MESOCELLULAR FOAM INCORPORATED WITH COPPER AND IRON AS CATALYST FOR AZO DYES DEGRADATION

By

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LIST OF ABBREVIATIONS

a.u.	Arbitrary unit
AAS	Atomic absorption spectrometer
AOPs	Advanced oxidation processes
ARB	Acid red B
BET	Brunauer-Emmet-Teller
BJH	Barret-Joyner-Halenda
CO_2	Carbon dioxide
COD	Chemical oxygen demand
CT	Calcination temperature
Cu	Copper
CuO	Copper oxide
Cu-FeMCF	Copper incorporated FeMCF
CuMCF	Copper incorporated MCF
E_a	Activation energy
EDX	Energy dispersive X-ray
$EO_{20}PO_{70}EO_{20}$	Polyethylene oxide-poly propylene oxide-polyethylene oxide
Fe^{2+}	Ferrous ions
Fe^{3+}	Ferric ions
$\equiv Fe^{2+}$	Immobilized ferrous ions
$\equiv Fe^{3+}$	Immobilized ferric ions
Fe_2O_3	Iron oxide (hematite)
Fe_3O_4	Iron oxide (magnetite)
Fe-CuMCF	Fe incorporated CuMCF
FeMCF	Fe incorporated MCF by direct synthesis method
Fe-MCF	Fe incorporated MCF by wet impregnation method
FeMCF0.023	Fe incorporated MCF synthesized with 0.023 g of NH_4F
FeMCF0.092	Fe incorporated MCF synthesized with 0.092 g of NH_4F
FeMCF0.184	Fe incorporated MCF synthesized with 0.184 g of NH_4F
FeMCF350	Fe incorporated MCF calcined at 350 °C
FeMCF450	Fe incorporated MCF calcined at 550 °C

FeMCF550	Fe incorporated MCF calcined at 550 °C
FeMCF600	Fe incorporated MCF calcined at 600 °C
FeMCF4	Fe incorporated MCF synthesized with 4 wt. % of Fe
FeMCF8	Fe incorporated MCF synthesized with 8 wt. % of Fe
FeMCF10	Fe incorporated MCF synthesized with 10 wt. % of Fe
FeMCF12	Fe incorporated MCF synthesized with 12 wt. % of Fe
FeMCFA1	Fe incorporated MCF synthesized by Approach 1
FeMCFA2	Fe incorporated MCF synthesized by Approach 2
FeMCFA3	Fe incorporated MCF synthesized by Approach 3
FeMCFA4	Fe incorporated MCF synthesized by Approach 4
Fe-MCFA	Fe incorporated MCF support synthesized using Set A
Fe-MCFB	Fe incorporated MCF support synthesized using Set B
Fe-MCFC	Fe incorporated MCF support synthesized using Set C
FT-IR	Fourier transformed infrared
IUPAC	International Union of Pure and Applied Chemistry
L-H	Langmuir Hinshelwood
MCF	Mesocellular foam
MCFA	MCF support synthesized using Set A
MCFB	MCF support synthesized using Set B
MCFC	MCF support synthesized using Set C
MCM-41	Mobil Composition of Matter no. 41
MS	Mesoporous silica
MSU-F-C	Mesocarbon
P123	Triblock copolymer Pluronic P123
PEO	Polyethylene
pH_{zpc}	pH zero point of charge
PL	Photoluminescence
PPO	Polypropylene
RB5	Reactive black 5
SBA-15	Santa Barbara Amorphous 15
SEM	Surface morphology microscopy
Si	Silica
SiO_2	Silica dioxide

TA	Terephthalic acid
TEM	Transmission electron microscopy
TiO_2	Titanium dioxide
TMB	1,3,5-trimethyl benzene
TOC	Total organic carbon
UV	Ultraviolet
UV-vis DRS	UV-vis diffuse reflectance spectroscopy
V	Vanadium
W	Tungsten
WO_3	Tungsten trioxide
WO_x	Oxides of tungsten
XPS	X-ray photoelectron spectroscopy
XRD	X-ray diffraction
ZSM-5	Zeolite Socony Mobil–5

LIST OF SYMBOLS

Symbol	Description	Unit
$-\frac{dC}{dt}$	Differential of concentration of organic dye with respect to time (t)	mg/L.min
°C	Degree Celsius	-
µm	Micrometer	-
•OH	Hydroxyl radicals	-
•OOH	Hydroperoxyl radicals	-
A	Arrhenius factor (pre-exponential factor)	-
cm	Centimeter	-
C_o	Initial concentration of organic dye	mg/L
C	Concentration of organic dye at time, t	mg/L
E_a	Activation energy	kJ/mol
K	Kelvin	-
k	Apparent reaction rate constant used in the Langmuir-Hinshelwood model	min^{-1}
K	Equilibrium constant	L/mg
k_{app}	Apparent reaction rate constant in the different reaction order models	(min^{-1} or L/mg.min for zero, first and second order, respectively)
n	Number	-
P	Pressure	atm
P/P_o	Relative pressure	-
R^2	Coefficient of determination	-
T	Temperature	°C
t	Time	min
θ	Diffraction angle	Degree (°)

BUSA MESO BERSEL MENGANDUNGI LOGAM KUPRUM DAN BESI SEBAGAI MANGKIN BAGI PENGURAIAN PENCELUP AZO

ABSTRAK

Penggunaan mangkin sokongan berasaskan besi (Fe) di samping hidrogen peroksida (H_2O_2) bagi proses penguraian bahan pewarna organik telah menarik perhatian disebabkan oleh kemampuan mangkin tersebut untuk mencapai kadar penguraian yang tinggi. Walau bagaimanapon, mangkin yang sedia mempunyai beberapa had yang menghalang untuk mencapai kadar penguraian yang terbaik. Untuk mengatasi kelemahan mangkin yang sedia ada, penggunaan busa meso bersel yang mengandungi logam Fe (FeMCF) sebagai mangkin telah disintesis bagi merawat bahan pewarna azo yang mempunyai strcutur yang berbeza iaitu asid merah B dan reaktif hitam 5. Perubahan dalam dos 1,3,5- trimetilbenzena (TMB) (TMB /P123 = 0.125, 0.5 dan 0.75) dan kepekatan asid hidroklorik (HCl) (2.5, 0.5 dan 1.5 M) telah membawa kepada perbezaan pembentukan sifat akhir bahan sokongan yang ketara untuk secara langsung mempengaruhi kandungan spesis Fe di dalam bahan sokongan. Mangkin busa meso bersel yang mengandungi Fe yang disintesis pada nisbah TMB/P123 0.75 dan kepekatan asid hidroklorik 1.5 M telah berjaya menggabungkan kebanyakan Fe di dalam struktur liang yang mempunyai saiz tingkap (6 nm) and saiz sel (13 nm) yang cukup besar untuk menampung Fe. Mangkin FeMCF yang disintesis dengan menggabungkan Fe semasa langkah pertama sebelum dimasukkan silika, kandungan Fe dengan peratus berat sebanyak 10 (10 wt. %), dos ammonium florida sebanyak 0.023 g dan pengolahan suhu setinggi 450 ^{0}C menunjukkan luas permukaan yang tinggi (814 m^2/g), isipadu liang yang luas

(1.808 cm^3/g), saiz tingkap (6 nm) dan sel (13 nm) yang besar, taburan Fe yang paling baik di samping kehadiran Fe yang berasingan di dalam struktur MCF dan saluran liang yang berkebolehan untuk mengatasi masalah yang berkaitan dengan pembentukan gumpalan Fe dan liang tersumbat. Secara tidak langsung, ini telah menyumbang kepada pembentukan radikal aktif bebas yang banyak dan dapat membantu dalam mendapatkan kadar peyahwarnaan (100 %) dan penguraian yang tertinggi (95 %) di samping dapat mengekalkan aktiviti dengan mempunyai kadar larut lesap yang rendah (< 0.1 mg/L). Penambahan logam kuprum terhadap mangkin FeMCF (Cu-FeMCF) menunjukkan kadar kecekapan yang tinggi (100 %) di dalam keadaan pH kurang berasid (pH 4). Mangkin Cu-FeMCF memerlukan tenaga pengaktifan yang lebih rendah (23.25 kJ/mol) berbanding mangkin FeMCF (62.57 kJ/mol). Ini adalah disebabkan oleh kesan sinergi antara Cu^{+}, Fe^{2+} dan Fe^{3+} di atas permukaan mangkin Cu-FeMCF dimana mereka dapat mengambil bahagian di dalam proses berbalik Fe^{3+} kepada Fe^{2+} yang dapat menggalakkan pembentukan radikal aktif bebas yang cukup dan berterusan dan menyumbang kearah sistem serupa-Fenton yang lebih aktif dan kemampuan kebolehgunaan semula sebanyak tujuh kali (penyahwarnaan > 95 %) didalam sistem serupa-Fenton.

MESOCELLULAR FOAM INCORPORATED WITH COPPER AND IRON AS CATALYST FOR AZO DYES DEGRADATION

ABSTRACT

The use of supported catalysts based on iron (Fe) in conjunction with hydrogen peroxide (H_2O_2) for catalytic degradation of organic dyes is attracting much attention due to their capability in achieving high rate of degradation. However, conventional Fenton-based heterogeneous catalysts still suffer from some limitations that could prevent them to achieve excellent performance in reusability study. To address the drawbacks of current types of heterogeneous Fenton-based catalyst, Fe incorporated mesocellular foam (FeMCF) catalyst have been developed to treat azo dyes with different structures namely acid red B (ARB) and reactive black 5 (RB5). Variations in the swelling agent (TMB) dosage (TMB/P123 = 0.125, 0.5 and 0.75) and hydrochloric acid (HCl) concentration (2.5, 0.5 and 1.5 M) led to significant differences in the final properties of the support materials to directly influence the distribution of Fe. Fe-MCFC catalyst developed at TMB/P123 ratio of 0.75 and 1.5 M HCl was capable of incorporating most of the Fe within the pore structure as the window (6 nm) and cell (13 nm) size were large enough to accommodate Fe particles. FeMCF catalyst developed by incorporating Fe during the first step before the subsequent addition of silica precursor, Fe concentration of 10 wt. %, ammonium fluoride dosage of 0.023 g and calcination temperature of 450 °C gave the largest surface area (814 m^2/g), pore volume (1.808), window/cell size (6/13 nm), highest Fe distribution with the presence of isolated Fe in the MCF framework and along with the pore channels that could overcome the formation of