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Coke dust enhances coke plant wastewater treatment

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HIGHLIGHTS

- We researched industrial adsorptive coke plant wastewater treatment.
- Two types of coke dust were examined in three scenarios each.
- COD, TOC, 16 PAHs, phenols, cyanide, thiocyanates, ammonium, nitrogen were analyzed.
- Coke dust addition enhanced many steps of coke plant wastewater treatment.
- The enhanced biological treatment proved the most effective.

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ABSTRACT

Coke plant wastewater contain many toxic pollutants. Despite physico-chemical and biological treatment this specific type of wastewater has a significant impact on environment and human health. This article presents results of research on industrial adsorptive coke plant wastewater treatment. As a sorbent the coke dust, dozen times less expensive than pulverized activated carbon, was used. Treatment was conducted in three scenarios: adsorptive after full treatment with coke dust at 15 g L^{-1}, biological treatment enhanced with coke dust at 0.3–0.5 g L^{-1} and addition of coke dust at 0.3 g L^{-1} prior to the biological treatment. The enhanced biological treatment proved the most effective. It allowed additional removal of 147–178 mg COD kg^{-1} of coke dust.

1. Introduction

A large quantity of wastewater is generated during coke production. Many processes are involved in production. These include: pyrolysis of a coal mixture in a coking chamber, and cooling and treatment of volatile products of coal pyrolysis. They are responsible for generation of by-products. Thus, the wastewater contains among others: ammonia, cyanides, thiocyanates, phenols and other organic compounds, such as PAHs (Ghose, 2002; Ning et al., 2005; Dameng et al., 2011; Burmistrz and Burmistrz, 2013; Zhang et al., 2013). Coke plant wastewater is characterized by low concentration of heavy metals and phosphorus. Concentration of main pollutants in coke plan wastewater depend on many factors (Ghose, 2002; Vazquez et al., 2007).

Conventional coke plant wastewater treatment utilize physico-chemical and biological methods. Among the proven biological treatment methods of coke plant wastewater, activated sludge processes is commonly used (Lee et al., 2002; Woo et al., 2009). In many countries, including Poland, preferred system is based on a single-sludge process with recycle of nitrified effluent, *i.e.* pre-denitrification process (Li et al., 2003; Kim et al., 2007). Despite complex treatment coke plant wastewater can have adverse impact on ecosystem and human health (Zhang et al., 2012; Zhang et al., 2013).

Increase of efficiency of coke plant wastewater treatment is possible by application of adsorption methods. Many research confirmed removal of phenols (Ahmaruzzaman and Sharma, 2005; Vazquez et al., 2007; Mohanty et al., 2008; Spiridon et al., 2013), cyanide (Zhang et al., 2010a,b), PAHs and other organic compounds (Zhang et al., 2010a,b; Zhang et al., 2012; Ali et al., 2013; Zhang et al., 2013) from industrial wastewaters after utilization of organic sorbents.

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Implementation of adsorption methods to treat coke plant wastewater runs up against a cost barrier emerging from high prices of commercial sorbents. High cost is due to troublesome regeneration of the used sorbents (Ghose, 2002). The most common thermal regeneration of coal consumes lot of energy and results in 5–10% mass loss of regenerated coal (Asghar et al., 2013). An attempt to overcome this barrier is made by using materials with fairly good sorptive properties and the lowest possible price, enabling a single application without need of regeneration (Burmistrz et al. 2003; Ali et al., 2013). Studies showed that coke dust generated in coke plants using coke dry quenching methods can be used as such material (Errera and Milanez, 2000; Diez et al., 2002). The coke dust is a by-product of the coke plant production process and is less expensive than commercial powdered activated carbons (PAC) by at least order of magnitude.

2. Material and methods

2.1. Characterization of coke dust

Coke dust samples were taken from two Polish coke plants in cities of Dąbrowa Górnicza (DG) and Zdzieszowice (Z). The studies used two types of coke dust. First one was acquired from coke plant with dry quenching installation (Q), and the second from coke plant without dry quenching installation (NQ). The NQ dust is not activated by contact with the cooling gas. The Q dust is activated in a certain way through contact with hot gas in the dry quenching installation.

Samples of coke dust underwent grain size, proximate and ultimate analysis in compliance with ISO standards. Based on experimental isotherms of argon adsorption and desorption at 77.5 K performed in automatic Sorptomatic apparatus the specific surface area (S_{BET}) and pore volume, including micropores and mesopores were determined for both studied coke dust types.

2.2. Adsorptive polishing of coke plant wastewater after biological treatment

The diagram of the technological system for research on adsorptive coke plant wastewater treatment is shown in Fig. 1A. Coke plant wastewater was treated in accordance with the technology used in the plant, encompassing physico-chemical and biological treatment using the activated sludge method. Wastewater was introduced to adsorption contactor at a rate of 14–17 $m^3\,h^{-1}$ (average 15 $m^3\,h^{-1}$) with a volume of 200 m^3, into which Q coke dust was added in quantity of 15 $g\,L^{-1}$ of treated wastewater. Mixing coke dust with treated wastewater was performed using air pumped through an aeration grate located at the bottom of the contactor. Average time wastewater spent in the contactor was 13 h. Effluent wastewater with the coke dust was pumped to a settling tank, from where it was transported to multi-purpose reactors. In order to speed up the settling of the particulate suspension, an aqueous solution of $Al_2(SO_4)_3$ was added to wastewater in sedimentation tank. Solution of polyelectrolyte was added to the multi-purpose reactor.

During that period, discrete 4 L samples of wastewater were taken at 8 h intervals in points A1 and A2. Analysis was performed as described in Section 2.5.

2.3. Enhancing the biodegradation process by coke dust addition

Research was conducted using coke dust Q. One of the biological reactors in wastewater treatment plant was used for the purpose of the test. Coke dust was added into biological reactors. Volume of the reactor was 750 m^3. Two scenarios were conducted in coke plant DG: addition of 0.3 and 0.5 $g\,L^{-1}$ of Q dust. The average time wastewater spent in the reactor was 22 h. Samples of wastewater leaving the biological reactor after biodegradation process enhanced with coke dust were taken. As a reference, samples from an adjacent biological reactor in which identical coke plant wastewater was treated without coke dust addition. Sampling sites are shown in Fig. 1B. Each sample was proceeded as described in Section 2.5, immediately after acquisition.

2.4. Addition of coke dust prior to the biological treatment

In this assay wastewater treatment system of coke plant Z was used. The coagulation is one of routine steps of physico-chemical treatment. As a coagulant, saturated aqueous solution of $FeSO_4$ in 20% $Fe_2(SO_4)_3$ with Fe^{2+}:Fe^{3+} molar ratio of 1:2 was used. Applied dose was 0.97 mg (density 1.55 $g\,L^{-1}$, pH 0.8–1.1, 69.8 $Fe^{3+}\,g\,L^{-1}$, 25.5 $Fe^{2+}\,g\,L^{-1}$) of coagulant L^{-1} of wastewater. Wastewater treatment system was enhanced by addition of coke dust NQ before biological treatment stage. The coke dust NQ was added in amount of 0.30 ± 0.05 $g\,L^{-1}$ into line mixer in which the coagulant solution is added. Water treatment plant scheme is presented in Fig. 1C. During a 14 d test composite wastewater samples were taken in points C1, C2 and C3 with average 8 h interval. During the first 2 d of test, the coagulation process remained unmodified. Starting with the 3rd d coke dust was added to treated wastewater. Sample proceeding, range of analysis and detailed methods were described in Section 2.5.

2.5. Scope and analysis method of coke plant wastewater samples

All samples of coke plant wastewater were analyzed as soon as possible after acquisition. If samples could not be analyzed immediately, they were preserved and placed in a refrigerator at a temperature of 4 °C. The scope and analysis methods included: COD index in accordance with US EPA procedure – method 410.4, 1983, total organic carbon (TOC) determination using Shimadzu TOC-5000A automatic analyzer, phenol – bromometric method in accordance with Polish Standard, colorimetric method with 4-aminoantipyrin if phenol concentration was below 5 $mg\,L^{-1}$, cyanide and thiocyanate content – colorimetric method using HACH 2000 spectrophotometer, ammonium content using Nessler method acc. to ASTM D 1426–96 – method A, 16 PAHs from the US EPA reference list were extracted using BAKER spe system and BAKERBOND octadecyl (C18) columns. The obtained eluates were analyzed using HPLC technique (Varian RES ELUT-ENV 4.5 μ SPHERICAL 150 × 4.6 mm column, eluent: acetonitrile/water, isocratic 80/100% acetonitrile) with UV detection at 254 nm (Burmistrz and Burmistrz, 2013).

2.6. Statistical description of achieved results

For quantitative analysis of wastewater components in specified sampling sites the following values were calculated for each tested parameter: arithmetic mean, standard deviation (SD) and 95% confidence interval.

For describing efficiency of studied sorption process, the percentage of removal for specified pollutant was used. To assess quality of research results SD or 95% confidence interval was used.

3. Results and discussion

3.1. Characteristics of coke dusts

Coke dust obtained in the coke dry quenching process (Q) has certain specific properties which differentiate it from dusts not