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Spent tea leaves: A new non-conventional and low-cost biosorbent for ethylene removal

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ABSTRACT

This characterizations of the low-cost microsized spent tea leaf powder (MSTLP) and the effectiveness on its use as biosorbent in the removal of ethylene from a gaseous stream was investigated. Microprocessing treatment of used tea leaves significantly enhanced its ethylene removal capability. The effects of MSTLP dosage, initial ethylene concentration, and temperature on the adsorption of ethylene on MSTLP were evaluated. Moreover, the surface area/pore size distribution, functional group, and morphology of MSTLP samples were characterized. The kinetic adsorption data conformed well to a pseudo-second-order equation, and this adsorption was involved in intraparticle diffusion. The negative value of ΔG^0 and the positive value of ΔS^0 indicate that the adsorption of ethylene onto MSTLP was a spontaneous process. The adsorption capacity was found to increase with decreasing temperature and increasing MSTLP dosage. The maximum adsorption capacities varied from 0.68 to 6.93 $\mu\text{mol/g}$ at 25 °C based on the fits of Langmuir isotherm. MSTLP could be regenerated, and reused ten times for ethylene adsorption. The results show that a suitable choice of the microprocessing procedure for agriculture byproducts permits production of cheap biosorbent with high sorption capacity of ethylene.

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1. Introduction

Ethylene (C_2H_4) is a gaseous phytohormone, which can stimulate the ripening process of climacteric fruits and vegetables. Fruits and vegetables will gradually lose their freshness failing the control of the environmental concentration of ethylene. Moreover, ethylene can increase respiratory activity, activity of some enzymes, alteration of metabolism, seed germination, and growth before harvesting (Saltveit, 1999). The adverse effects of ethylene during postharvest include a reduced storage life of many fruits, vegetables, and decorative crops. These effects occur very rapidly even in the presence of very small amounts of excess ethylene after harvest, i.e., during shipping and storage. Consequently, an over-ripe state of the fruit is reached easily with a loss of its quality, and such fruits are then considered as unmarketable owing to the consequently reduced benefits of their shelf life (Alver, 2013). Thus, control of the ambient concentration of ethylene and removal of its

excess quantities from an ethylene-sensitive environment of fresh products are both important to delay over-ripening and to extend postharvest storage life. Various conventional methods have been proposed to remove ethylene from the ethylene-sensitive environment, such as adsorption and oxidation processes. Chemical ethylene scrubbers are the most commonly used instruments to control the ethylene concentration during postharvest storage (Elsgaard, 1998). In the adsorption process, zeolites and carbon-based adsorbents can be used for ethylene removal (Wills and Warton, 2004; Keller et al., 2013). However, chemical scrubbers have a major drawback in that they are costly in terms of their maintenance and replacement of chemical agents (Reddy et al., 2012; Zhao et al., 2012). To lower operation costs, a bioscrubber was introduced in the last two decades for pollutant removal without regeneration of a chemical agent (Kim, 2006; Liu et al., 2006; Arantes et al., 2009; Fu et al., 2011; Kazmierczak et al., 2013; Soltani et al., 2013; Xiong et al., 2013; Yu et al., 2013; Zolgharnein et al., 2013; Chand et al., 2014). Development of new valuable adsorbent using environmental-friendly materials is most desirable.

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Because agriculture byproducts and wastes are rich in cellulose, hemicelluloses, and lignin, they are assumed to be potential adsorbents on account of their biodegradability, nontoxicity, and separation efficiency (Sharma and Bhattacharyya, 2004; Hossain et al., 2005; Babu and Gupta, 2008; Reddy et al., 2012; Zhao et al., 2012; Kazmierczak et al., 2013; Nowicki et al., 2013). Tea is the most important agricultural commodity in the world. During the preparation of a tea beverage or the manufacturing of instant tea and bottled tea drinks, raw tea leaves is contacted with hot water or steam. From these operations, a solid residue known as spent tea leaves (STL) waste is produced. STL have no commercial value and are usually discarded as solid waste. Recent increases in tea consumption around the world have led to the production of large amounts of tea waste that requires disposal (Cai et al., 2015). Moreover, the increasing awareness of the need for waste reduction and environmental protection has stimulated the search for possible methods of using this waste (Zuorro and Lavecchia, 2012). However, STL are a non-conventional and cost-effective adsorbent (Hossain et al., 2005). Recently, Weng et al. (2013) explored the application of STL powder as a biosorbent for the elimination of pollutants from aqueous solution. STL saturated with organic contaminants can be composted. The use of STL as a potential source of biosorbent for phytohormone removal is another promising. However no study has focused on the phytohormone removal using STL. To obtain further insight into the potential application of STL, we studied the adsorption behavior of ethylene on STL powder.

An attempt was made to use microsized STL powder (MSTLP) as a biosorbent for the removal of the plant hormone ethylene in postharvest. To the best of our knowledge, no such study has been conducted thus far, despite the importance of control of ambient ethylene concentration from the viewpoint of preserving the freshness of fruits and vegetables. We expected the micro-processing treatment of STL to result in an increase in the specific surface area with an increase in ethylene adsorption capacity. We then investigated the effectiveness of MSTLP as a biosorbent for the removal of ethylene from a gaseous stream on the basis of its kinetics, adsorption equilibrium prospects, and regeneration capacity.

2. Material and methods

2.1. Preparation of MSTLP

STL was collected from a beverage chain store in Taiwan. It was washed thoroughly several times with double-distilled water to remove dust remaining on the STL surface. After pretreatment, STL was first dried at room temperature and then in an oven (80 °C) overnight. Dried STL, referred to as raw STL (RSTL), was then converted into fine STL powder (FSTLP) by grinding in a mechanical grinder and sieving through a 9-mesh sieve (2-mm mesh). FSTLP had an average particle size of about 830 μm. FSTLP was crushed into microsized powder via high-energy ball milling (PM100 Retsch, Germany) to obtain MSTLP. This MSTLP was kept in an airtight storage bag for further use in the adsorption study.

2.2. Characterization of MSTLP

The specific surface areas of RSTL, FSTLP, and MSTLP were measured by the Brunauer–Emmett–Teller nitrogen adsorption (BET-N₂) method using a Quantasorb surface area analyzer (BET Micromeritics ASAP 2020, USA). Functional groups of MSTLP were characterized using a Fourier transform infrared spectrometer (FTIR Spectrum 100 PerkinElmer, USA) in the solid state with KBr.

Microstructural and morphological characterizations of FSTLP and MSTLP were conducted using a scanning electron microscope (SEM, S2700 Hitachi, Japan) at an acceleration voltage of 20 kV after gold sputtering under vacuum. The average particle size and size distribution were determined using a dynamic light scattering (DLS) analyzer (Nano-ZS, Malvern Instrument Ltd., Malvern, UK).

2.3. Kinetic and equilibrium adsorption experiments

Kinetics experiments of C₂H₄ adsorption onto MSTLP were performed to establish the effect of time on the adsorption process and to quantify the adsorption rate. Experiments were conducted with an initial C₂H₄ concentration of 2580 μmol/L and adsorbent dosage of 5 g. The effects of adsorbent dosage and ethylene concentration on the adsorption kinetics were determined by varying the adsorbent dosage from 1 to 5 g and using different initial C₂H₄ concentrations (6–3350 μmol/L) at 25 °C. Ethylene was obtained in a compressed-gas cylinder. The typical experimental procedure was as follows. The desired amount of MSTLP was placed in a reactor, following which C₂H₄ was injected into the reactor. The rotation rate of the mixer (RM-2 Elmi, USA) was 50 rpm. When the reactor was rotating, the gas was withdrawn at designed time intervals. Temperature was controlled to isothermal conditions at 5, 15, 25, 35, and 45 °C by placing the entire assembly in an isothermal incubator (RIN-45 RISEN, Taiwan). The reactors were first placed in the incubator for 1 h prior to the experiment to establish thermal equilibrium. Samples were analyzed using a gas chromatograph (GC, PerkinElmer, Claurus 500, USA) equipped with a flame ionization detector (FID) and a thermal conductivity detector (TCD). The GC was equipped with an Elite Plot Q capillary column (Agilent Technologies) with a length of 30 m, film thickness of 0.25 μm, and internal diameter of 0.32 mm. Each run of the experiment was replicated, and the average values were used for data analysis. Blank tests without adsorbent were run in parallel to avoid possible adsorption on the reactor. Isotherm experiments were performed to determine the maximum adsorption capacity and thermodynamic parameters. The experimental procedure in these experiments was the same as that described above. The amount of C₂H₄ adsorbed per unit mass of the adsorbent (*q*, in μmol/g) was computed using the following expression:

$$q = \frac{(C_0 - C_t)V}{m}, \quad (1)$$

where *C*₀ and *C*_{*t*} are the C₂H₄ concentrations in μmol/L before and after adsorption, respectively, for time *t*; *m* (g) is the amount of biosorbent; and *V* is the volume (L) of the reactor.

2.4. Mathematical model

2.4.1. Kinetics of adsorption

The kinetic data were analyzed using the pseudo-second-order (PSO) model, expressed as

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

where *q*_{*e*} and *q*_{*t*} denote the amounts of ethylene adsorbed on MSTLP (μmol/g) at equilibrium and at time *t* (min), respectively, and *k*₂ is the PSO rate constant (g/μmol·min). After integration of the above equation and application of boundary conditions *q*_{*t*} = 0 at *t* = 0 and *q*_{*t*} = *q*_{*t*} at *t* = *t*, the integrated form of the equation becomes

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