Vera</i> leaves wastes-based activated carbon (TT550)	Aniline	3/60 min	F	Ps2	106.38	(Basiri et al., 2015)
Activated carbon prepared from the <i>Aloe Vera</i> leaves and modified with sulfuric acid (TT550-s)	Aniline	3/60 min	F	Ps2	185.18	(Khaniabadi et al., 2016b)

Beigzadeh and Moeinpour (2016). Optimum adsorption conditions were obtained at pH 5, 30 min of contact time and 0.20 g/50 mL solid/liquid ratio. The activation energy (E), evaluated from the Dubinin-Radushkevich (D-R) model, was less than 8 kJ/mol, indicating a physisorption mechanism. Repeated adsorption/desorption experiments were conducted using 0.1 mol/L Na₄-EDTA proving that the adsorbent didn't lose its adsorption capacity.

Namavari and Moeinpour also examined the use of Ni_{0.5}Zn_{0.5}Fe₂O₄ magnetic nanoparticles supported by *Aloe vera* shell carbon (TT700-nzf) for the uptake of Pb²⁺ (Namavari and Moeinpour, 2016). The pH was found to control the removal process and maximum adsorption was found at pH 5. The effect of adsorbent dose was studied in the range from 0.01 g/50 mL to 0.4 g/50 mL solid/liquid ratio and the best removal was about 99.8% using 0.2 g/50 mL solid/liquid ratio. The mean free energy (E) was 2.673 kJ/mol which confirmed the physical nature of adsorption process.

The use of *Aloe vera* leaf powder in raw (AD-Mal) (Malik et al., 2015b) and its modified with 1 M H₃PO₄ form (AD-Mal-p) (Malik et al., 2015b) were examined for the uptake of Pb²⁺. For both adsorbents, the thermodynamic study was explored in a temperature range of 273–333 K and the adsorption was found to be spontaneous, feasible and exothermic in nature. For raw material, optimum adsorption conditions were observed at pH 4 (pH studied range from 1 to 7), temperature of 30 °C (temperature studied range from 0 to 100 °C) and 30 min of contact time (contact time studied range from 5 to 160 min) whereas for the modified adsorbent were at pH 4.5, 40 °C and 25 min for the same ranges of adsorption parameters, respectively.

Aloe vera leaves in raw (AD-Moo) and acid (different mixtures of HNO₃ and H₂SO₄) activated *Aloe vera* leaves (AD-Moo-sn) were used to adsorb Pb²⁺ (Moosa et al., 2016a) and Zn²⁺ (Moosa et al., 2016b) from artificially contaminated water. Acid activation was found to increase the BET surface area. More specifically, the BET surface area for the raw material was found to be 13.8 m²/g, whereas for the acid modified materials was 22.7, 21.9 and 21.9 m²/g for 1: 1, 1: 2 and 2: 1 HNO₃: H₂SO₄ acid mixtures. The highest adsorption occurred using 2.2 g and 1.6 g for raw and acid treated adsorbent, respectively. For both ions and adsorbents, optimum adsorption conditions were noticed at pH 5 (in the pH range from 2 to 7) and after 6 h of contact time (in the contact time range from 2 to 10 h).

Singh et al., examined the uptake of Cu²⁺ by powder derived from *Aloe vera* leaves after air drying (AD-Sin) from aqueous solution (Singh et al., 2017). The highest removal percentage were observed in the following adsorption conditions: 2 h of contact time (tested range from 0.5 to 3 h), 20 mg/L of initial Cu²⁺ concentration (tested range from 20 to 70 mg/L), adsorbent dose 2 g (tested range from 0.5 to 3 g), pH 5 (tested range from 4 to 9) and temperature 45 °C (temperature studied at 25 °C, 35 °C and 45 °C). The adsorption of Cu²⁺ ions was feasible, spontaneous and endothermic in nature. The negative ΔS^0 value denoted decreased randomness at the solid/liquid interface during the adsorption process.

Aloe vera leaf after only milling (NT-P) was also used to adsorb Cr⁶⁺ by Pragathiswaran et al. (2013). Maximum adsorption (60%) was obtained at pH 2 (pH studied range from 2 to 11) and the equilibrium was attained after 180 min of contact time (in the range from 30 to 180 min). The raise of initial concentration from 20 to 80 mg/L caused a reduction of removal uptake and optimum removal % was observed at 20 mg/L.

Tripathi et al. also applied *Aloe vera* leaves to remove arsenic from water. They concluded that 30 g of biosorbent was sufficient to clean 1 L of polluted water including arsenic up to 43 ppb, in a period of 4 h (Tripathi et al., 2016).

5.1.1. Multi metal removal

Batch equilibrium studies were carried out with the scope to test the uptake of Pb²⁺, Cu²⁺, Zn²⁺, and Cr³⁺ from magnetically modified *Aloe vera* leaves ash (TT500) (Abedi et al., 2016). The increase of pH from 2 to 7 was found to positively affect the removal process and maximum adsorption was obtained at pH 6–7. The effect of contact time (contact time ranged from 1 to 15 min) revealed that the adsorption equilibrium was reached in 10 min and remained constant up to 15 min for all studied metals. The negative values of the Gibbs free energy change (ΔG^0) showed that the adsorption was feasible and spontaneous, whereas the negative values of enthalpy change (ΔH^0) suggested the exothermic nature of uptake process. Desorption studies were conducted satisfactorily by using 0.001 M HCl and after five adsorption/desorption cycles the removal percentages of Pb²⁺, Cu²⁺, Cr³⁺ and Zn²⁺ were 71.64, 88.15, 92.08 and 86.67%, respectively.

5.2. *Aloe vera* waste-adsorbents for other pollutants removal

5.2.1. Dyes removal

Khaniabadi et al., examined the removal of methylene blue dye by *Aloe vera* waste activated carbon (TT550) from aqueous media (Khaniabadi et al., 2016a). The pHPZC was estimated to be 11.3 and the best adsorption capacity appeared at pH 12 (adsorption capacity 22.98 mg/g). The increase of adsorbent dose from 1 to 10 g/L led to a decrease in adsorption capacity so that the best adsorption was found using 1 g/L. Adsorption was fast and equilibrium was achieved in 40 min. The q_{max} obtained from the Langmuir isotherm reported to be 129.87 mg/g. This value is an acceptable one, especially by taking into consideration the low temperature of activation. However, carbons obtained by various other sources of agricultural wastes (like cashew nut shells, tea seeds, corncob, pecan nut shell, etc.) have exhibited higher removal performances (Spagnoli et al., 2017).

Khaniabadi et al. tested the use of activated carbon (TT550) prepared from *Aloe vera* leaves shell with the aim to sequester congo red (Khaniabadi et al., 2017). SEM images of the activated carbon showed that the adsorbent had irregular cavities via fine open holes. After the adsorption, a smoother configuration, regular surface, and expanded pores were formed. The expansion of the pores was ascribed to the

uptake of dye onto the sorbent surface that filled the pores. Optimum adsorption conditions were obtained to be contact time of 20 min, pH 2.0, and adsorbent dose of 0.5 g/L. Both the raise of pH (from 2 to 12) and adsorbent dose (from 0.5 to 2 g/L) negatively affected the dye uptake.

The adsorption of congo red, malachite green, rhodamine B and rose bengal by acid activated carbon derived from *Aloe barbadensis* (TT400) was studied by Arivoli et al. (2008). In their work, batch equilibrium experiments were conducted studying the effect of initial dye concentration, pH, temperature, and contact time. The point of zero charge was estimated to be 6.7. The pH seemed to play a significant role in adsorption process. Maximum adsorption was found at pH 3 for acid (anionic) dyes (congo red, rose bengal) and at 10 for basic (cationic) dyes (malachite green, rhodamine B). Both surface adsorption and intra-particle diffusion participated in the dye removal process. Desorption studies were carried out using different desorption eluents (such as sulfuric acid, hydrochloric acid, nitric acid and water) and it was noticed that 0.2 M HCl gave the best desorption % with more than 65% of adsorbed dyes.

Aloe vera plant ash was explored to adsorb reactive red 198 and blue 19 from water (Malakootian et al., 2014). The effects of pH (3–12), adsorbent dose (0.1–1 g/L), contact time (10–60 min), initial dye concentration (10–160 mg/L) and temperature (25–55 °C) were discussed in detail. For both dyes, the highest removal capacity (~50 mg/g) was obtained at pH 3, 0.4 g/L of adsorbent dose, 20 min of contact time, 60 mg/L of initial concentration and 55 °C of temperature.

5.2.2. Fluoride removal

The removal of fluoride was studied by Shyam and Kalwania (2014) using a mixture of the gel part of the *Aloe vera* leaves and CaCl₂ (NT-J). The optimum removal was estimated to be 88% at pH 7.4, using 40 g of *Aloe vera* derived juice, 3 g of CaCl₂ in 1000 mL sample and 40 min of contact time. The removal of fluoride ions by *Aloe barbadensis* as adsorptive material (Rayappan et al., 2014) was also investigated. The best uptake efficiency (40 mg/g) was noticed at pH 7 (studied range from 3 to 11.5), adsorbent dose 10 mg/50 mL (studied range from 10 to 200 mg/50 mL), 60 min of contact time (studied range from 10 to 60 min) and 120 rpm of shaking speed (studied range from 100 to 300 rpm). The presence of carbonate, sulphate and chloride ions showed a minimal negative effect on fluoride removal, whereas the presence of phosphate ions led to a negative impact. On the contrary, the co-existence of nitrate ions affected positively the fluoride adsorption.

Aloe vera plant material was also used by Murugan and Subramanian, to adsorb fluoride (Murugan and Subramanian, 2002). The pH at the point of zero charge of the adsorbent was found to be 7.43 and maximum fluoride adsorption was noticed at pH around 7. The equilibrium of adsorption was attained in less than 60 min and the effect of temperature affected negatively the adsorption capacity. More specifically, the rise in temperature from 20 to 50 °C resulted to a decrease in removal ability from 85.9% to 52.63%. One possible explanation is that at high temperatures a) deterioration and surface deactivation of the adsorbent biomass and b) partial destruction of active sites due to bond rupture may have occurred (Anastopoulos et al., 2018). The negative value of ΔH^0 and negative value of ΔG^0 indicated that the removal process was endothermic and spontaneous in nature, respectively. The negative ΔS^0 value for adsorbent indicated decreased randomness at the solid/liquid interface.

5.3. Adsorption of 4-chlorophenol and aniline

Khaniabadi et al. examined the use of activated carbon fabricated from *Aloe vera* waste (TT550) in order to sequester 4-chlorophenol from aqueous solution (Khaniabadi et al., 2015). Maximum adsorption was achieved at pH 2 (in the studied pH range from 2 to 8) and equilibrium was accomplished in 40 min (in the studied contact time range

from 0 to 100 min). The increase of adsorbent dose from 1 g/L to 9 g/L led to a decrement on the adsorption capacity, so that the optimum dose was 1 g/L.

Aloe vera leaves wastes-based activated carbon (TT550) (Basiri et al., 2015) and sulfuric acid modified activated carbon-based *Aloe vera* leaves (TT550-s) (Khaniabadi et al., 2016b) were fabricated in order to test their ability to remove aniline. SEM images revealed that the use of sulfuric acid as modification agent resulted to a material with highly regular pores which indicates a smoother surface and higher surface area. The pH_{ZPC} of TT550 and TT550-s were estimated to be 11.3 and 5.8, respectively. SEM-EDX analytical results of TT550 and TT550-s showed that the modification of activated carbon with sulfuric acid led to significant decrease in weight percentage of oxygen (from 43.82% to 28.30%) and sulfur (24.07%–17.9%), and an increase in weight percentage of calcium (27.98%–29.82%) and manganese (3.66%–13.18%). The effects of pH, adsorbent's dose and contact time on the adsorption process were studied at pH values between 3 and 11, using 1–5 g/L adsorbent: solution ratio, and 0–90 min as contact time. The optimum conditions were at pH 3.0, 1 g/L of adsorbent dose, and the equilibrium was reached at the contact time of 60 min. The use of sulfuric acid was found to improve the adsorptive ability of TT550 (q_{max} obtained from Langmuir for TT550: 106.38 mg/g and for TT550-s: 185.18 mg/g).

6. Conclusions

This review article summarizes in brief the history, taxonomy, botany, and applications of *Aloe vera*, and predominately focuses on the alternative use of *Aloe vera* waste as a basis to fabricate novel and efficient adsorptive materials. This study has collected all the reports in which *Aloe vera* waste-biomass has been used as precursor for the synthesis of biosorbents. These reported in the literature biosorbents were synthesized following various strategies. The herein studied materials were categorized in five groups, based on the applied treatment. These groups include:

1. Non-treated materials,
2. Air dried biosorbents,
3. Chemically treated dried powder,
4. Thermally treated *Aloe Vera* biomass,
5. Carbonized and functionalized materials.

The biosorption performance was found to be governed by different parameters such as initial pollutant concentration, initial solution pH, contact time, adsorbent dosage, temperature, etc. Regarding the examined pollutants, heavy metals and dyes were predominantly explored. Optimum pH and contact time ranges were identified between 4 and 6, and 7–360 min for the studied metals, whereas for dyes (depends on the type of dye: cationic/basic or anionic/acidic) were between 2 and 12, and 20–60 min, respectively. Langmuir isotherm and pseudo-second kinetic model were found to give the best fit to the experimental data.

7. Challenges and future prospects

As a new and not so widely explored topic, in order the application of *Aloe vera* waste biomass-based adsorbents to be competitive with other commercial adsorbents, and for being economically feasible and environmentally sustainable, future work should give emphasis to the following points:

7.1. Laboratory experiments

In order to understand the holistic performance of these fabricated materials in wastewater treatment process, their applicability on a variety of other pollutants (such as endocrine disruptors, pharmaceutical compounds, radionuclides, and rare earth metals), column studies,

and reusability studies should be performed in every study.

A more advantageous and in-depth discussion of the equilibrium data, adsorption mechanism/isotherm/kinetic modelling should be followed, in addition to appliance of the Langmuir and Freundlich isotherm models or pseudo-first and pseudo-second order models. For these purpose, other models like intra particle diffusion model (Weber-Morris model) or the application of three-parameter isotherm equations is highly recommended.

Other issues that should be addressed is the comprehensive characterization of the adsorbents, the alteration of their physicochemical properties after multiple adsorption/desorption cycles, or even in cases that the raw materials were used only after minimal/mild treatment, the microbial degradation.

7.2. Real-life application evaluation

The evaluation of the biosorbents removal capabilities against real wastewater and in pilot scale are also of a high priority.

The cost arises from the collection up to the final step of the fabrication of the *Aloe vera* waste biomass-based adsorbents should be estimated in order to answer if the application is economically feasible. The use of the chemical solutions to recover the pollutants from surface and application cost should also be estimated.

Future work should focus on the engrave strategy regarding disposal of the polluted loaded wastes and on the handle of the chemical solution wastes obtained from adsorption/desorption studies. Life cycle analysis of the fabrication of *Aloe vera* waste biomass-based adsorbents is mandatory.

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