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## CONTENTS

### 6<sup>th</sup> INTERNATIONAL ACADEMIC CONFERENCE “CHEMISTRY & CHEMICAL TECHNOLOGY 2017”

<i>Antoniuk V., Krasinskyi V., Khamula N., Yurkiv I.</i> Rheological Properties of Compositions Based on Modified Polyvinyl Alcohol.....	22
<i>Saiuk D., Bukartyk N., Shevchuk O., Tokarev V.</i> Cross-Linked Polymer Hydrogels with Embedded Semiconductor Nanocrystals.....	24
<i>Znak Z., Sukhatskii Y., Zin O., Mnykh R., Khomiak S., Hrabarovska A.</i> Peculiarities of Benzene Decomposition in Cavitation Fields.....	26
<i>Skoretska I., Beznosyk Yu.</i> Modeling of Heterogeneous Catalytic Recovery Process of Ketones .....	28
<i>Dziaman I., Kycherenko A., Moravskiy V.</i> Investigation of the Activation Efficiency of Powdered Polyvinylchloride .....	30
<i>Kopak N., Stasevych M., Zvarych V., Nahide Gulsah D., Cigdem S., Ozyurek M., Guclu K., Vovk M., Novikov V.</i> Anthraquinonylhydrazones of $\alpha$ -Active Ketones and $\beta$ -Carbonyl-containing Compounds: Synthesis and Antioxidant Activity.....	32
<i>Skydanenko M., Kononenko M., Kurdes Yu.</i> Influence of the Vibration Source Location on the Modes of Jet Disintegration in the Priller and on Monodispersity of the Finished Product.....	34
<i>Chaykivska R., Sozanskyi M., Stadnik V., Guminilovych R., Shapoval P., Yatchyshyn Yo.</i> Investigation of HgSe Films, Deposited from Aqueous Solutions with Different Complexing Agents .....	36
<i>Korchak B., Kochubei V., Chervinskiy T., Hrynshyn O.</i> Changes in the Thermal Stability of Mineral Motor Oils After its Using in Diesel Engine .....	38
<i>Shepida M., Dobrovetska O.</i> Sedimentation of Silver and Palladium on the Silicon Surface by Galvanic Replacement .....	40
<i>Melnyk R., Serdiuk V., Shevchuk O., Tokarev V.</i> Synthesis of Microcapsules with Encapsulated Magnetic Nanoparticles for $\alpha$ -amylase Immobilization .....	42
<i>Husarova O., Shapar R.</i> The Features of Fruits Drying in the Production of Natural Chips .....	44
<i>Sachuk O., Valery Zazhigalov, Diyuk O.</i> The Mechanochemical Influence on the Physico-chemical Properties of CeO <sub>2</sub> -MoO <sub>3</sub> System .....	46
<i>Sanzhak O., Brazhnyk D., Azimov F.</i> Photodegradation of Aqueous Solution of Benzene and Phenol Using Nitrogen Doped TiO <sub>2</sub> Supported on Silica Gel.....	48
<i>Paiuk O., Volianiuk K., Sobko I., Nadashkevych Z., Mitina N., Zaichenko O.</i> Synthesis, Properties and Application of Amphiphilic Copolymers Based on Poly(fluoroalkylmethacrylate)s With Terminal Peroxide Group.....	50
<i>Nagorniak M., Oleksa V., Dron I., Stasiuk A., Maikovich O., Bordeniuk O.</i> Esterification of Dextrine by the N-derivatives of Glutamic Acid Using Steglich Reaction.....	52
<i>Hlad R., Shapoval P., Stadnik V., Sozanskyi M., Guminilovych R., Yatchyshyn Yo.</i> Influence of the Deposition Time on the Structure and Optical Properties of Indium Sulfide Films (In <sub>2</sub> S <sub>3</sub> ).....	54
<i>Tupychak M., Shyyka O., Pokhodylo N., Obushak M.</i> New Heterocyclic Scaffolds with Thiophene and 1,2,4-triazole Rings.....	56
<i>Makarova L., Pliushko O., Zhyl'tsova S., Opeida Io.</i> Effectiveness of Fenton Reagent in Oxidation Process of Methyl Violet Dye .....	58
<i>Shved M., Pyshyev S., Prysiazhnyi Yu., Sagan O.</i> Effect of Oxidant Composition on Obtaining Raw Material for Pulverized Coal Production from High-sulfuric Low Grade Coal.....	60
<i>Melnyk Yu., Tsvyk V., Skorokhoda T.</i> Diffusion-transport Properties of Hydrogel Membranes Based on Copolymers of 2-hydroxyethyl Methacrylate with Polyvinylpyrrolidone.....	62
<i>Chopyk N., Vashchuk K., Melnyk S., Skorokhoda V.</i> Composite Hydrogel Materials of Biomedical Application with Fungibactericidal Properties .....	64
<i>Demchuk Yu., Gunka V., Pyshyev S., Bratychak M., Lypko Yu.</i> Bitumen Modified by Phenol-Cresol-Formaldehyde Resins Obtained From Coking By-products.....	66
<i>Orobchuk O., Pidsadiuk M., Subtelnyi R., Dzinyak B., Fuch Y.</i> Emulsion Co-oligomerization of Hydrocarbon Fraction C <sub>9</sub> With the Polyvinylchloride Production Waste as a Dispersion Medium.....	68
<i>Masyuk A., Paraschak O., Levytskyi V.</i> Influence of the Nature of Metal-Containing Polymer-silicate Filler on the Physico-mechanical Properties of Polypropylene .....	70
<i>Katruk D., Levytskyi V.</i> The Effect of Poly (vinyl chloride) and Filler on Technological Properties of Polyester Composites.....	72

# Effectiveness of Fenton Reagent in Oxidation Process of Methyl Violet Dye

Lyubov Makarova<sup>1</sup>, Olena Pliushko<sup>1</sup>,  
Svitlana Zhyl'tsova<sup>1</sup>, Iosyp Opeida<sup>1,2</sup>

1. Department of Biochemistry and Physical Chemistry,  
Faculty of Chemistry, Vasyl' Stus Donetsk National University,  
UKRAINE, Vinnytsia, vul. 600-richchia 21, E-mail:  
makarova.l@donnu.edu.ua

2. Department of Physical Chemistry of Fossil Fuels,  
L. M. Litvinenko Institute of Physical Organic and Coal  
Chemistry, National Academy of Sciences of Ukraine,  
UKRAINE, Lviv, Naukova Str. 3a, E-mail:  
opeida\_i@yahoo.co.uk

**Abstract** – Decoloration methyl violet dye by Fenton reagent was investigated spectrophotometrically. Reagents concentration, pH value, and metal ion were varied. It was shown that substrate oxidation rate depends non-linearly on dye concentration,  $Fe^{n+}$ ,  $H_2O_2$ , and pH value. The composition of the reaction mixture providing maximum oxidation rate and MV conversion is proposed.

Keywords – kinetics, oxidation, Fenton reaction, dye, methyl violet, hydrogen peroxide, UV/Vis-spectroscopy.

## I. Introduction

Dyes are a common model in the studying of water purification processes. Oxidation of dyes is not only a methodological but also a practical interest, since it is of great importance for reducing the impact of textile, paper, food, and pharmaceutical industry wastes on the environment. Fenton reagent may be effectively used as oxidizing agent because it is environmentally friendly and rather cheap. In classical Fenton system, Fe(II) ions are used for the catalytic decomposition of hydrogen peroxide [1]. Fenton reaction produces Fe(III) and hydroxyl radical, the latter initiates radical-chain oxidation process. It is known that this process consists of number of stages, including oxidation–reduction reactions. Some authors showed that Fe(III) could also be used in Fenton-like systems instead of Fe(II) [2]. The goal of this work was investigation of regularities of oxidation reaction of organic compounds with both classical Fenton reagent and  $H_2O_2/Fe(III)$  system under different conditions using methyl violet dye as the substrate.

## II. Experimental part

Methylene violet (MV),  $FeSO_4 \cdot 7H_2O$ ,  $FeCl_3 \cdot 6H_2O$ ,  $H_2O_2$  (60 % w/v),  $H_2SO_4$ , were all G.R. grades and used as received. All solutions were made in distilled water. The pH value of the  $FeSO_4$  solution was adjusted using 0.25 M  $H_2SO_4$ .

The kinetics of dye decoloration was studied using SPEKOL® 1500 UV/Vis spectrophotometer (Analytik Jena AG, Germany) at 585 nm. Digital pH-meter was used for pH measurements. The experiments were carried out at  $21 \pm 2$  °C.

It was shown that in the range  $1.2 \cdot 10^{-5}$ – $2.7 \cdot 10^{-5}$  M, the absorbance of methyl violet solution at 585 nm versus concentration plots were linear ( $r \sim 0.99$ ) with zero intercept for both Fenton systems.

## III. Results and discussion

The kinetics of oxidative destruction of methyl violet (MV) by hydrogen peroxide in the presence of Fe(II) or Fe(III) ions was investigated. It was shown that such Fenton-like reactions could be effectively controlled by initial concentrations of dye,  $Fe^{n+}$ ,  $H_2O_2$ , and pH value of the reaction media.

Analysis of kinetic curves demonstrated that the decoloration of MV solution by Fenton reagent occurs in two stages (Figs. 1 and 2). The first stage is faster (1–2 min or less), and the second one is ~4 times slower, regardless  $Fe^{n+}$  ion used. The rate of each stage essentially depends on the concentration of the components of the reaction mixture. It has been shown that the initial rate of the oxidative destruction of dye and process duration depends on each component concentration (substrate, hydrogen peroxide,  $Fe^{n+}$ ). It should be noted that type of metal ion used is also important – change of  $Fe^{2+}$  in classical Fenton system to  $Fe^{3+}$  results in changes of kinetic curves form (see Figs. 1 and 2). The duration of the first faster stage is much shorter for  $Fe^{3+} + H_2O_2$  system.

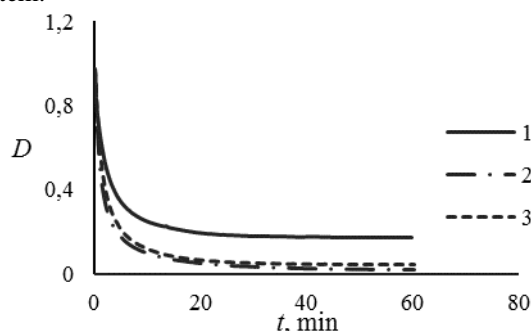


Fig. 1 Change in optical density ( $D$ ) at different  $[H_2O_2]_0$ : (1)  $1.15 \cdot 10^{-4}$  M; (2)  $2.28 \cdot 10^{-4}$  M; (3)  $4.57 \cdot 10^{-4}$  M;  $[MV]_0 = 1.75 \cdot 10^{-5}$  M;  $[Fe^{2+}]_0 = 1.0 \cdot 10^{-4}$  M; pH = 3.

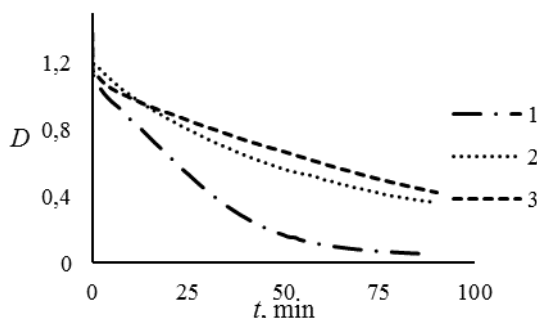


Fig. 2 Change in optical density ( $D$ ) at different  $[H_2O_2]_0$ : (1)  $0.99 \cdot 10^{-3}$  M; (2)  $2.01 \cdot 10^{-3}$  M; (3)  $3.99 \cdot 10^{-3}$  M;  $[MV]_0 = 1.75 \cdot 10^{-5}$  M;  $[Fe^{3+}]_0 = 1.0 \cdot 10^{-4}$  M; pH = 3.

To investigate the effect of hydrogen peroxide concentration on dye degradation kinetics, experiments were conducted at different  $H_2O_2$  concentrations. From Figs. 1 and 2 it can be seen that change of  $H_2O_2$  concentration significantly influences the initial rate of



dye oxidation reaction. At comparatively low concentrations of  $\text{H}_2\text{O}_2$ , relatively high initial rates are observed. When the concentration of  $\text{H}_2\text{O}_2$  increased significantly ( $0,02 \text{ M}$ ) (not given), the form of the kinetic curve has changed: the initial rate decreased, but remained almost constant throughout the measurement period. This may indicate a nonproductive decomposition (in terms of the formation of  $\bullet\text{OH}$  radicals, which are essential in the degradation of the dye) of hydrogen peroxide in such conditions. The analysis of the received dependences demonstrated that the initial rate of dye decomposition ( $V_0$ ) and substrate conversion depended non-linearly on  $\text{H}_2\text{O}_2$  concentration.  $V_0$  reached maximum values at  $[\text{H}_2\text{O}_2]_0 = 5,7 \cdot 10^{-4} \text{ M}$  for  $\text{Fe}^{2+} + \text{H}_2\text{O}_2$ , and  $[\text{H}_2\text{O}_2]_0 = 6,02 \cdot 10^{-3} \text{ M}$  for  $\text{Fe}^{3+} + \text{H}_2\text{O}_2$ .

Fig. 3 demonstrates the kinetic curves of MV decoloration by  $\text{H}_2\text{O}_2$  in the presence of  $\text{Fe}^{2+}$ . At high  $\text{Fe}^{2+}$  concentrations gradual decrease in the initial rate of dye destruction process is observed. The possible explanation is the growth of hydrogen peroxide decomposition rate, which means that the concentration of reactive  $\text{OH}^\bullet$  radicals in the system decreases:  $2\text{Fe}^{2+} + \text{H}_2\text{O}_2 + 2\text{H}^+ \rightarrow 2\text{Fe}^{3+} + 2\text{H}_2\text{O}$ . As for system  $\text{Fe}^{3+} + \text{H}_2\text{O}_2$ , larger concentrations of reagents should be taken to achieve similar results (Fig. 4).

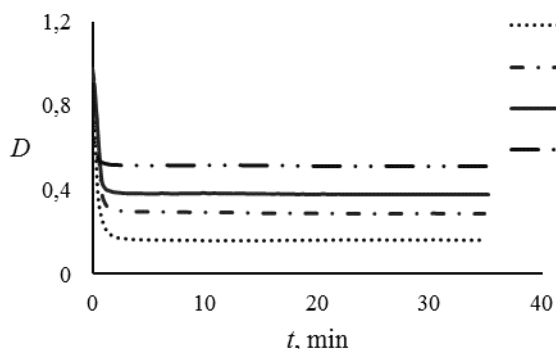


Fig. 3 Change in optical density ( $D$ ) at different  $[\text{Fe}^{2+}]_0$ : (1)  $- 0,91 \cdot 10^{-4} \text{ M}$ ; (2)  $- 1,82 \cdot 10^{-4} \text{ M}$ ; (3)  $- 22,8 \cdot 10^{-4} \text{ M}$ ; (4)  $- 27,4 \cdot 10^{-4} \text{ M}$ ;  $[\text{MV}]_0 = 1,75 \cdot 10^{-5} \text{ M}$ ;  $[\text{H}_2\text{O}_2]_0 = 4,0 \cdot 10^{-4} \text{ M}$ ;  $\text{pH} = 3$ .

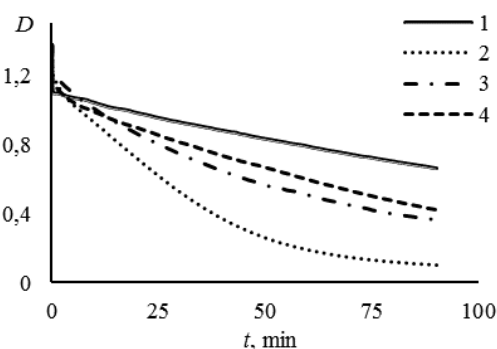
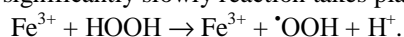


Fig. 4 Change in optical density ( $D$ ) at different  $[\text{Fe}^{3+}]_0$ : (1)  $- 1,0 \cdot 10^{-3} \text{ M}$ , (2)  $- 1,85 \cdot 10^{-3} \text{ M}$ , (3)  $- 2,0 \cdot 10^{-3} \text{ M}$ , (4)  $- 4,0 \cdot 10^{-3} \text{ M}$ ;  $[\text{H}_2\text{O}_2]_0 = 1 \cdot 10^{-3} \text{ M}$ ;  $[\text{MV}]_0 = 2 \cdot 10^{-5} \text{ M}$ ;  $\text{pH} = 3$ .

In the presence of only  $\text{Fe}^{3+}$  ions and hydrogen peroxide, significantly slowly reaction takes place:



Generated  $\bullet\text{OOH}$  radical is less active than hydroxy radicals, therefore, they react with MV much more slowly. For the investigated concentrations range initial decoloration rate shows non-linear dependence: maximum initial rate of decoloration and dye conversion was observed at  $[\text{Fe}^{3+}]_0 = 1,85 \cdot 10^{-3} \text{ M}$ .

Additional experiments were made to determine the influence of pH value on the studied process. It was established for  $\text{Fe}^{2+} + \text{H}_2\text{O}_2$  system, that maximum conversion and rate of dye decomposition was observed for  $\text{pH} 2,2-3$ .

In order to determine the optimum ratio of  $[\text{Fe}^{n+}]/[\text{H}_2\text{O}_2]$  for dye degradation, experiments were conducted by varying the  $\text{Fe}^{n+}$  and  $\text{H}_2\text{O}_2$  concentrations, keeping the dye concentration fixed. The results of the experiments are listed in Table 1.

TABLE 1  
COMPARISON OF THE INITIAL RATES OF MV OXIDATION ( $V_0$ ) BY FENTON SYSTEMS DEPENDING ON  $[\text{Fe}^{n+}]/[\text{H}_2\text{O}_2]$  RATIO AND METAL ION USED ( $[\text{MV}]_0 = 2 \cdot 10^{-5} \text{ M}$ )

$[\text{Fe}^{n+}]/[\text{H}_2\text{O}_2]$	$V_0 \cdot 10^7, \text{ mol} \cdot \text{l}^{-1} \cdot \text{min}^{-1}$	
	$\text{Fe}^{2+}$	$\text{Fe}^{3+}$
2/1	1.8	1.0
3/1	1.5	4.9
4/1	1.3	3.4
6/1	0.9	2.9

It can be seen from the Table that for  $\text{Fe}^{2+}$  as the catalyst maximum initial oxidation rate is observed at  $[\text{Fe}^{2+}]/[\text{H}_2\text{O}_2] = 2/1$ , while for  $[\text{Fe}^{3+}]/[\text{H}_2\text{O}_2] = 3/1$ . The higher the concentration of  $\text{Fe}^{n+}$  the lower the  $V_0$  value is observed.

## Conclusion

The oxidation of methylene violet dye by Fenton reagent at ambient temperature was studied spectrophotometrically. The effects of the initial concentrations of dye,  $\text{Fe}^{2+}$  ( $\text{Fe}^{3+}$ ), and  $\text{H}_2\text{O}_2$ , pH of the solution on dye decoloration reaction were established. It was shown that the initial rate of dye decoloration changes non-linearly with concentration of hydrogen peroxide and  $\text{Fe}^{n+}$ . Substitution of classical  $\text{Fe}^{2+} + \text{H}_2\text{O}_2$  system to  $\text{Fe}^{3+} + \text{H}_2\text{O}_2$  showed lower efficiency. In order to reach similar effect in dye conversion and reaction rate the concentrations of reagents should be increased approximately by order of magnitude. It was shown That the higher the concentration of  $\text{Fe}^{n+}$  in the system compared to  $\text{H}_2\text{O}_2$  the lower the dye decoloration rate was observed.

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Shkromyda M. 105  
Shpyrka I. 76  
Shvachko S. 262, 389  
Shved M. 60  
Shylo O. 202  
Shylo Ye. 202  
Shyyka O. 56  
Skoretska I. 28  
Skorokhoda T. 62  
Skorokhoda V. 64  
Skydanyenko M. 34  
Slipchuk A. 252  
Sliusar V. 144  
Sobko I. 50  
Solonets D. 312  
Soloviy Kh. 122  
Sosed A. 283  
Sotnikova A. 285  
Sozanskiy M. 36, 54  
Stadnik V. 36, 54  
Stanasyuk N. 348  
Starchevskyy V. 81  
Stasevych M. 32  
Stasiuk A. 52  
Strypa N. 350  
Subin-Kozhevnikova A. 224  
Subtelnyi R. 68  
Sukhatskii Y. 26  
Svyryda I. 146  
Sydoruk V. 76  
Tesliuk R. 362  
Tkachuk H. 232  
Tokarev V. 24, 42  
Torbych B. 258  
Tsvyk V. 62  
Tupychak M. 56  
Tur N. 331  
Turchyn Ja. 381  
Tykhonenko V. 152  
Ustyianovych T. 375  
van der Waaij H. T. 442  
van Geelkerken F.W.J. 458  
Vashchuk K. 64  
Vashkurak U. 74  
Vasylyev V. 111  
Verba V. 226  
Verkhoturova M. 389  
Vilshanetska K. 279  
Vishtak I. 265  
Vlasenko M. 180  
Volianiuk K. 50  
Vovk M. 32  
Vovk Yu. 275  
Vronska N. 144  
Werner Ł. 97  
Wierzba P. 101  
Yaculchak G. 87  
Yahodynets O. 126  
Yakubik V. 78  
Yaremko I. 317  
Yatchyshyn Yo. 36, 54  
Yavorska N. 136, 140  
Yemchenko I. 352  
Yevchuk I. 79  
Yevchuk Yu. 279  
Yurasova O. 188  
Yurkiv I. 22  
Zaichenko O. 50  
Zavaliy K. 76  
Zavuschak I. 416  
Zayats S. 78  
Zazhigalov V. 46  
Zholubak I. 420  
Zhyhailo M. 79  
Zhyl'tsova S. 58  
Zhytenko A. 267  
Zielińska A. 354  
Zin O. 26  
Zin`ko O. 140  
Znak Z. 26  
Zvarych V. 32

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*вул. Ф. Колесси, 4, Львів, 79013*

тел. +380 32 2582146, факс +380 32 2582136

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