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Review

에너지 저장 및 환경 분야에 응용되는 바이오매스 기반 활성탄

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Biomass-based Carbon Materials for Energy Storage and Environmental Applications

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초 록

에너지 저장 및 환경 관련 분야에 응용 흡착매질로 바이오매스 기반 활성탄의 중요성을 살펴보았다. 지금까지 발표된 연구 결과는 바이오매스 기반 활성탄의 표면적과 기공부피 이외에 이들의 표면 화학 특성 또한 다양한 분야에 응용될 수 있는 중요한 역할이 있음이 확인된다. 바이오매스 기반 활성탄의 용량은 바이오매스의 특성 및 이들의 활성화 공정에 따라 달라지므로 다양한 응용 분야에 맞게 제조할 수 있다. 따라서 본 리뷰에서는 다양한 분야에 이용되고 있는 바이오매스 기반 활성탄의 역할을 정리하였다.

Abstract

The importance of the biomass-based activated carbon as an adsorbent has been reviewed with emphasizing on the application in the fields of energy storage and environmental related problems. It is clear from the literature survey that beside surface area and pore volume, surface chemistry also plays important role in determining their usage in various field. The capacities of biomass-based activated carbon can be increased depending upon the choice of the biomass used and the pathway taken for their activation and hence they can be tailored for various applications. Accordingly, this review summarizes the role of biomass based activated carbon in different applications.

***Keywords:** biomass, activated carbon, gas storage, EDLC, VOC removal*

1. Introduction

Activated carbons (ACs) are classic carbons, which have high surface area, large pore volume and a wide range of pore size from the Angstrom scale of the micropores to the micrometer scale of macropores. According to the International Union of Pure and Applied Chemistry (IUPAC) recommendations, “Activated carbon is a porous carbon material, a char which has been subjected to reaction with gases, sometimes with the addition of chemicals, e.g. $ZnCl_2$, before, during or after carbonization in order to increase its adsorptive properties”[1].

The high adsorption capacities of ACs are mainly related with their structural characteristics such as surface area[2], pore volume, pore size and pore size distribution[3]. It is highly recommended that the controlled pore size and pore size distribution are necessary for the utilization of ACs for specific usages[4]. In general, the highly microporous AC are used for the gas adsorption, whereas the mesoporous AC are utilized for the adsorption of large molecules[5]. It has been reported that the physical properties of ACs are mostly determined by the nature of the precursors as well as the adopted preparation processes[3,4,6].

In general, physical and chemical activation methods can be used for the preparation of ACs[4,7-13]. During the physical activation, the process starts with the carbonization of raw material and followed by the activation of the resulting char or direct activation of the precursor in the presence of an activating agent such as steam, carbon dioxide, air or their mixtures at higher temperature[2,4,7]. In the case of chemical activation processes, the activating agent is selected based on the nature of the precursors. For example, the activating agents such as

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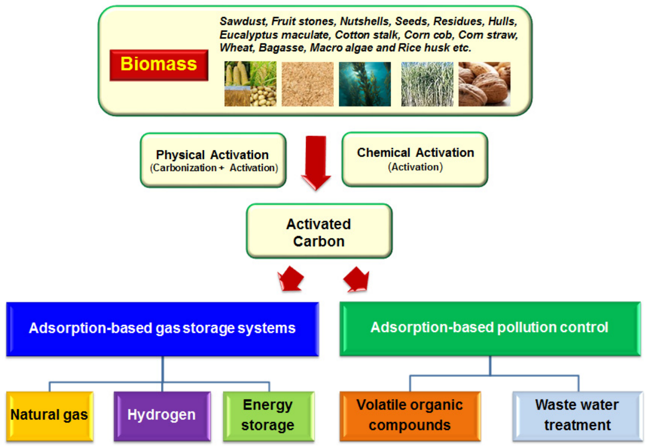

Figure 1. Pathways of producing biomass-based activated carbon and their application.

phosphoric acid and zinc chloride are used for the activation of non-carbonized lignocellulosic materials[14], whereas alkaline metal compounds, more importantly KOH, are employed for the activation of coals and carbonized chars[4,12]. During the chemical activation, chemical activating agents, which are basically dehydrating agents, prevents the formation of tar and hence the yield of carbon produced is usually high compared to that of the physical activation process. The physical activation is generally done through the interaction between the carbon atom and oxidizing gas and here typically a huge amount of internal carbon is removed from the structure. Each activation process has its own advantages and disadvantages. The physical activation processes are eco-friendly and corrosion free processes, although they need higher activation temperature and time compared to the chemical activation. On the other hand, the chemical activation processes require low activation temperature and activation time. Moreover, it usually helps to get high yield, surface area, microporosity along with higher reduction in higher mineral content. However, the chemical activation processes have some disadvantages such as the more complex reaction mechanism, the corrosiveness of the process and the requirement of washing stage for the removal of activating agent[4,13].

Numerous precursors have been used for the preparation of ACs. The precursors are usually selected based on the carbon and inorganic contents, availability, cost, bulk density, mechanical strength, degradation upon storage, ease of activation procedure and the specific application of the resulting AC[15]. Mostly, carbon-containing organic materials are being used as the precursors. The available precursors are natural biomasses, coals, resins, synthetic polymers, petroleum coke, phenolic resins, peat and *etc*.

A proper characterization of surface properties of ACs plays a key role in their successful synthesis and application[16]. The gas adsorption (N_2, Ar or CO_2) based characterization technique appears to be the most suitable compared to that of liquid adsorption studies (Immersion calorimetry). Among the all characterization techniques, N_2 adsorption measurement is most common, simple and convenient[17]. The obtained data from the adsorption experiment are analyzed with several methods such as the Brunauer-Emmett-Teller (surface area), Barrett-Joyner-Halenda (mesopore size and mesopore size distribution), Dubinin-Radushkevich (micropore size and micropore size distribution), *t*-plot (micro pore volume), α s-plot (external surface area, micropore volume) density functional theory (pore size distribution) to obtain the detailed information about the concern AC[18].

ACs have been successfully employed in a number of adsorption studies, as shown in Figure 1, according to their surface characteristics. Recently, there has been a growing interest in using activated carbon as an adsorbent for the design of an efficient gas storage system for vehicle, in particular, for the storage of natural gas[19,20] and hydrogen[21,22]. The activated carbon with very high surface area, large pore volume and narrow pore size distribution with controlled pore size is considered to be an efficient storage media. Beside, activated carbons have been shown most attractive electrode materials for the electrical double layer capacitors, as a consequence of their apt textural characteristics[23,24]. ACs are also effectively reduced the emission of various volatile organic compounds[25,26]. Apart from these applications, activated carbons are widely used for the wastewater treatment owing to their unique surface characteristics[27,28].

Activated carbons (ACs) have been extensively used in various processes including gas separation, gas storage, solvent recovery, energy storage, waste water treatment, air pollution control and catalyst support owing to their high adsorption capacities, relatively high mechanical strength, low cost and easy availability[3,4,6-8,29-31]. The biomass is relatively cheap and widely available precursor in all over the world. Several natural biomasses such as wood, sawdust, fruit stones, nutshells, seeds, residues, hulls, eucalyptus maculate, cotton stalk, corn cob, corn straw, wheat, bagasse and rice husk have been used for the preparation of ACs[32]. It should be noted that the huge quantities of agricultural by-products result from the annual harvesting and processing of various agricultural crops grown around the world. Also, the agricultural by-products based ACs show the comparable adsorption capacities compare to that of the commercial ACs[33]. This article mainly aims at making concise review report on applicability of biomass based ACs for energy and environmental applications. The same has been done by conducting a detailed literature review based on the research papers published in this field.

2. Adsorption-based Gas Storage Systems

2.1. Natural gas

Environmentally and economically satisfied natural gas is one of the most wanted fuels now in energy field compare to the liquid fuels such as gasoline and diesel[34]. Natural gas (NG) is an abundant and the cleanest burning alternative transportation fuel today. Natural gas vehicles (NGVs) have surpassed all other competitors in delivering the superior emissions performance. Moreover, NGVs have been certified to the most demanding environmental emission standards. Although it is most convenient than liquid fuel, due to its low energy density at standard temperature and pressure (STP) condition, storing huge amount of methane in a given volume is very difficult[19,20]. Thus, it has become the perfect challenge for active researchers[35]. Also, the developments of NG storage and transportation technologies are very important for the future. Well known conventional methods (e.g. liquefied natural gas (LNG) and compressed natural gas (CNG)) may be the solution for storage purpose even though they have some problems in both safety and cost wise[19,20,36]. For example, storing high density NG in LNG vehicles is possible at cryogenic temperature. However, the specialized container design and refueling procedure required are undesirable for vehicular fuel application. In CNG, multi stage compression increases the cost of the process and furthermore relatively huge tank is required[37]. To overcome the problems inherent in the conventional methods, a newly upcoming adsorbed natural gas (ANG) storage system has been used for the NG storage[19,20,35-39]. ANG is a method in which natural gas is stored by physisorption at 298 K and up to 35 atm[19,40-42]. Recently the Advanced Research Projects Agency-Energy (ARPA-E) of US DOE has set the target of 263 V/V[43,44], equivalent to the gravimetric value of 0.5 g of CH_4/ g of sorbent, for the ANG storage system so that ANG can have the amount of energy density, which is similar to CNG. This target could be achieved only by selecting an appropriate adsorbent. The highly microporous AC is a promising adsorbent, which satisfies most of the requirements for the better design of an ANG vessel[19,20,37,38,45-53]. However, it is also noted that some ACs show low adsorption capacity on a volume basis (volume of methane adsorbed/ volume of adsorbent) owing to their low packing density, even though their capacities are high on a mass basis (moles of methane adsorbed/ grams of adsorbents)[38,47]. Therefore, the monolithic forms of ACs have been employed in the ANG storage system to achieve the universal target[38,47,48,50]. The monolith form offers the increase in density by reducing the excess void volume[39], compactness and easy handling. In general, the carbon monolith has been prepared by compressing the AC[50] or a mixture of AC and binder[38,39,47,48].

Biomass based activated carbons synthesized by several researchers have shown high surface area and high volumetric storage capacity[11,34]. Bagheri and Abedi synthesized corn cob based ACs having surface area 1320 m^2/g with the optimum methane storage capacity of 160 V/V at 298 K and 103 bar[49]. Similarly, Rios et.al. prepared ACs from coconut shell having specific surface area of 1441 m^2/g and natural gas storage capacity of 95 V/V at 303 K and 35 bar[50]. The biomass based Activated carbon synthesized by Zhang et.al shows the specific surface area of 2000 m^2/g and the methane storage capacity of 0.7 g/cm^3 at 298 K and 34.5 bar[51].

2.2. Hydrogen

Hydrogen (H_2) fuel is one of the proper options to meet the energy demand of our world. Moreover, H_2 is considered as an eco-free and efficient fuel[54,55]. In spite of several advantages, the development of a viable storage system for H_2 is still challengeable task for the researchers due to its low energy density[56]. The US Department of Energy (DOE) has also fixed the strict materials based storage targets for H_2 storage systems for vehicular applications as follows 5.5 wt% by 2020 and 7.5 wt%, ultimate storage target [US DOE]. Hence, in the last two decades there has a growing interest in the design of new H_2 storage systems with high capacity, light mass and high stability for mobile applications[21,54].

The methods such as liquefaction (20-30 K and 0.5-1 MPa), compression (35-70 MPa and room temperature), and formation of metal hydrates are currently available for the storage of H_2[57-59]. However, the mentioned storage techniques have different drawbacks as follows, potential H_2 losses due to evaporation in the liquefied hydrogen storage system, safety problems in the compressed storage system, and high weight and cost as well as the requirement of high temperature to release H_2 in the metal hydrates system[42,60]. On the other hand, it has been noted that the adsorption-based storage system offer the possibility to achieve the reversible storage of H_2 with higher volumetric and gravimetric densities at much lower pressures than compression and higher temperature than liquefaction[61-63].

Abundant adsorbents such as ACs, carbon fibers, carbon nanotubes, carbon nanohorns, fullerenes, zeolites, and metal-organic frameworks have been used as adsorbents for the adsorption of H_2[42]. Among the various adsorbents, the AC is also considered as a good candidate due to its high surface area, abundant pore volume, acceptable pore size, large microporosity and controlled pore size distribution, even though it has relatively low density[60,62-65]. Especially, highly microporous AC seems to be a good candidate for H_2 storage at 77 K also because of its unique surface characteristics, accessibility at industrial level and low cost[22,56,60,66].

Various researchers synthesized the biomass based ACs and showed their applications in hydrogen storage. Lignin based ACs prepared by carbonization and KOH activation showed the surface area of 1946 m^2/g and having hydrogen storage capacity of 1.89 wt%[67]. Sevilla et. al. prepared ACs derived from glucose, starch, cellulose, and eucalypt sawdust achieving maximum surface area of 2700 m^2/g. These materials have high hydrogen uptake which is around 6.4 wt% with large isosteric heat of adsorption (8.5 kJ/mol)[68]. Liou synthesized ACs from sugarcane baggase and sunflower seed hull by phosphoric and zinc chloride activation and achieved surface area up to 2289 m^2/g for sugarcane bagasse and 2240 m^2/g for sunflower hull[69]. Hydrogen storage capacity of zeolite like carbon material is found to be 6.9% by Yang et. al. at 77 K and 20 bar[70]. ACs synthesized by chemical activation of fungi-based chars with KOH has shown the optimal surface area of 2500 m^2/g and hydrogen uptake up to 4.7 wt% at 77 K and 35 bar[71]. Activated carbon prepared by chemical activation of cannabis sativa stem with KOH has shown surface area of 3241 m^2/g and hydrogen adsorption capacity of 3.28 wt% at 77 K at 1 bar[72] Activated carbon synthesized by Wang et.al. have hydrogen storage capacity of 7.08% at 77 K and 20 bar[73]. Further, Fiero et.al. indicated that ACs synthesized from high-rank mineral coal have hydrogen storage capacity of 6 wt% at 77 K and 40 bar[74]. Bader et.al. synthesized ACs from five different biomasses using KOH activation and have shown the hydrogen storage capacity of 6 wt% at 77 K and 200 bar[75]. These results show that the porosity, large micropores volume and well developed internal surface area play key role in the hydrogen storage[76-78]. KUA5 which is considered to be the most effective AC

Table 1. Biomass-based Activated Carbons for EDLC Applications

Carbon Source	Surface area (m^2/g)	Capacitance (F/g)	Reference
Banana peel	1650	206	96
Cherry stones	1300	230	97
Birch wood	1000	308	98
Firewood	~1100	120	99
Tobacco	3326	190	100
Paper pulp mill sludge	2980	166	101
Waste tea leaves	2841	330	102

with the specific surface area of 3000 m^2/g and can store up to 1.2 wt% hydrogen at 200 bar whereas KUA6 displayed hydrogen storage capacity of 56 wt% at 40 bar[60]. It was pointed out that number of narrow micropores (< 1 nm), narrow pore size distributions are very important in determining the hydrogen storage capacity[60,79].

3. Energy Storage

The energy demand in our world is steeply increasing due to the less resource of fossil fuels and growing population. It is also expecting that, the world energy demand will be doubled within-50 years. Along with this, the various environmental issues including global warming are caused by CO_2 and other toxic gas emission from vehicle and various industries, especially, from electricity producing industries. Hence, we are in a desperate position to find some other alternative fuel resources that would generate nil or less of toxic pollutants. The renewable fuel such as solar and windmill is promising energy and the production of energy from these sources is currently hot research area. The productions of energy in a large manner from these renewable fuels are currently in need of efficient storage system.

Electrochemical capacitors (ECs) have been used as energy storage devices because of fast energy delivery and long durability. ECs store the electric energy in an electrochemical double layer formed at a solid and electrolyte interface. Positive and negative ionic charges within the electrolyte accumulate at the surface of the solid electrode and compensate for the electronic charge at the electrode surface. In other words, based on the separation and accumulation of charge at the interface between the electronically conductive electrode and the ionically conductive electrolyte, the energy is stored in ECs[80-83]. EC is also known as supercapacitor, electrical double layer capacitor (EDLC)[84]. The physical mechanism of charge differentiates ECs from chemical energy storage systems (Batteries) and leads to their high power density, high cycle efficiency, long cycle life, and high efficiency. At that same time, the energy density of ECs is lower than batteries. Moreover, EDLCs are eco-friendly and low cost energy storage device than the conventional capacitors. The various applications of EDLCs are given as follows, backup sources (memories, microcomputers, system boards, batteries and clocks), main power sources (toys and started application), alternative power sources (solar watch, solar lanterns), energy storage (from windmills, solar cell), recover energy from various repetitive processes (braking in cars or descending elevators) and electric vehicles[81,84-88].

It has been known that the carbon materials are incorporated into the electrodes of energy storage device as electro-conductive additives, supports for active materials, electron transfer catalysts, intercalation hosts, substrates for current leads, and as agents for the control of heat transfer, porosity, surface area and capacitance. According these above mentioned reasons, carbons are also well suited as electrode materials for EDLCs[89]. Carbon nanotubes, carbon nanofibers, templated carbon graphene have been explored for their application supercapacitor electrode application, however still AC has its reputation to be used for commercial purpose because of their low cost and possibility to produce on the large scale. Moreover, ACs have better electrochemical properties compared to other. Biomass sources become extremely important because of the large availability and less cost.

The porous carbon based electrodes are widely used for the preparation of supercapacitor or EDLC application, because of their low cost and tunable micro texture and surface functionality, accessibility, a high electrical and thermal conductivity, processability and compatibility[23,90,91]. For the feasible operation of EDLC, carbon material with high surface area, tailored pore size[92], controlled pore size distribution, high micropore volume, high density, good corrosion resistance and controlled surface functionalities is necessary. It has been also recommended that the obtain the above mentioned properties of AC, the selection of the proper precursor as well as the appropriate carbonization and activation processes conditions are highly important[81,85-92].

ACs prepared from sugarcane bagasse have been tested by several researchers EDLC applications. The sugarcane bagasse based AC (surface area of 2871 m^2/g) synthesized by the Rufford et.al showed specific capacitance of 109 Fg^{-1}[93]. Hierarchically porous carbon materials (HPCMs) made by Huang and Doong have specific surface areas in the range 487-544 m^2 g^{-1} and displayed specific capacitance values in the range 190-234 F g^{-1} at the scan rate of 5-50 mV s^{-1}[94]. The ACs porous aerogel derived from the sugarcane bagasse has shown the specific capacitance of 142.1 F g^{-1} at a discharge current density of 0.5 A g^{-1}[95]. Apart from the biomass based ACs from sugarcane bagasse, several biomass sources have been consider for making ACs for EDLC application as shown in Table 1.

4. Adsorption-based Pollution Control Processes

Organic compounds which evaporate readily to the atmosphere are called volatile organic compounds (VOCs) as defined by the Environment Protection Agency (EPA). The main origins of VOCs emissions include the burning of coal and gasoline and also other sources such as solvents, paints, and cleaners[25,26]. Benzene, toluene and other secondary petrochemicals from various sources not only pollute our environment but also threaten human health[103-106]. Especially, the following significant effects such as climatic change, destruction of ozone stratospheric layer, acid rain, carcinogenic and mutagenic effects are observed due to the emission of VOCs into the environment[107]. The recovery of valuable VOCs is an economically and ecologically

Table 2. Adsorptive Dye Removal Using Biomass-based Carbon Adsorbents

Biomass	Compound to remove	Percent removal	Reference
Banana pith	Acid violet, Congo red + Rhodamine B mixture, Congo red + Acid Violet + Rhodamine-B mixture	80%	122
Orange peel	Congo red, Procion orange, Rhodamine-B	92%	123
Apple pomace	Cibacron Yellow C-2R, Cibacron Red C-2G, Cibacron Blue C-R, Remazol Black B, Remazol	81%	124
Wheat straw	Red RB	80%	124
Rosewood sawdust	Methylene blue	83%	125
Wheat shell	Direct blue 71	99%	126

Table 3. Biomass-based Carbonaceous Materials for Heavy Metals Removal

Adsorbents	Metal ion	Reference
Wheat straw	Cadmium, Copper, Chromium	127
Orange peel	Copper, Lead, Zinc, Cadmium	128
Litchi chinensis seeds	Nickel	129
Walnut shells	Lead, Cadmium, Nickel	130
Cashew Nut shell	Copper, Cadmium, Zinc, Nickel	131

feasible method rather than their destruction. The available methods for treating gaseous VOCs are absorption, adsorption, condensation, thermal oxidation and catalytic oxidation[107-110]. Among all the existed processes, a non-destructive adsorption process is one of the most appreciate methods for removing VOCs, because of easy operation, low operating cost and efficient recovery[25,105,111-114].

Compared to other adsorbents, AC is the most applicable adsorbent for the removal of VOCs because of its high surface area, hydrophobic properties, and high adsorption capacity[26,104-107,115,116]. In particular, the adsorption of VOCs is fairly good in micropores compared to mesopores because of the interaction between the attractive forces of the pore walls[116-118]. Hence, the preparation of highly microporous ACs is important for the removal of toxic gases.

The substantial amount of dyes and water are consumed in many industries such as dyestuffs, paper, textile and plastic in order to color their products. Consequently, they generate a considerable amount of colored wastewater[119] and mostly discharged in surface water resources[120]. The small amount of dyes in wastewater can also visibility noticed and they not only simply affects aesthetic merit but also inhibits the light penetration and photosynthesis[120-121]. Also, the most of the dyes give severe threat to our environment because of their mutagenic and carcinogenic behaviors[120]. Hence, it is environmentally important to discover cheap and effective methods for the wastewater treatment[119].

Of the above mentioned techniques, the adsorption has been recognized as a promising, cost-effective and efficient process for the removal of pollutants from aqueous solution[120]. Also it has been proven that the AC is an effective adsorbent for the removal of wide variety of pollutants dissolved in aqueous media due to its extensive surface area, high degree of porosity, effectiveness and versatility[119-121]. The activated carbon is used not only to adsorb various types of dyes but also different types of organic and inorganic pollutants as shown in the Tables 2 and 3. Many studies report that the waste water treatments including dye, heavy metal, and organic pollutant removal by ACs are economically favorable and technically more feasible[122-131].

5. Conclusions

The various possible applications of biomass-based carbon materials were studied in this review paper. Biomass based activated carbons have been recognized as potential adsorbents not only for gas/energy storage but also for removal of pollutants from the air as well as from the waste water due to their decent physical properties and relatively less cost of production. It was also observed in this review work that preparation of biomass carbon materials with high surface area, controlled pore size, larger pore volume and narrow pore size distribution is quite highly possible for the usage of numerous energy and environmental applications. Further, the review also finds from the published papers that the surface physical/chemical properties of biomass based activated carbon can be customized by carefully selecting conditions for carbonization and activation. Finally, it was clearly observed from the recently published papers that the carbon materials, especially prepared from various biomasses, are still being considered one of the potential adsorbents for real time storage, separation and purification applications.

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