Pengembangan fotokatalis bermagnet ($Fe_3O_4/SiO_2/TiO_2$) untuk eliminasi zat organik metilen biru dan paraquat dalam air = Development of magnetic photocatalysts ($Fe_3O_4/SiO_2/TiO_2$) for elimination of organic compounds methylene blue and paraquat in water

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Abstrak

[ABSTRAK
Fotokatalis bermagnet Fe3O4/SiO2/TiO2 telah dibuat dengan cara heteroaglomerasi. Fotokatalis bermagnet ini diterapkan dalam reaktor sistem slurry untuk eliminasi zat organik (metilen biru dan paraquat) dalam air. Dan juga, fotokatalis bermagnet ini memberi kemudahan untuk dikumpulkan kembali dengan bantuan medan magnet luar sehingga fotokatalis bekas pakai ini dapat digunakan kembali secara berulang-ulang.
Sintesis fotokatalis bermagnet Fe3O4/SiO2/TiO2 diawali dengan membuat nanopartikel Fe3O4 dengan cara presipitasi menggunakan campuran Fe(III)/Fe(II) (rasio mol 2:1) dalam larutan amonia dan kemudian dilapisi dengan SiO2 dengan cara hidrolisis ion silikat. Fe3O4/SiO2 yang terbentuk dicampurkan dengan TiO2 dengan cara hetero-aglomerasi, untuk memperoleh fotokatalis bermagnet Fe3O4/SiO2/TiO2. Dalam penelitian ini, ada dua jenis TiO2 komersil yang digunakan, yaitu nanopartikel TiO2 Aldrich dan nanopartikel TiO2 P25 Evonik. Fotokatalis bermagnet yang telah dibuat dikarakterisasi dengan berbagai teknik, di antaranya, difraksi sinar-x (XRD) untuk menentukan fasa kristal, zeta potensial meter untuk menentukan muatan permukaan partikel, VSM untuk menentukan sifat kemagnetan, spektrometer FTIR dan Raman untuk identifikasi gugus fungsi dan ikatan logam-oksida, EDS untuk menentukan komposisi unsur permukaan, TEM untuk mengetahui morfologi dan adsorpsi-N2 untuk menetukan luas permukaan. Hasil karakterisasi XRD menunjukkan bahwa TiO2-Aldrich mengandung fasa anatase 55% dan rutil 45% sedangkan TiO2-P25 Evonik mengandung fasa anatase 86% dan rutil 14%. Fasa magnetit (Fe3O4), TiO2 anatase dan rutil dipertahankan dalam fotokatalis bermagnet yang terbentuk. Dalam tahapan pembentukan bahan, ke dua jenis TiO2 telah berhasil menempel secara permanen pada Fe3O4/SiO2 melalui interaksi elektrostatik gugus hidroksil permukaan masing-masing oksida. Spektromentri FTIR mengamati ikatan Si-O-Ti yang terbentuk, hasil interaksi elektrostatik TiO2 dengan Fe3O4/SiO2, selanjutnya dinotasikan sebagai Fe3O4/SiO2/TiO2-Ald dan Fe3O4/SiO2/TiO2-P25. Kedua jenis bahan mempunyai sedikit perbedaan sifat fisika-kimia dan morfologi. Semakin banyak fraksi TiO2 dalam bahan maka luas permukaannya semakin turun. Fotokatalis bermagnet yang diperoleh mempunyai sifat magnet cukup baik dan nilai remanent magnetization dan coercivity yang rendah, artinya bahan ini dapat

dengan mudah dikumpulkan kembali dari cairan dengan bantuan medan magnet
luar dan tanpa medan magnet dapat terdispersi kembali dengan baik dalam air. Fe3O4/TiO2-Ald dan Fe3O4/TiO2-P25 yang telah dibuat mampu mengeliminasi
metilen biru dan paraquat dalam air melalui proses fotokatalitik, namun demikian aktivitasnya dianggap masih rendah. Diduga terjadi pelemahan aktivitas TiO2 yang ada dalam fotokatalis bermagnet akibat dari efek fotodisolusi. Adanya barrier SiO2 di antara Fe3O4 dan TiO2, metilen biru yang terdegradasi meningkat dari 44,2% menjadi 52,3% oleh Fe3O4/SiO2/TiO2-Ald dan paraquat yang terdegradasi meningkat dari 16,3% menjadi 45,8% oleh Fe3O4/SiO2/TiO2-P25. Aktivitas kedua fotokatalis bermagnet yang punya barrier SiO2 ini setara dengan aktivitas rasio fraksi aktif TiO2 terhadap TiO2 murninya. Tidak ada kehilangan aktivitas TiO2 dalam fotokatalis bermagnet setelah diberi barrier SiO2. Adanya SiO2 dalam fotokatalis bermagnet ini juga mampu mengeliminasi senyawa metilen biru dan paraquat melalui proses adsorpsi. Maka dari itu, lebih banyak lagi metilen biru dan paraquat tereliminasi melalui kedua proses. Secara total, Fe3O4/SiO2/TiO2-Ald mampu mengeliminasi metilen biru 87,3% dan paraquat 71,5%. Sedangkan Fe3O4/SiO2/TiO2-P25 mampu mengeliminasi paraquat 82,6%. Kapasitas adsorpsi Fe3O4/SiO2/TiO2-Ald lebih rendah dari kapasitas adsorpsi Fe3O4/SiO2/TiO2-P25, tetapi aktivitas fotokatalitik Fe3O4/SiO2/TiO2-Ald lebih tinggi dari aktivitas fotokatalitik Fe3O4/SiO2/TiO2-P25. Fotokatalis bermagnet yang dikembangkan menunjukkan kestabilan fotokatalitik paling tidak sampai empat kali pemakaian berulang dan juga masih dapat dikumpulkan kembali dengan mudah dengan bantuan medan magnet luar
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ABSTRAK
Magnetic photocatalysts of Fe3O4/SiO2/TiO2 have been prepared using heteroagglomeration method. The magnetic photocatalysts were applied in slurry reactor
system for elimination of organic compounds (methylene blue and paraquat) in water. In addition, the magnetic photocatalysts are able to be recollected easily with the assistance of an external magnetic field so that the spent composite can be used repeatedly.

Synthesis of magnetic photocatalysts of Fe3O4/SiO2/TiO2 were preceded by preparing Fe3O4 nanoparticles through precipitation method using mixture of Fe(III)/Fe(II) (2:1 mole ratio) in ammonia solution and further coating with SiO2 through hydrolysis of silicate ion. The formed Fe3O4/SiO2 were mixed with TiO2 in hetero-agglomeration manner, to obtain Fe3O4/SiO2/TiO2 magnetic photocatalysts. In this study, two type of commercial TiO2 nanoparticles were used, namely; TiO2 Aldrich and TiO2-P25 Evonik.

The prepared magnetic photocatalysts were characterized with various techniques, i.e, x-ray diffraction (XRD) to determine the crystal phase, zeta potensial meter to determine the surface charge of particles, VSM to determine the magnetic properties, FTIR and Raman spectrometer to identify the functional groups and

metal-oxide bond, EDS to determine the surface chemical composition, TEM for morphological examination and N2-adsorption to determine surface area. The results of XRD characterization showed that TiO2-Aldrich contains 55% of anatase and 45% of rutile, while TiO2-P25 Evonik contains 86% of anatase and 14% of rutile. Magnetite (Fe3O4), anatase and rutile phase of TiO2 were retained in the formed composites. In the stage of composite formation, both TiO2 types have been successfully attached to Fe3O4/SiO2 via electrostatic interaction of surface hidroxyl group of oxides. FTIR spectrometry analysis revealed the formed Si-O-Ti bond, resulting of electrostatic interaction both TiO2 and Fe3O4/SiO2. Hence, they were denoted as Fe3O4/SiO2/TiO2-Ald dan Fe3O4/SiO2/TiO2-P25. The both magnetic photocatalysts have a slight different physico-chemical properties and morphology. The more the fraction of TiO2 in magnetic photocatalysts, the lower its surface area. The obtained magnetic photocatalysts have high saturation magnetization and low coercivity and remanent magnetization value. It means that the magnetic photocatalysts can be still recollected from water with assistance of external magnetic field. In a non-magnetic field, magnetic photocatalyst can be well dispersed in water again.

The formed Fe3O4/TiO2-Ald and Fe3O4/TiO2-P25 was able to eleminate methylene blue and paraquat in water by photocatalytic process, nevertheless itsactivities was low. Allegedly it occurs weakening of TiO2 activity in magnetic photocatalyst caused by photodissolution effects. The presence of SiO2 barrier between Fe3O4 and TiO2, the degraded methylene blue was increased from 44.2% to 52.3% by Fe3O4/SiO2/TiO2-Ald and the degraded paraquat was increased from 16.3% to 45.8% by Fe3O4/SiO2/TiO2-P25. The photocatalytic activity of the both magnetic photocatalysts which having SiO2 barrier was equivalent to the activity of fraction ratio of TiO2 to its pure TiO2. There was no loss of photocatalytic activity of TiO2 in magnetic photocatalysts after SiO2 barrier being introduced. The presence of SiO2 on magnetic photocatalysts was also able to eliminate methylene blue and paraquat compounds through adsorption process. Therefore, more methylene blue and paraquat eliminated via both process. Totally, the Fe3O4/SiO2/TiO2-Ald was able to eliminate 87.3% of methylene blue and 71.5% of paraquat. Meanwhile Fe3O4/SiO2/TiO2-P25 was able to eliminate 82.6% of paraquat. Adsorption capacity of Fe3O4/SiO2/TiO2-Ald was lower than that of Fe3O4/SiO2/TiO2-P25, but photocatalytic activity of Fe3O4/SiO2/TiO2-Ald was higher than that of Fe3O4/SiO2/TiO2-P25. The developed magnetic photocatalysts show its activity photocatalytic stability and still can be well magnetically separated after being repeatedly used for four times.;Magnetic photocatalysts of Fe3O4/SiO2/TiO2 have been prepared using heteroagglomeration method. The magnetic photocatalysts were applied in slurry reactor system for elimination of organic compounds (methylene blue and paraquat) in water. In addition, the magnetic photocatalysts are able to be recollected easily with the assistance of an external magnetic field so that the spent composite can

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