

Transformation of natural and synthetic dyes in pulsed electric discharge in the granular steel bed

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ABSTRACT

Humic substances in groundwaters containing dissolved iron form stable colloids. Pulsed spark discharge in a granular steel bed was applied for abatement humic substances and synthetic indicators mimicking the target molecule fragments. A reversible red-ox cationic methylene blue, an anionic eosin Y and irreversible red-ox indicator furacilin were used to disclose the reactions and adsorption on the discharge erosion products. Partial, for about 40%, mineralization of organic molecules was observed for all substances, whereas rapid red-ox reactions also took place between organic molecules and dissolved ferrous iron. Humic substances adsorbed at the spark erosion products with their anionic groups.

1. Introduction

In marsh-ridden territories of Western Siberia, the ground water sources are predominantly used for potable water supply. Concomitantly with high contents of humic substances (HS), ground waters contain substantial amounts of dissolved ferrous ions. Numerous carboxylic and phenolic groups of HS macromolecules make those chemically active in bonding metallic ions resulting in formation of stable colloids [1]. Conventional treatment of such ground water, targeting oxidation of ferrous iron with its precipitation in the ferric hydroxide form, includes air stripping with subsequent settling and filtration. This approach, however, is ineffective against colloids stabilized with HS [2]. The removal of colloids with coagulation-flocculation or electrocoagulation [3] has its drawback of massive sludge formation requiring further handling, which may also appear insufficient with HS present due to restabilization of colloids and, thus, requiring substantial coagulant doses for sweep coagulation [4]. Adsorption on activated carbons and membrane filtration effectively remove dissolved and colloid HS, although high cost of treatment [5–8] and irreversible fouling of membranes are problematic. Application of ozone and other advanced oxidation processes (AOPs) to the HS oxidation are well known for their positive discoloration and colloid particle destabilization effect [9]. More detailed studies in ozone impact to the HS colloid stability showed strong aggregation effect at mineral surfaces [10]. The costly character of ozone application is, nevertheless, well known [11].

One of the promising methods applied to the treatment of waters is application of various electric discharges realizing an integral physico-chemical action on the organic and inorganic pollutants [12,13]. The authors of the present research earlier proposed application of the pulsed spark discharge (PSD) in the iron and aluminium granular beds applied for the removal of coloured organic compounds [14]. The discharges in the bed of iron granules appeared beneficial for practical application due to the bivalent character of iron ions resulting in their red-ox reactions forming insoluble hydroxides and salts in a wide pH diapason.

The objective of the present study is establishing the regularities in red-ox, decomposition and adsorption processes taking place in the PSD reactor containing iron granules applied to elimination of aqueous coloured natural and synthetic organic compounds. Humic substances and synthetic methylene blue, furacilin and eosin Y resembling the fragments of natural HS with their specific reactive and adsorption properties were chosen to be the research objects. The synthetic dyes showing red-ox and cation- and anion-adsorption properties were used as indicators of adsorption by the spark erosion products, extrapolated to the adsorption of HS.

2. Materials and methods

2.1. Chemicals

The 4-% HS solution was extracted with alkali from the peat of Tomsk Region at the Siberian Research Institute of Agriculture and Peat of

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Table 1
Coloured organic substances used in the study and their characteristics [16].

Title	Formula	Properties
Humic substances	See Fig. 1	Yields to red-ox and acid-base reactions, chelating agent for heavy metals
Methylene blue	$C_{16}H_{18}ClN_3S \cdot HCl$	Reversible red-ox and cation-adsorption indicator. Dissociates forming $[C_{16}H_{18}ClN_3S]H^+$ cation
Eosin Y	$Na_2[C_{20}H_6O_5Br_4]$	Irreversible red-ox and anion-adsorption indicator. Dissociates forming an anion $[C_{20}H_6O_5Br_4]^{2-}$
Furacilin	$C_6H_6N_4O_4$	Yields to red-ox reactions, oxidized with destruction

Russian Academy of Agricultural Sciences. A peat sample was extracted with 0.1-M NaOH solution at 80 °C for 1 h with subsequent soaking at ambient temperature overnight. Humic substances were precipitated from the alkaline centrifugate with 10-% HCl solution with subsequent centrifugation, rinsing with 0.5-% HCl solution and distilled water to the neutral reaction, and dried at 80 °C. Obtained HS were dissolved in distilled water [15]. Synthetic dyes simulating the fragments of the HS molecular structure, methylene blue (3,7-bis(dimethylamino)-phenothiazin-5-ium chloride), furacilin ((2E)-2-[(5-nitro-2-furyl)methylene]hydrazine carboxamide) and eosin Y (2-(2,4,5,7-tetrabromo-6-oxido-3-oxo-3H-xanthen-9-yl)benzoate) (see Table 1 and Fig. 1) were studied in the experiments, having the starting concentration of the dyes of

20 mg L⁻¹. The concentration was chosen for reliable spectrophotometric quantification in treated samples. Methylene blue and furacilin were provided by JSC CosmoKhim, Russia; eosin Y was supplied by JSC KhimReaktiv, Ukraine. The chemicals were of “chemically pure” grade identical to *Purissimum*, i.e. > 99% of the assay content.

2.2. Experimental equipment

The outline of the pulsed electric discharge reactor is given in Fig. 2. The experimental device consists of the 1.5-L rectangular reactor vessel made of acrylic glass with the horizontal rectangular rod electrodes, and the source of pulsed voltage fed from the 220-V 50-Hz mains. The

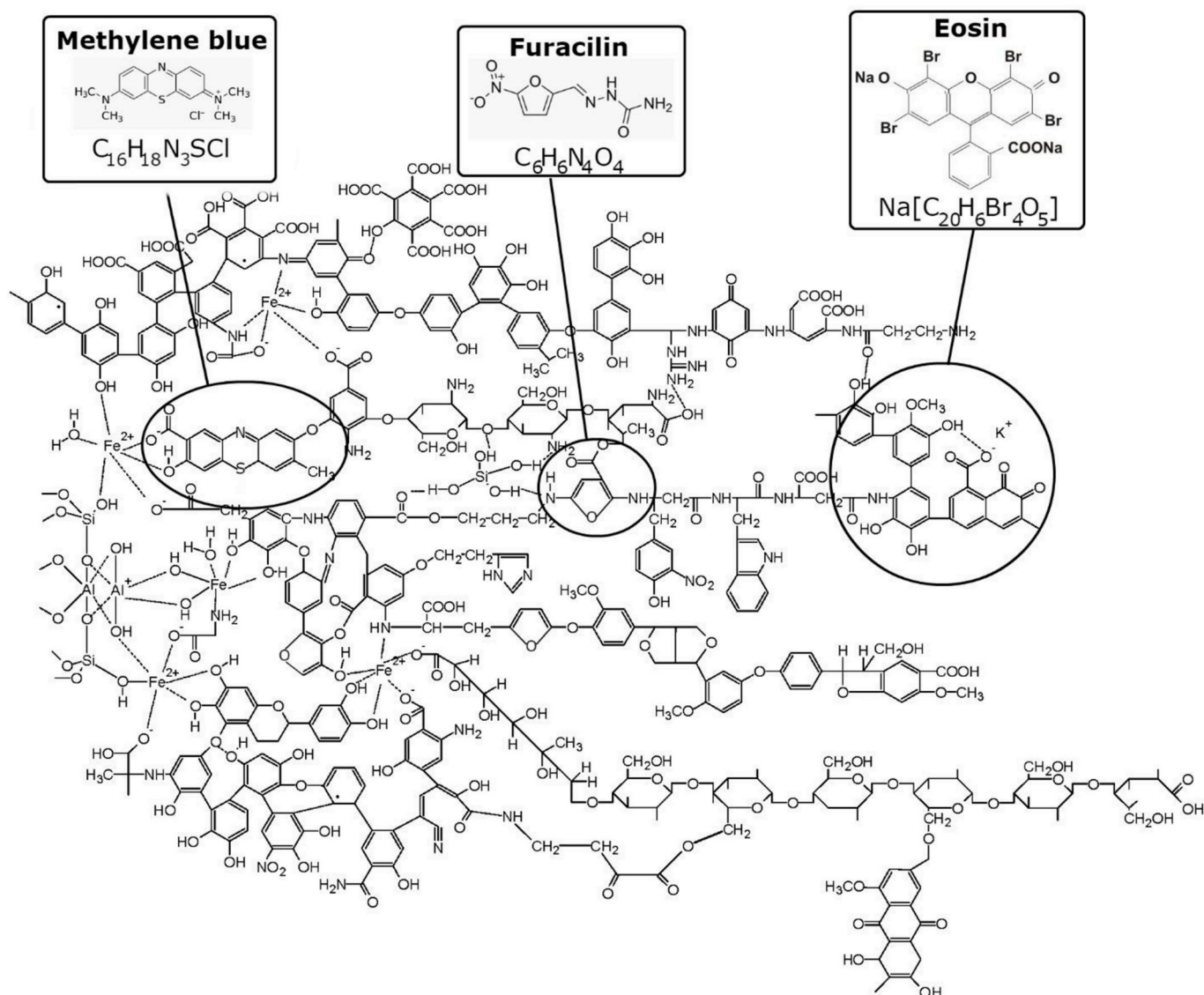


Fig. 1. Structural element of humic substance [23] with fragments resembled by methylene blue, furacilin and eosin Y.

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