

Highly Active Delaminated Ti-MWW for Clean Production of Epoxides

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The use of titanosilicate / H₂O₂ system in place of the hazardous organic oxidants is expected to lead to the greener process. In this respect, a new titanosilicate, named del-Ti-MWW, has been prepared in order to develop an environmentally friendly catalyst suitable for bulky alkenes. Del-Ti-MWW has a greatly enlarged surface area and open reaction space, which can reduce the restrictions on the diffusion of bulky molecules. The delaminated sample also exhibited good performance for small alkene molecules as a result of the retention of zeolitic structure.

Catalytic epoxidation of alkenes using hydrogen peroxide as a terminal oxidant

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Epoxides are highly useful intermediates for the manufacture of a range of important commercial products. The use of stoichiometric amounts of peracids for direct oxidation of alkenes is the major procedure for the synthesis of epoxides in the fine chemicals industry. However, the use of peracids is not environmentally benign as it produces equivalent amounts of acid waste and additionally there are associated safety handling issues.

Environmentally Benign Catalytic Oxidation Using Apatite Disperse Phase

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Apatite disperse phase containing a catalytic amount of polyoxometalates with solid urea-hydrogen peroxide complex (urea-H₂O₂) is effective reaction system for solvent-free oxidation of organic compounds. Our solid phase system is convenient and green process because of solvent-free catalytic reaction without special apparatus under mild conditions, the use of a harmless solid disperse phase and reuse of solid catalyst phase.

Synthesis of Dimethyl Carbonate as a Green Chemical by Vapor-Phase Oxidative Carbonylation of Methanol

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To develop a vapor-phase dimethyl carbonate synthesis process, effects of reaction conditions and gas additives on reaction performance have been investigated. Based on experimental and simulation results, a fluidized bed reactor has been selected, considering the temperature control and continuous catalyst regeneration.

A2-05

Liquid Phase Oxidation of p-Xylene on SBA-15 immobilized Transition Metal Ions

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Transition metal ions immobilized on SBA-15 were tested for the selective oxidation of p-xylene in a 250 ml a pressurized semi-batch reactor, in which the consumed oxygen can be successively supplied, with and without the addition of bromine. We studied the effects of the number of organic functional groups on the surface of SBA-15 and the type of transition metal ions (Co, Mn, Ni, Cu and Fe), and the addition of bromine. The conversion and the selectivity to organic acids (p-toluic acid and terephthalic acid) on a Co/SBA-15 catalyst were higher than those on the other M/SBA-15 [M=Mn, Ni, Cu, and Fe]. The addition of bromine in heterogeneous catalytic system also greatly improved the conversion and the selectivity to organic acids.

A2-06

SBA-15 immobilized Cobalt Catalyst for Oxidation of Ethylbenzene to Acetophenone

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Cobalt() complex immobilized on a mesoporous material SBA-15 was tested for the selective oxidation of ethylbenzene without any solvent in a 250 ml a pressurized semi-batch reactor, in which the consumed oxygen can be successively supplied. The selectivity to acetophenone at a typical condition of reaction temperature of 130 °C, partial pressure of O₂ of 2 atm, total pressure of 10 atm, and reaction time of 3 h was high as ca. 75%, even though the conversion at the condition was comparatively low as ca. 21%. The deactivated catalyst was easily recovered by simple drying. The conversion and selectivity to acetophenone in the oxidation on the reactivated catalyst returned to very similar levels with the original values.

A2-07

Characteristics of Spherical NiO/Al₂O₃ Medium Materials Prepared by A Novel Granulation Method for Chemical-Looping Combustion

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Chemical-looping combustion (CLC) has been focused as a very promising combustion technology for high energy utilization with easy separation of CO₂ and no formation of NO_x. The key of this combustion technology is to develop medium materials with high oxygen capacity, and high mechanical and thermal stability. We developed a novel oil-drop granulation method for the preparation of spherical NiO/Al₂O₃ particles with a uniform particle size of ca. 2mm. In cyclic tests of the reduction with H₂ and the oxidation with air, it is found that NiO/Al₂O₃ particles with the weight ratios between 2/3 and 4/1 show high reaction rate, regenerability, and mechanical and thermal stability. The reaction characteristics are also discussed in connection with the physical and chemical properties of these particles.

A2-08

Silica Gel/*N*-Oxyl/Aq. NaOCl System for Oxidation of Alcohols

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An environmentally benign silica gel/*N*-oxyl/aq. NaOCl oxidation system was developed and applied to the oxidation of alcohols without using any organic solvent. A totally closed recycle system was achieved by immobilization of *N*-oxyl on silica gel and electro-regeneration of NaOCl.

A2-09

Catalytic deNO_x Process Using Hydrogen as a Reducing Agent

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A new catalytic lean-deNO_x process using hydrogen as a reducing agent has been studied over platinum catalyst supported on various metal oxide support materials. The advantage of the process is very low reaction temperatures (<100°C), but considerable emission of N₂O is a problem for the practical applications. Our study is directed towards the development of efficient catalyst systems for the title reaction.

Greener Synthetic Routes to Carbonyl Compounds and Imines: Au(I) Catalyzed Highly Efficient Hydration and Intermolecular Hydroamination of Alkynes

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Hydration of alkynes was efficiently catalyzed by 0.005-1 mol% of $(\text{Ph}_3\text{P})\text{AuCH}_3$ -acid to afford a wide variety of carbonyl compounds in aqueous methanol. (II) Intermolecular hydroamination of alkynes catalyzed by 0.01-1 mol% of $(\text{Ph}_3\text{P})\text{AuCH}_3$ -additive proceeded to afford ketimines in good yields under solvent free conditions.

Environmentally Benign Process for Aromatic Polycarbonate Synthesis by Oxidative Carbonylation Catalyzed by Pd-carbene Complexes

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A direct oxidative carbonylation of Bisphenol A to polycarbonate catalyzed by Pd-carbene complex systems has been developed. This catalyst systems gave much higher molecular weights and yields than conventional Pd catalyst systems. Highest molecular weight ($M_w=29,400$) and yield (77%) were obtained using the $\text{PdBr}_2(\text{c}1\text{-tBu})$ system.

**Transition Metal-Catalyzed Oxidation of Organic Substrates
—O₂ or Air as Oxidant, Recyclable Catalyst—**

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Transition metal-catalyzed oxidation of organic substrates, such as alcohols and amines, using an atmospheric pressure of oxygen or air as oxidant is presented. The successful application of the homogeneous catalyst system to the catalyst recyclable system is also referred from the viewpoint of green and sustainable chemistry.

A Highly Effective Pd- (t-Bu)₃ Catalyst for the Synthesis of Triarylaminines

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A Pd/P(*t*-Bu)₃ catalyst system has revealed very high activity and selectivity for the synthesis of triarylaminines. This system allows the use of unactivated aryl bromides, aryl chloride. This catalyst can be useful for the synthesis of the new hole transport materials of OLED.

Green Baeyer-Villiger Oxidation with Hydrogen Peroxide : Sn[N(SO₂C₈F₁₇)₂]₄ as a Highly Selective Lewis Acid Catalyst in a Fluorous Recyclable Phase

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Tin(IV) bis(perfluoroalkanesulfonyl)amide complex catalyst for Baeyer-Villiger (BV) oxidations of cyclic ketone by 35% H₂O₂ was shown to give an excellent yield and selectivity in a fluorous biphasic system. Furthermore, the catalyst was completely recovered and reused in the recyclable fluorous immobilized phase without loss of its catalytic activity.

New Heterogeneous Olefin Metathesis Catalyst: Mesoporous Alumina-supported Rhenium Oxide

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Olefin metathesis is a unique carbon skeletal rearrangement between two olefins catalyzed by metal carbene complexes. With the recent development of homogeneous catalysts, this reaction has emerged as a powerful tool for the formation of C-C bonds in modern organic synthesis. We were interested in heterogeneous catalysis for olefin metathesis and succeeded in developing a new heterogeneous catalyst of rhenium oxide on mesoporous alumina.

A2-16

**Reactivity Study of NO_x-free Chemical-Looping Combustion
with Double Metal Oxide**

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The single and double metal oxide were prepared to gain high reactivity and regenerability for chemical-looping combustion (CLC). In both reduction and oxidation, the reaction rates of double metal oxides were slightly lower than the single metal oxides. Fe₂O₃ is relatively less expensive than NiO and CoO. Thus double metal oxide of NiO-Fe₂O₃ will be a substitute of single metal oxide NiO.

A2-17

**Effect of Various Kinds of Binders in Oxygen Carrier
for Chemical-Looping Combustion**

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For combustion with CO₂ capture, chemical-looping combustion (CLC) has the advantage that no energy is lost for the separation of CO₂. Oxygen carrier consists of metal oxide and binder. NiO particle is used as metal oxide. Oxygen carriers are prepared by dissolution method. The feasibility of using various kinds of binders has been investigated in a thermogravimetric reactor (TGR).

A2-18

CATIONIC POLYMERIZATION WITH SUPPORTED BF₃

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Hydrocarbon resins are produced from the acid catalysed (typically AlCl₃ or BF₃) cationic polymerization of a complex mix of monomers derived from petroleum plant cracking fractions. Resins produced have found applications in adhesives, rubbers, and paints. However, the necessity to quench the polymerized feed after

reaction, generates vast quantities of aqueous acidic waste and is of major concern for the industry. The use of supported catalytic systems, which offer ease of separation, reuse and avoid the need to quench, have been proposed as a way of eliminating this waste. We wish to report the first use of silica supported BF_3 catalyst with methanol co-catalyst in the polymerization of aromatic alkenes and mixed C9 petroleum feed.

A2-19

INVOLVEMENT OF SOLID ACID OF HETEROATOM-DOPED POROUS SILICA IN THE DIELS-ALDER REACTION OF ISOPRENE WITH METHYL ACRYLATE

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Based on physico-chemical characterizations, it is clearly demonstrated that weak solid Lewis acid sites are effective for the Diels-Alder reaction of isoprene with methyl acrylate.

A2-20

Oxidation of Nitrotoluenes with Air by *N*-Hydroxyphthalimide Analogues as key Catalysts

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The oxidation of nitrotoluenes with air to nitrobenzoic acids was successfully achieved by the use of *N*-hydroxyphthalimide analogues as key catalysts. Thus, *p*- and *m*-nitro- toluenes were oxidized under 10 atm of air in the presence of *N*-acetoxyphthalimide combined with $\text{Co}(\text{OAc})_2$ and $\text{Mn}(\text{OAc})_2$ at 130 °C to afford *p*- and *m*-nitrobenzoic acids in 81% and 92% yields, respectively. *o*-Nitrotoluene was converted into *o*-nitro- benzoic acid in 51% yield by the aid of NO_2 .

A2-21

Improved Vapor-Phase Propene Epoxidation over Silylated Au-Titanosilicate Catalysts

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Enhanced propene epoxidation using H_2 and O_2 , over Au nanoparticles dispersed on silylated 3-D wormhole-like mesoporous titanosilicates prepared by modified sol-gel technique is reported. Silylation was found to have marked influence in preparing efficient and regenerable catalysts for epoxidation due to

hydrophobicity effect.

A2-22

**Studies on Selective Adsorbents for Oxo-anions.
Nitrate Ion-sieve Property of Layered Double Hydroxides**

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Layered double hydroxide (LDH) consists of a group of anion exchanger with exchangeable interlayer anions. They are attractive in the field of green sustainable chemistry, since they can be applied to the removal of harmful oxo-anions (phosphate, nitrate, etc.) from aqueous solutions. In the present study, we prepared chloride-type LDHs consisted different metal ions Li^+ , Mg^{2+} , Al^{3+} , Fe^{3+} , Co^{2+} or Ni^{2+} in the brucite layers.

A2-23

**Environmentally Benign Removal of Pollutant Oxyanions by Fe Adsorption Center in
Functionalized Mesoporous Silica**

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We have synthesized the specific molecular adsorption sites on the pore wall surface in MCM-41 by means of fixation of ethylenediamine group followed by cationization with Fe^{3+} . The iron-anchored surface sites result in fast and selective adsorptions of pollutant oxyanions, such as arsenate, chromate, selenate and molybdate.

A2-24

Selective Oxidation of Alkane with Carbon Dioxide Using Diamond Catalyst

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Oxidation of methane and ethane with CO_2 was conducted with Vanadium oxide loaded diamond or SiO_2

catalyst. Silica for methane and diamond for ethane afforded excellent activity in the conversion of respective alkane to corresponding aldehydes.

A2-25

Oyster Shell as a Novel Green Material for Gas Clean Up

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In order to develop an efficient absorbent of SO₂ in coal combustion, coal char-supported oyster shell was proposed. The absorption behavior of the absorbent for SO₂ was examined using a fixed bed reactor at 900 °C. In all the absorption runs, no SO₂ leakages in the tail gas were observed before the breakthrough point (less than 1 ppm). The utilization efficiency of commercial CaO was 27.1%. When Oyster shell/YL char was used in the absorption of SO₂, the utilization efficiencies of calcium increased to 85.1%.

A2-26

A Green Process for The Production of Carbon Nanotubes in Organic Liquid

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Well-aligned carbon nanotubes arrays were grown on silicon substrates by using a novel catalytic method in organic liquid such as alcohols. Un-equilibrium catalytic deposition gave significantly pure carbon nanotubes with very little soot within a few minutes in methanol and/or ethanol. Scanning electron microscope images indicate that the nanotubes arrays are grown perpendicular to the silicon substrate surface with a significantly high density like a flower arrangement. This very fast and dense production of carbon nanotubes can be attributed to the great difference of chemical potential between the substrate surface and reactant liquid.

A2-27

Direct methane conversion to ethane and hydrogen at room temperature over SiO₂-Al₂O₃-TiO₂ photocatalysts

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Methane can be directly converted into ethane and hydrogen over novel SiO₂-Al₂O₃-TiO₂ photocatalytic system at room temperature. The active sites on the catalyst was suggested to be synergistic pair sites of Ti and Al in silica matrix.

A2-28

Selective Oxidation of Ethylene-Glycol by Active Carbon Catalyst

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Selective oxidation of ethylene glycol was examined with a batchwise reaction system and metal supported active carbon catalysts under pressurized conditions. Main products by oxidation were glycol aldehyde and glycolic acid, and these yields were maximized with a catalyst whose metal loading ratio was Pb:Pt=3:7.

A2-29

Selective Isomerization of m-Xylene using a Catalytic Zeolite Membrane

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It is a very fact that p-xylene as the feedstock of polyethyleneterephthalate (PET) is industrially important and therefore is desired to be produced more than m- and o-xylene. Therefore, we demonstrate in this study that a selective isomerization of m-xylene to p-xylene using a ZSM-5 membrane synthesized on a porous alumina support becomes possible.