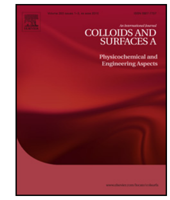
Colloids and Surfaces A: Physicochem. Eng. Aspects 484 (2015) 99–107

Contents lists available at ScienceDirect

Colloids and Surfaces A: Physicochemical and Engineering Aspects

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



Development of a novel integrated membrane system incorporated with an activated coke adsorption unit for advanced coal gasification wastewater treatment

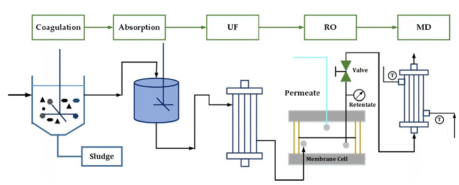
Lili Xu[a,b,c], Jun Wang[a,b,*], Xiaohui Zhang[a,b], Deyin Hou[a,b], Yang Yu[d]

[a] Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China
[b] Beijing Key Laboratory of Industrial Wastewater Treatment and Reuse, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China
[c] Membranes@Bath and Department of Chemical Engineering, University of Bath, Bath BA2 7AY, United Kingdom
[d] Department of Civil and Environmental Engineering, National University of Singapore, 119260, Singapore

HIGHLIGHTS

- Activated coke (AC), a low-cost substitute to the more expensive activated carbon, is firstly used as the pretreatment of integrated membrane processes for the advanced coal gasification wastewater treatment.
- The effect of AC adsorption pretreatment on the UF, RO, and MD units is investigated.
- The mechanism that AC adsorption alleviates membrane fouling is discussed.
- TOC, FEEM, MWD, and GC–MS give a systematic characterization of water quality.
- The techniques that activated coke used as pretreatment of integrated membrane processes has been applied in a local coal gasification plant in China.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 14 June 2015
Received in revised form 26 July 2015
Accepted 29 July 2015
Available online 1 August 2015

Keywords:
Coal gasification wastewater
Activated coke
Adsorption
Integrated membrane systems

ABSTRACT

Lurgi–Ruhrgas coal gasification process is commonly used to produce methane in many regions. However, such a process produces wastewater containing toxic pollutants that are difficult to remove by the conventional integrated membrane system consisting of a coagulation unit, an ultrafiltration (UF) unit, a reverse osmosis (RO) unit, and a membrane distillation (MD) unit. In this regard, an activated coke was added prior to the UF unit in order to remove the organic matters and meet the wastewater discharge regulation. The activated coke is an abundant, low-cost substitute to the more commonly used expensive activated carbon. The effect of the adsorption unit on the removal efficiency and membrane fouling

* Corresponding author at: Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China. Fax: +86 10 62849198.
E-mail address: junwang@rcees.ac.cn (J. Wang).

http://dx.doi.org/10.1016/j.colsurfa.2015.07.062

mitigation was systematically studied with real coal gasification wastewater. The results revealed that majority of the organic matters were effectively removed by the adsorption unit. Consequently, the fouling phenomenon of the subsequent membrane units was suppressed. The permeate fluxes of the UF, RO, and MD units increased by 31.6%, 21.9%, and 23.1%, respectively, when comparing the fluxes between the processes with and without the adsorption unit. The results demonstrated that coupling activated coke adsorption with integrated membrane units was an attractive and feasible option for the advanced treatment of coal gasification wastewater.

1. Introduction

Lurgi–Ruhrgas coal gasification process is commonly used to produce large quantity of syngas (methane as the primary component) in the regions rich in coal but poor in oil or gas (e.g., China) [1,2]. The wastewater discharged from the Lurgi–Ruhrgas gasifier usually contains large amount of toxic organic and inorganic pollutants, including phenolic compounds, polynuclear aromatic hydrocarbons, heterocyclic compounds, ammonium, sulfate, cyanide, and thiocyanate which are required to be removed to meet the wastewater discharge regulation [3,4]. However, most of the currently available treatment processes, such as anoxic–oxic (A/O) and anaerobic–anoxic–oxic (A^2/O), were proven to be insufficient to reduce these contaminants to an acceptable level [5,6]. In this regard, the need for an effective and efficient process to treat the coal gasification wastewater is certainly warranted.

Among all the research efforts aiming for maximally reducing the contaminant contents in the coal gasification wastewater, the so-called “integrated membrane system (IMS)” combining various types of membrane processes and other separation processes has received considerable attention mainly because such an integrated configuration synergistically enhances the advantages of each individual process, and subsequently improves the contaminant removal efficiency from the coal gasification wastewater [7–12]. A typical IMS configuration for coal gasification wastewater treatment consists of a coagulation pre-treatment unit for colloids removal, an ultrafiltration (UF) unit to remove suspended solids, a reverse osmosis (RO) unit for metal ions and small organic matters removal, and a complimentary membrane distillation (MD) unit to further increase the global water recovery of the process. However, despite the great benefits derived from this process, the undesired fouling behavior commonly observed in the UF, RO, and MD unit processes causes a substantially loss in clean water productivity and quality, thus impeding the further advance of IMS in coal gasification wastewater treatment, where large quantity of wastewater is required to be purified and strict discharge regulation needs to be complied with [13–15]. With this concern, developing an appropriate strategy and IMS configuration not only delivering higher contaminant removal efficiency but also suppressing the undesirable membrane fouling phenomenon is of great importance for the IMS process.

The organic matters present in the coal gasification wastewater are believed to be the main foulants that (i) foul the UF membrane thus causing the flux decline in the UF process, (ii) wet the RO membrane surface and consequently lower the salt rejection, and (iii) accumulate on the MD membrane surface thus encouraging the crystallization and scaling of the metal ions on the membrane surface, and consequently lead to a lowered salt rejection. As such, the addition of a pre-treatment step to eliminate these organic foulants thus minimizing the fouling propensity in each individual membrane unit process has become a research focal point and several separation techniques have been considered as the potential pre-treatment candidates. Among them, the use of activated carbon to adsorb contaminant compound (mainly organic matters) in the wastewater shows great process flexibility and potential to be incorporated in the IMS process as a pre-treatment to safeguard the subsequent membrane processes. However, several issues associated with the activated carbon substantially limit the feasibility of using such a material in the wastewater treatment, including: (i) the difficulty to regenerate the activated carbon, and (ii) the high cost of these adsorbents [16–19]. In this regard, it is essential to search for a low-cost, more efficient and easily accessible adsorbent. Activated coke, a low-cost adsorbent, is often used as a substitute adsorbent for activated carbon, due to (i) its excellent SO_X and NO_X adsorption ability in the gas phase, and (ii) its exceptional adsorption capacity to remove organic pollutants from the liquid phase. Furthermore, activated coke is produced from naturally occurring carbonaceous materials such as lignite, petroleum coke, wood and other biomass [16,18,20,21]. Therefore the abundant resource of activated coke in a coal gasification plant guarantees a secured and low-cost supply of such absorbent in the IMS process. However, to the best knowledge of the authors, no studies have yet been reported on the incorporation of an activated coke adsorption unit in an IMS process for the coal gasification wastewater treatment.

In this study, the addition of an adsorption unit with activated coke as a pre-treatment step prior to the UF unit process in a typical IMS configuration was studied. Real coal gasification wastewater provided by a local coal gasification plant instead of synthetic model wastewater was used to demonstrate the feasibility of using such a system in an industrial-relevant environment. The effect of the activated coke adsorption pre-treatment on (i) contaminant removal, and (ii) membrane fouling mitigation was systematically examined by comparing the performance of two different IMS configurations (with and without activated coke adsorption unit process). The effluent properties of each unit process were characterized by total organic carbon (TOC), fluorescence excitation-emission matrix (FEEM), molecular weight distribution (MWD), and gas chromatograph/mass spectroscopy (GC–MS) analysis. In addition, the surface morphology of the activated coke as well as the fouling layer formed on the membrane surface was characterized by the scanning electron microscopy (SEM).

2. Materials and methods

2.1. Materials

The wastewater influent used in the experiments was provided by Yima gasification plant (Henan, China). The existing treatment processes in the plant included a phenolic compounds extraction unit, an ammonia striping unit, and a sequencing batch reactor (SBR) treatment. After being treated by a series of chemical and biological treatments, the wastewater was still deep brown in color with a bad odor. The chemical and physical properties of the wastewater are showed in Table 1.

The activated coke from lignite was obtained from the same gasification plant, with particle sizes ranging from 0.038 mm to 0.15 mm, and the Brunauer–Emmett–Teller (BET) surface area of