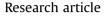
#### Journal of Environmental Management 181 (2016) 261-268

Contents lists available at ScienceDirect

### Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman





# Remediation of aquaculture water in the estuarine wetlands using coal cinder-zeolite balls/reed wetland combination strategy



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#### ARTICLE INFO

Article history: Received 20 January 2016 Received in revised form 16 June 2016 Accepted 21 June 2016

Keywords: Estuarine wetlands Aquaculture water Coal cinder-zeolite balls (CCZBs) Reed wetland Remediation

#### ABSTRACT

In this paper, the modified coal cinders and zeolite powders in proportion of 2:1 were mixed with modified polyvinyl alcohol (PVA) with a ratio of 20:1 (w/v) to make a new sorbent and biological carrier-the coal cinder-zeolite balls (CCZBs). The maximum absorption capacities of ammonia nitrogen and Chemical Oxygen Demand (COD<sub>Cr</sub>) on CCZBs, adsorption process were evaluated in batch experiments. And then they were combined with reed wetland for bioremediation of micro-polluted aquaculture water in estuarine wetlands. The results showed that the removal efficiencies of ammonia nitrogen and COD<sub>Cr</sub> improved with the decrease in water inflow and increase in inflow concentrations. Efficiencies of 67.3% and 71.3% for ammonia nitrogen and COD<sub>Cr</sub> under water flow of 10 L/h were obtained when their inflow concentrations were 1.77 and 56.0 mg/L respectively. This strategy can be served as a model system for bioremediation in situ of aquaculture water and other organic polluted or eutrophic water. © 2016 Elsevier Ltd. All rights reserved.

#### 1. Introduction

Estuarine wetlands play a significant role in improving climatic conditions, maintaining biodiversity, saving water, purifying pollutants (Yang et al., 2011; Cheng and Zhou, 2012). But the estuarine delta is an active land-sea interaction zone affected by the interactive processes of marine, terrestrial and riverine ecosystems, so it is also a comparatively sensitive and fragile ecosystem (Wang et al., 2007; Haines, 2013; Tian et al., 2014). In recent years, the estuarine wetlands, an important component of the estuarine delta, have become the sites of increasing economic activity. Many estuarine wetlands are exploited for commercial aquaculture and farming. These projects produce increasing pressure on harsh environmental pollution and the degradation of wetland ecosystem services.

The Liaohe estuarine wetland, located in the northern Liaodong Bay of the Bohai Sea, is the largest reed (*Phragmites communis* Trin.) wetland in Asia and the second largest in the world. And it covers a geographic area of approximately  $8 \times 10^4$  ha (Yang et al., 2011; Lin et al., 2013). The reeds, as the dominant plant species in this

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wetland, are used for raw materials to make paper. So, to increase the quantities of reeds, some field and irrigation engineering constructions were built. The wetland was divided into many little wetland cells with similar size and shape according to terrian condition. And circular ditch was equipped in each wetland cell. The river flowed through every cell via a main canal and two inflow ditches during the irrigation season. Before harvesting reeds the irrigation water finally converged at the outlet ditch and discharged into Bohai Sea through the nearest river. The retention of water in circular ditches provided favorable conditions to develop commercial aquaculture in the reed filed wetland. However, large quantities of organic matter and nutrients such as nitrogen were deposited in the water because of excessive inputs of manure/feed. The contaminants accumulating in the reed filed wetland brought certain influence to the wetland ecological system and coastal environment. According to our previous research ammonia nitrogen, total phosphorus and chemical oxygen demand (COD<sub>Cr</sub>) levels in the aquaculture water of the reed wetland were in the range of 0.87-1.65 mg/L, 0.037-0.189 mg/L and 37.8-106.9 mg/L, respectively. The concentrations of ammonia nitrogen and COD<sub>Cr</sub> of most sites in the reed wetland exceeded class IV 'Environmental Quality standards for Surface Water' in China (MEP and AQSIQ, 2002).

Rotating biological contactors, trickling filters, fluidized bed reactors, membrane filters, aeration stripping, ozone oxidation,

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bead filters and biofilters are conventionally used in purification of intensive aquaculture water (Yang et al., 2001; Tango and Gagnon, 2003; Nora'aini et al., 2005; Eding et al., 2006; Crab et al., 2007; Sánchez and Matsumoto, 2012). Besides these conventional water treatment systems, many studies have reported on bioremediation for aquaculture water such as phytoremediation, microalgae ponds, photosynthetic bacteria treatment, etc. and the application of these methods has received better removal effects. (Lin et al., 2002; Ghalv and Mahmoud, 2005; Lin et al., 2005; Lymbery et al., 2006; Metaxa et al., 2006; Sindilariu et al., 2007; Snow and Ghaly, 2008; Meng et al., 2009). But aquaculture in the reed filed wetland is different from the conventional aquaculture system. It belongs to a distributed aquaculture model. Water quantities in circular ditches change with the seasons, and there are four irrigations and drainages during one reed-growing season. In addition, water in circular ditches cannot be pumped out for purification according to the mode of high yields of reed. So the traditional water treatment technologies are not applicable to the aquaculture water in reed field wetland. Therefore, a new method must be studied for removal of organic matters and nutrients accumulated in estuarine wetlands.

In this paper, the coal cinder-zeolite balls (CCZBs) were developed according to the aquaculture characteristics of reed field wetland. The adsorption process and the maximum absorption levels of these CCZBs for ammonia nitrogen and  $COD_{Cr}$  were evaluated by batch experiments. Then, they were combined with reed wetland to remove ammonia nitrogen and  $COD_{Cr}$  in aquaculture water. And the removal efficiencies and optimal conditions were investigated.

#### 2. Materials and methods

#### 2.1. Raw materials

The coal cinders, obtained from Qinghemen (Liaoning province, China), were dried in a drying closet for 24 h at 105 °C prior to use. The chemical compositions of the raw coal cinders were given in Table 1. Zeolite was purchased from Huazhijie Aquipment Co., Ltd., Qingdao, China. The other solvents and chemicals used in this study were of an analytical grade.

#### 2.2. Preparation of coal cinder-zeolite balls

Coal cinders and zeolite were used as carrier materials because of their honeycombed, inexpensive and easily available properties. Coal cinders and zeolite were crushed and sieved through a 0.15mm mesh. It is known that coal cinders contained a large number of deleterious metals such as Al, Cr, Cu, As, Hg, Cd and Pb (Zhang et al., 2013). To avoid involving secondary pollutants, coal cinders were soaked in 6 mol/L hydrochloric acid (HCl) for 24 h to remove heavy metals, and then were repeatedly washed with sterile distilled water until the pH came back to 6–7. The treated materials were then dried in a closed container at room temperature. The mixture of the coal cinders and zeolite powders in proportion of 2:1 were mixed with modified polyvinyl alcohol (PVA) with a ratio of 20:1 (w/v) to make CCZBs in a balling plate (BY-4). PVA was modified by succinic acid with the mass ratio of 5.6:1 at 85 °C to improve their water resistance and adherence (Zhou et al., 2015). The CCZBs were

Table	1
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Composition con	tent of raw (	coal cinders.
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Composition	SiO <sub>2</sub>	$Al_2O_3$	TFe	TiO <sub>2</sub>	CaO	MgO	CaF	$MnO_2$
Content/%	62.25	20.72	5.75	0.59	1.33	2.05	0.35	0.85

dried at 40 °C for 48 h in a closet for the surface areas and interior structures analysis. The weight of each CCZB was 19.7  $\pm$  0.3 g.

#### 2.3. Batch sorption experiments

#### 2.3.1. Ammonia nitrogen and COD<sub>Cr</sub> sorption kinetics

The ammonia nitrogen and  $COD_{Cr}$  sorption kinetics of CCZBs were determined using batch experiments. Two CCZBs as a group were separated placed into two 500 mL capped conical flasks containing 400 mL standard ammonia nitrogen and  $COD_{Cr}$  solutions with initial concentration of 10 and 50 mg/L. The mixtures were then shaken by a horizontal oscillator (XJ-III, Shaoguan Taihong medical instrument Co., Ltd, China) at 25 °C for 550 min. Then the concentrations of ammonia nitrogen and  $COD_{Cr}$  were measured at different times.

#### 2.3.2. Ammonia nitrogen and COD<sub>Cr</sub> sorption isotherms

For sorption isotherms studies, eight ammonia nitrogen solutions (2, 4, 6, 8, 10, 15, 20, 25 mg/L) were prepared with NH<sub>4</sub>Cl. And a series of COD<sub>Cr</sub> solutions, ranging from 30 to 350 mg/L (30, 40, 50, 60, 80, 100, 120, 180, 250, 350 mg/L) were prepared with potassium hydrogen phthalate (KHP). Each CCZB was added to 200 mL different ammonia nitrogen and COD<sub>Cr</sub> solutions, respectively. Then mixtures were shaken by a horizontal oscillator at 25 °C for 24 h. The concentrations of ammonia nitrogen and  $COD_{Cr}$  left in the water samples were tested. The ammonia nitrogen and  $COD_{Cr}$  adsorption capacities were calculated by Eq (1).

$$Q_t = \frac{(c_o - c_t)V}{m} \tag{1}$$

where,  $Q_t$  (mg/g) is the adsorption capacity of ammonia nitrogen and COD<sub>Cr</sub>,  $c_o$  (mg/L) and  $c_t$  (mg/L) are the initial and final concentrations of ammonia nitrogen and COD<sub>Cr</sub>, respectively. *V*(L) is the solution volume. *m*(g) is the mass of CCZBs.

#### 2.4. Kinetic and equilibrium models

To study the controlling mechanism of the adsorption process, the well-known Pseudo-first-order kinetic model Eq. (2) (Lagergren, 1898), pseudo second-order kinetic model Eq. (3) (Ho and McKay, 1998), and intral-particle diffusion equation Eq. (4) were used to test experimental data. These models were presented below.

$$ln(q_e - q_t) = lnq_e - k_1 t \tag{2}$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t$$
(3)

$$q_t = k_3 t^{0.5} + c \tag{4}$$

where,  $q_t$  and  $q_e$  (mg/g) are the adsorption capacities of CCZBs at time *t* (h) and at equilibrium, respectively;  $k_1$ ,  $k_2$ ,  $k_3$  are the equilibrium rate constants of pseudo-first-order sorption pseudo-second-order sorption and intral-particle diffusion expressed in min<sup>-1</sup>, g/(mg·min), g/(mg·min<sup>0.5</sup>) respectively; and *t* is time (min).

Langmuirand Freundlich isotherm models (Langmuir, 1918; Freundlich, 1906) were applied to fit the equilibrium data of ammonia nitrogen and  $COD_{Cr}$  adsorption.

The Langmuir model was expressed as follows:

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