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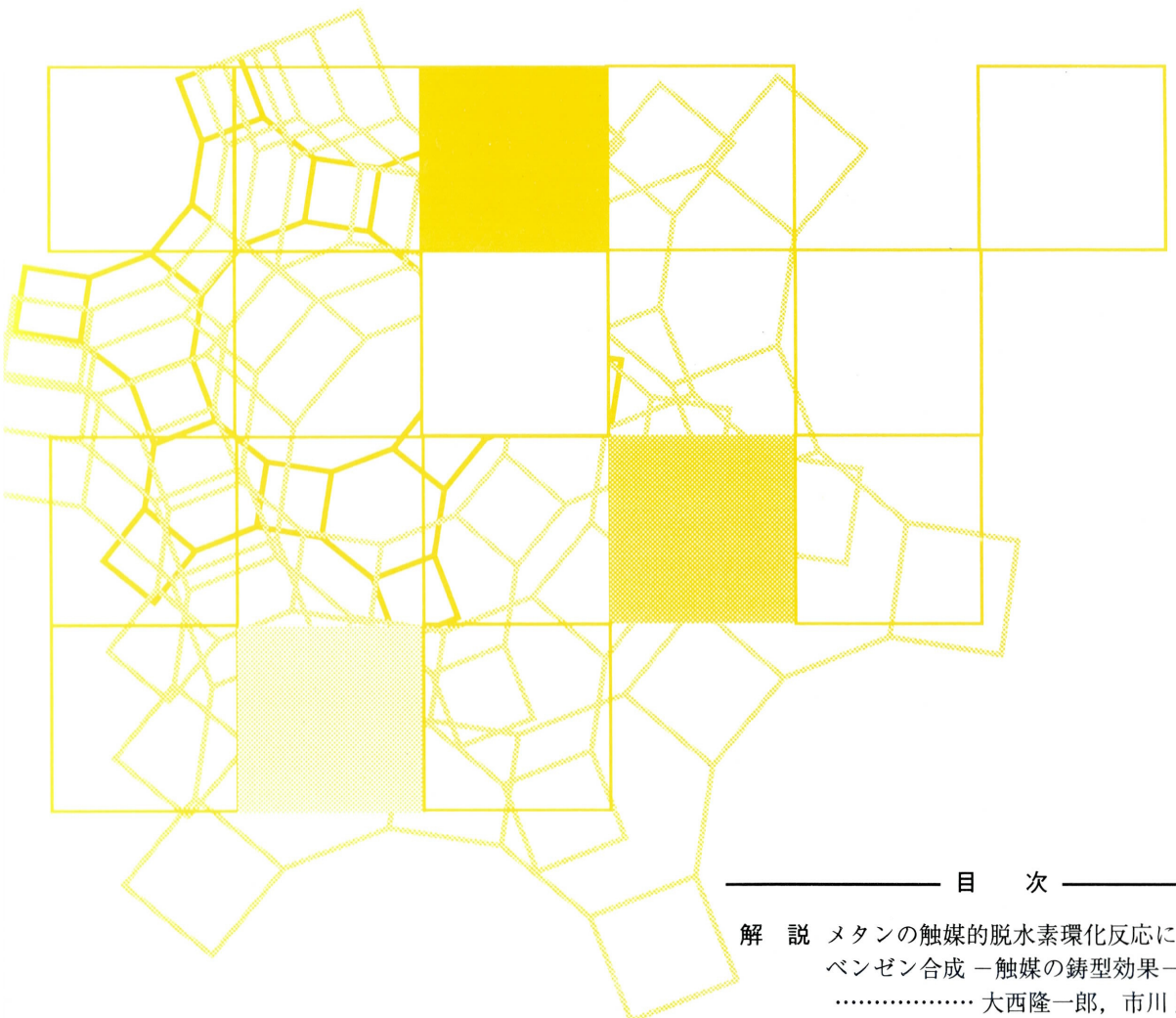

目　次

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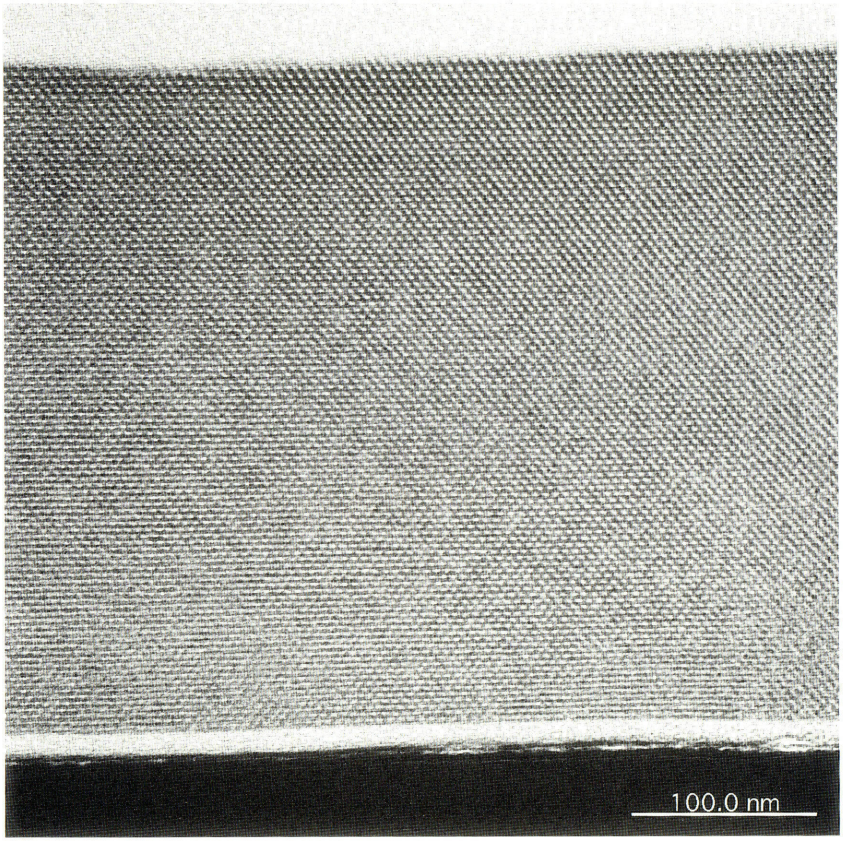

配向性メソポーラスシリカ薄膜
（提供：キヤノン株式会社 中央研究所　渡邊壮俊，宮田浩克）

《解 説》

メタンの触媒的脱水素環化反応によるベンゼン合成 —触媒の鋳型効果—

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メタンから芳香族化合物と水素を合成する脱水素環化反応について概説する。その中でまず，多くの触媒探索の結果得られた本反応を活性及び選択率高く促進する中心金属種とゼオライト担体が持つべき要件を記述する。次いで，メタンフィード中に少量の炭酸ガスや一酸化炭素を添加，或いは全圧を2～3気圧にすることで，触媒を不活性化する第一の要因である触媒表面への炭素蓄積を抑制し長時間安定な活性を維持できることを明らかにする。さらに，メタンから芳香族化合物を生成するルートと活性な触媒種の生成過程につい述べる。

1. はじめに

石油は，高分子樹脂や医農業・食品などの化学原料として，また熱源或いはエネルギー源として我々の生活になくてはならない資源である。しかし，その埋蔵量の推算から，ここ半世紀内に掘り尽されると言われている。そこで，石油を代替する炭化水素資源として天然ガスが考えられる。メタンを主成分とする天然ガスは，含有する硫黄分や窒素分が極めて少なく化石燃料中一番クリーンなエネルギー源であり，可採年数も石油の1.7倍あるとされている。さらに，糞尿，ゴミ，材木などの腐敗発酵によって発生するメタンはリサイクル可能な炭素資源である。また，近年見いだされたメタン水和物（メタンハイドレート）は，世界各地の深さ数百メートルの海底に存在することが確認され，推算であるが数百年の可採年数があるとの報告もある。以上の観点から，メタンを原料とした未来産業の展開を図1に示す。しかし，安定なメタンを有用な化合物に転換するのは至難であった。ここでは，この安定なメタンからベンゼンと水素への触媒的直接合成プロセスについて筆者らの仕事を中心に紹介する。

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2. メタンの炭化水素資源への転換反応

メタンの活性化には，439 kJ/molという大きな結合エネルギーをもつC-H結合を解離する必要があり，メタンを直接有用な炭素資源に転換するには大変厳しい反応条件が必要となる。そこで，より容易な反応条件でメタンを有用な炭素資源に転換するため，酸素との反応について精力的に研究された。その例として，methane coupling反応（$CH_4 + 1/2O_2 = 1/2C_2H_4 + H_2O$，或いは$CH_4 + 1/4O_2 = 1/2C_2H_6 + 1/2H_2O$）によるエタンやエチレン，また酸素含有生成物（HCHOやHCOOH）の生成反応がある。しかし，反応条件及び触媒開発の多様な改良努力にもかかわらず，多量の炭酸ガスや一酸化炭素の生成によって，目的生成物の収率を20 %以上にすることが出来ず，工業化の水準に達する成果は得られていない[1]。

2.1 メタンからベンゼンと水素の直接合成

表1に各種アルカンの各種反応に対する熱力学的な計算結果を示す[2]。同表には，メタン（C_1），エタン（C_2），プロパン（C_3），ブタン（C_4），ヘキサン（C_6）の脱水素反応，ベンゼンの生成反応，部分分解反応，完全分解反応のΔG_fが0になる温度（$T_{\Delta G_f=0}$）を計算した結果を示す。表から$T_{\Delta G_f=0}$が，完全分解<部分分解<芳香族化<脱水素の順に高くなることが分かる。即ち，炭素を生成する完全分解反応が，ベン

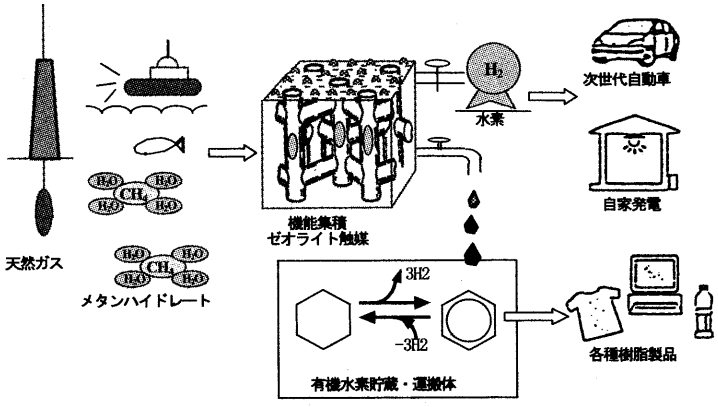


図1 メタンからベンゼンと水素の触媒的合成システムと産業展開の可能性

表1 各種n-アルカンの各種反応のΔG_fが0になる温度，K

アルカン	メタン	エタン	プロパン	ブタン	ヘキサン
脱水素[a]	−	＜1100	930	930	−
Bz生成[b]	＜1200	850	720	740	650
部分分解[c]	−	−	600	500	−
完全分解[d]	820	380	360	340	＞300

a) $C_nH_{2n+2}=H_2+C_nH_{2n}$
b) ベンゼン生成反応
c) $C_nH_{2n+2}=CH_4+C_{n-1}H_{2n-2}$
d) $C_nH_{2n+2}=nC+(n+1)H_2$

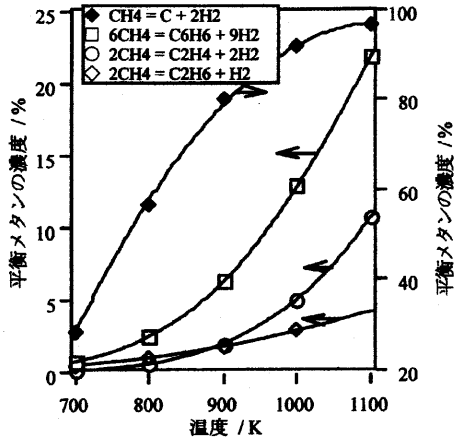


図2 各種メタン関連反応における平衡メタン濃度

ゼンを生成する反応より，容易に起こる反応であり，これを如何に抑制するかが安定で高選択的に芳香族化合物を得るための鍵となる。また，この表から，アルカンの炭素鎖が長くなるに従って，反応が起こり易くなることも解る。

炭素数が6以上のアルカンを環化脱水素してBTXなどの芳香族化合物を生成する反応は，比較的容易であるためか50年以上前に発見された。それでも，芳香族化合物を生成する反応条件では，重合，異性化，クラッキング，水素化分解など多様な副反応が起こる。これらの反応を抑制し高収率で目的化合物を得るために，担体の酸点を中和し，中心金属であるPtの小粒子化を図ったL-ゼオライト担持Pt触媒(Pt/BaK-L或いはPt/K-L)が調製され，n-ヘキサンから70％以上の収率で芳香族化合物を生成することに成功した[1]。一方，炭素数が5以下(C_{5-})のアルカンから芳香族化合物を得るには炭素数6以上(C_{6+})の場合とは違い，より高い反応温度とC_{5-}のアルカンを増炭してC_6以上の炭素鎖を持つ中間生成物に転化する必要がある。そこで，触媒に酸性質の付与が必要となる。しかし，触媒の酸性質は，重合，異性化，クラッキングなどを引き起こすため，芳香属化合物の生成選択率が落ちると同時に触媒表面への炭素蓄積による活性の低下が激しくなる。この問題を解決の方向に導いたのが，ZSM-5ゼオライト担体である。BP/UOP社やMobil社が開発したHZSM-5にGa，ZnやAgを担持した触媒は，ブタンからベンゼン，トルエン，キシレン（BTX）を65～75％の高収率，長寿命で生成すると報告されている[1]。

メタン関連反応の，平衡メタン濃度を計算し図2に示す。図から，平衡では，エチレン生成<エタン

表2 ZSM-5($SiO_2/Al_2O_3=50$)触媒によるメタンからベンゼン生成反応[a)]

触媒	メタン転化率	ベンゼン選択率
HZSM-5	1.0	100
2%Mo/HZSM-5	7.2	100
2%Mo/NaZSM-5	0	0
MoO_3	0	0

a) 973 K, 0.2 MPa, メタンF/W = 1440 ml/g/h

生成<ベンゼン生成<完全分解（炭素生成）の順に反応が容易になることがわかる。また，メタンからベンゼンへ反応は高温ほど高い平衡メタン濃度を示し，1000 Kで13 %，1100 Kで22 %と求められる。従って，1000 K以上の高温で，メタンの完全分解を引き起こさない触媒系が探索された。その結果，1989年に乾ら[3)]によるPtでイオン交換したH-gallosilicateが，また1993年にWangら[4)]による，HZSM-5に担持したMo触媒が高い選択率で連続的にベンゼンを生成することを見いだした。Wangらの結果の一部を表2に示す。表から，HZSM-5担体あるいはMoO_3単独で殆ど進行しない反応が，それらを組み合わせた触媒では促進され，7.2 %のメタン転化率，100 %の生成選択率でベンゼンを生成した。しかし，酸性の無いNaZSM-5担体にMoO_3を担持した触媒では，全く反応性が無いことから，本反応は酸触媒反応であることがわかる。その後，多くの研究者によって追試，触媒および反応条件の改良がなされた。

2.2 活性な中心金属と担体との組み合わせ

我々は，触媒の酸性質と反応活性との関連についてより定量的に検討した。即ち，SiO_2/Al_2O_3比の異なるHZSM-5にMoを3 wt%担持した触媒を調製し，そのピリジン吸着のIR吸収強度から測定した酸性質とベンゼンの生成速度との関連について調べた[5)]。結果を図3に示す。図から，ベンゼンの生成速度とMo/HZSM-5触媒のプロトン酸（B-acid）量との間に良い相関関係があるが，ルイス酸量とは関連が無い。即ち，メタンの芳香族化反応を引き起こすためにはゼオライト担体のプロトン酸が必要であると理解できる。

酸性質以外に担体が持つべき条件について，我々

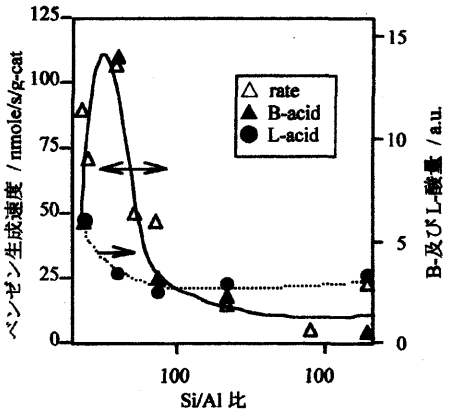


図3 Mo/HZSM-5の酸性質と反応活性（973 K, 1 atm, メタンF/W = 1440 ml/g/h）

表3 各種ゼオライトに担持したMoとRe触媒[a)]

ゼオライト	Mo		Re		細孔径, Å
	反応率	速度	反応率	速度	
ZSM-5	9.5	1.24	10.1	1.63	5.1-5.6
ZRP-1	8.9	0.95	–	–	5.1-5.6
MCM-22	8.0	0.55	5.3	0.61	4.0-5.5
ZSM-11	6.3	0.57	5.1	0.35	5.3-5.4
Beta	4.1	0.14	4.6	0.11	5.5-7.6
ZSM-12	4.3	0.11	2.9	0.10	5.5-5.9
FER	5.2	0.12	3.9	0.08	3.5-5.4
L	0.4	0.05	4.3	0.03	7.1
SAPO-5	4.3	0.05	3.1	0.01	7.3

a) 993 K, メタンF/W = 3000 ml/g/h, 3気圧, 2%CO_2;
反応率=メタン反応率,
速度=炭素数で数えたベンゼン生成速度 (μmole/g/s)

はシリカ／アルミナ比が36～39（但し，ZSM-12とFERでは約60）の各種ゼオライト担体にMoを担持した触媒を調製し，メタンの芳香族化反応に使用した。結果を表3に示す。表からメタンの芳香族化反応に対し，5.3～5.5 Åの狭い範囲の細孔径を持つZSM-5, MCM-22, ZSM-11およびZRP-1（希土類元素を含むMFI型）[6)]が特異的に高い活性を示すことが分かる。これら細孔の中では，大きな分子（例えば，coke前駆体重合物）の成長を物理的に妨げ活性点上への炭素生成を抑制するが，ほぼ同じ大きさの芳香族化合物の生成は可能にする鋳型効果が働く。そのため，5.3～5.5 Åの細孔を持つゼオライト担体

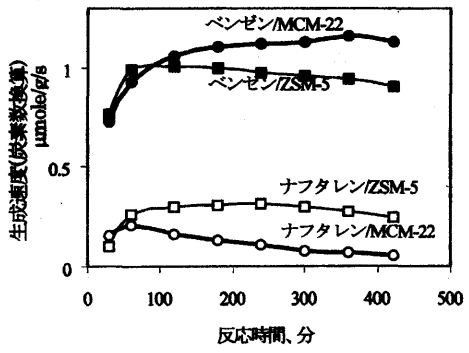


図4 6%Mo/HZSM-5と6%Mo/HZSM-5触媒の活性比較
(973 K, 3気圧, メタンF/W＝2700 ml/g/h)

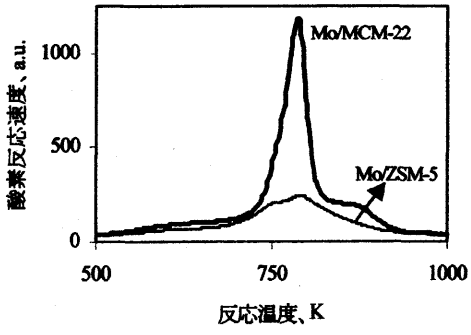


図5 図4の条件で6時間反応後のTPOスペクトル

は，ベンゼンやナフタレンの直接合成に有効であると考える。また，シリカ・アルミナ系ゼオライトでは，電荷バランスを保つため，Alの近傍にプロトン酸が発生する。低アルミナ或いは高シリカZSM-5，MCM-22，ZSM-11では，活性点である酸点同士が隣り合う機会が少なく重合反応が起こり難いことも，炭素生成抑制の原因となっている。

更にここで，ZSM-5担体とMCM-22担体の比較を行う。図4には，6 %MoをZSM-5とMCM-22担体に担持した触媒を使い，反応温度973 K，メタン2700 ml/g/h，全圧3気圧で反応させた結果を示す。いずれの触媒も長時間安定な活性を維持するが，Mo/MCM-22触媒が，より高いベンゼン生成速度と低いナフタレン生成速度を持つことが特徴である。これは，MCM-22がZSM-5に比べわずかに小さな細口径を有することに起因すると考えられる。図5には，反応6時間後の触媒に蓄積した炭素を酸素と昇温しながら反応させるTPO（Temperature

表4 各種金属を担持したHZSM-5の触媒特性[a)]

wt% 金属	Temp K	メタンF/W ml/g/h	conv %	arom. sel %	ref.
2Mo	973	1440	7.2	100	4
5Re[b)]	973	1440	7	95	10
2W[d)]	1023	1500	7.1	100	8
2W[b,c)]	1023	800	2.4	51	9
2Fe[b)]	1023	800	4.1	62	9
2V[b,c)]	1023	800	3.2	32	9
2Cr[b,c)]	1023	800	1.1	72	9
2Zn	973	1500	1.0	79	7
2Cu	973	1500	0.6	53	7
2Pt	973	1500	0.03	0	7
2Ni	973	1500	0.01	−	7

a) conv＝メタン転化率，arom sel.＝芳香族化合物選択率
b) 高沸点芳香族化合物を含む
c) COで前処理
d) 硫酸で前処理

Programmed Oxidation）実験の結果を示す。明らかに，Mo/MCM-22触媒がMo/ZSM-5触媒より，多くの炭素を蓄積している。蓄積炭素は活性点を被覆し触媒の活性を奪うと言われているが，図4に見るように両者にさほどの違いはない。この理由として，MCM-22では細孔内にスーパーケイジがあるため，反応生成物の移動に影響なく，多量の炭素を細孔内に保持できるためであると考えた。

メタンの脱水素縮合反応でベンゼンを合成するには，Mo/HZSM-5触媒が特に優れていることが知られている。そこで，Mo以外の活性な金属の探索がなされた[4,7-11]。表4には，HZSM-5に担持された各種金属触媒の探索結果を示す。著者らが見出したReとTsaiらの硫酸処理したWをHZSM-5に担持した触媒が，Moに匹敵する活性ならびに選択性を示すことが分かる。最近，β-Mo_2C上である種のアルキリデン種が900 Kでも安定に存在するとの報告があり[12]，メタンの脱水素芳香族化反応に活性なMo，Re，Wが，メタセシス反応に対しても特異的に活性な金属であることと考え合わせ，反応機作の面から興味深い。

以上の結果から，メタンの脱水素芳香族化反応に特異的に活性を示す触媒は，

① 中心金属としてMo，ReあるいはWを含み

② 担体は5.3～5.6 Åの細孔径を持ち
③ SiO_2/Al_2O_3比が40程度であり
④ 最適なプロトン酸量を持つゼオライト
であると結論された。

2.3 安定で高活性な反応条件

長い探索の結果見出されたMo/HZSM-5やRe/HZSM-5触媒でも，メタンと長時間反応させるとメタン反応率，ベンゼン生成速度ともに低下する。これは，副生する炭素によって触媒活性点が被覆されるためと解釈される。事実，反応後の触媒を酸素と反応させると多量の炭酸ガス生成を観測した。このような活性低下は，メタンの流速や反応温度を上げるなど，反応条件を厳しくするとより明瞭に現れる。我々は，原料メタンへのCOやCO_2の添加や反応圧力の変化によって活性低下の緩和あるいは防止を計った。また，単位時間当たりのベンゼン生成量を引き上げるため，高い反応温度での実験を行った。それらの結果を次に述べる。

2.3.1 CO/CO_2の添加効果

原料メタンに温和な酸化剤であるCOやCO_2を添加しRe/HZSM-5触媒上で反応させた結果を図6に示す[13]。図のタイトルに示すような反応温度1023 K，メタンF/W＝5000 ml/g/hという厳しい反応条件では，メタンのみを流通させながら反応すると（×），反応開始5時間後には初期活性の1/10にまで活性が低下する。一方，メタンにCOを添加することにより（○）反応5時間後において，まだ初期活性の半分の活性が保持される。さらに顕著な安定化がCO_2の添加によってもたらされる（■●▲）。即ち，わずか1～3％のCO_2添加で反応活性の低下が殆ど見られなくなり，5時間後も初期活性が保たれるようになった。しかし，過剰にCO_2を添加すると活性は低下した。同様なCOやCO_2の添加効果は，Mo触媒の場合も観測された[11]。この実験によって，長時間安定にメタンから水素とベンゼンなどの芳香族化合物を生成するための道が開かれた。

2.3.2 全圧と触媒の安定性

図7に反応温度1073 K，メタンF/W＝2700 ml/g/hに5％炭酸ガス添加という反応条件で求めたベンゼン生成速度の全圧依存性を示す[14]。全圧1気圧では

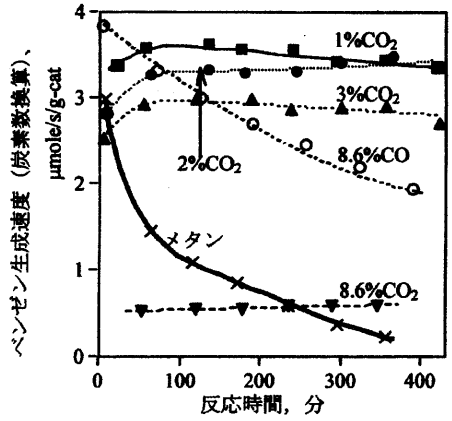


図6 メタン中にCOやCO_2の添加（Re/HZSM-5触媒，1023 K，3気圧，メタンF/W＝5000 ml/g/h）

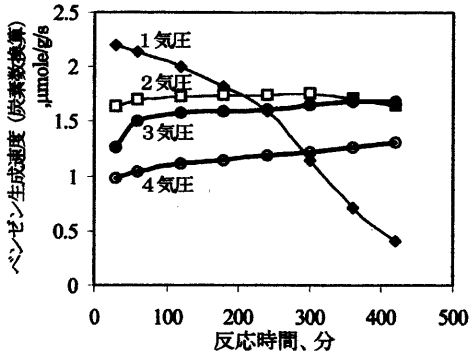


図7 全圧とベンゼン生成速度（6%Mo/HZSM-5，1073 K，メタンF/W＝2700 ml/g/h with 5 % CO_2）

触媒活性が反応時間と共に徐々に低下していくが，全圧2気圧で長時間安定な触媒活性を維持する。さらに圧力を上げても一定活性が長時間保たれる。しかし，圧力を上げるほど活性が低下した。この結果は，メタンからベンゼンを生成する反応が，$6CH_4 = C_6H_6 + 9H_2$と分子数が増える反応であるため，圧力を上げると反応平衡がメタン側によるためであると考えられる。1気圧以下の低圧領域で安定な活性が得られない理由は，炭素生成反応（$CH_4 = C + 2H_2$）が，ベンゼン生成反応より優位になるためであると考える。以上の結果から，本反応の最適な全圧は2～3気圧と求められた。

2.3.3 反応温度の影響

図2で見るように，反応平衡では高い温度ほどベ

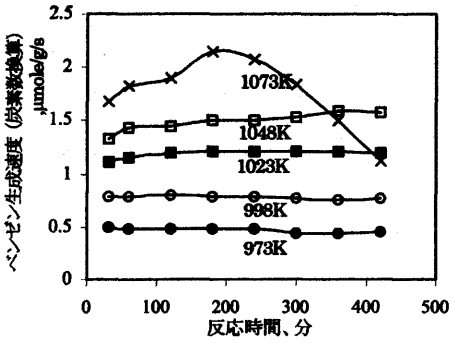


図8 反応温度とベンゼン生成速度（6%Mo/HZSM-5, 3気圧, メタンF/W＝2700 ml/g/h with 3 % CO_2）

ンゼンが多く生成する。しかし，高温ほど炭素生成も多くなり長時間安定な活性を得るのは難しい。そのためか，反応温度973 K以上の実験は，現在まであまり報告されていない。我々は，図6に見るようなメタンフィードに少量のCOやCO_2を添加すると反応活性が安定化することを利用し，メタンに3 %の炭酸ガスを添加した反応ガスを使い，973～1073 Kの反応温度で実験を行った。結果を図8に示す[14]。図から初期活性は，期待されたように反応温度の上昇と共に増加した。また，3 %の炭酸ガスの添加で，1048 Kまで安定な活性を得ることが可能となった。しかし，1073 Kで安定な活性を得るためには，3 %炭酸ガス添加では足りず，図7で見るように5 %の炭酸ガス添加が必要であった。さらに，温度を上げた1098 Kの反応で安定な活性を得るためには，7 %の炭酸ガスの添加が必要であった。しかし，1073 K, 5 %炭酸ガス添加と1098 K，7 %炭酸ガス添加の活性は，炭酸ガス添加による活性の目減りのため後者の方が低く，メタンF/W＝2700 ml/g/hの条件下で安定で最大の活性を得る反応温度と炭酸ガス濃度は，1073 K，5 %と求まった。

2.3.4 メタンへのCOやCO_2の添加による反応活性安定化のメカニズム

メタン中にCOを添加した場合，入り口と出口でのCO流速は殆ど変化しなかったが，CO_2をメタンに添加した場合，反応管の出口でCO_2は殆ど検知されず，添加CO_2の約2倍量のCO量を観測した。これらの結果は，CO_2は系中に多量にある水素と反応せず，炭素含有化合物（例えば，CH_x炭素($x=0$～4)）と$CO_2+CH_x=2CO+x/2H_2$のように反応することを示している。COは，Boudart反応によって一部CO_2となり，これがCH_x炭素と反応すると考えた。この際副生するCは，生成物であるベンゼンやエチレンなどの炭化水素に取り込まれることを，^{13}COの炭素を追跡することから明らかにした[13]。

表5 TPO実験によって求められた蓄積炭素量とそのH/C比[a]

番号	反応温度 K	CO_2 %	全圧 atm	炭素量 mol/g-cat	H/C
1	1073	0	3	0.00446	0.23
2	1073	3	3	0.00228	0.73
3	1073	5	3	0.00191	0.91
4	1073	6	3	0.00151	0.97
5	973	0	3	0.00134	0.76
6	1023	0	3	0.00220	0.73
7	1073	0	3	0.00446	0.23
8	1073	5	1	0.00263	0.68
9	1073	5	2	0.00203	0.76
10	1073	5	3	0.00191	0.91
11	1073	5	4	0.00175	0.97

a) メタンF/W＝2700 ml/g/h, 反応6時間後の触媒

どのような炭素含有物が炭酸ガスと反応するのかを反応使用後の触媒のTPO実験で明らかにしようとした[14]。この際，TCD検知器で反応した酸素の量だけでなく生成した水の量も定量した。これらの値を使い，平均としてCH_yと表した蓄積炭素が酸素と，$CH_y+(1+y/4)O_2=CO_2+y/2H_2O$の式に従って反応するとして，蓄積炭素量と蓄積炭素中のH/C比を求め表5に示す。表にはメタン中へCOやCO_2を添加した実験（番号1～4）に加え，全圧（番号5～7）及び反応温度（番号8～11）を変化させた実験で生成する炭素種についても記した。反応温度1073 K，全圧3気圧，メタンF/W＝2700 ml/g/hに固定してメタン中の炭酸ガス濃度を0から6 %に増やすと，予期されたように蓄積炭素量が約1/3に減少した。同時にH/C比は大きく増加した。全圧や反応温度の変化によっても同様であり，反応活性が安定化する（全圧を増す，反応温度を減ずる）方向で蓄積炭素が減少し，H/C比が増加した。これらの結果は，メタンが脱水素して$CH_3\rightarrow CH_2\rightarrow CH\rightarrow C\rightarrow C_n$（炭素重合物）となる過程で，$CO_2$が$CH_x$と反応し$C_n$に行

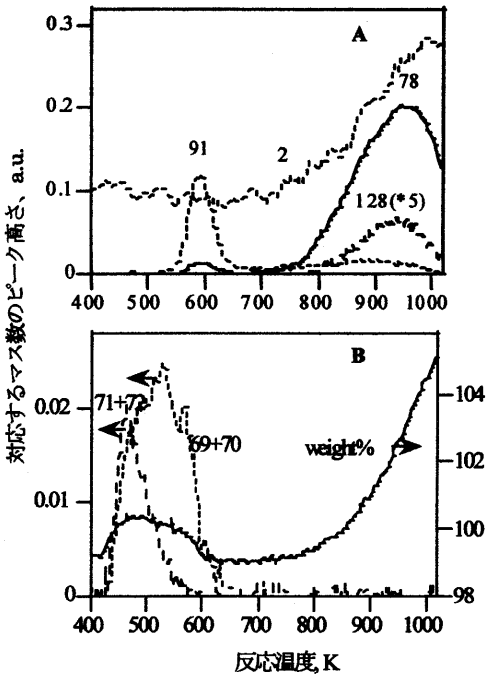
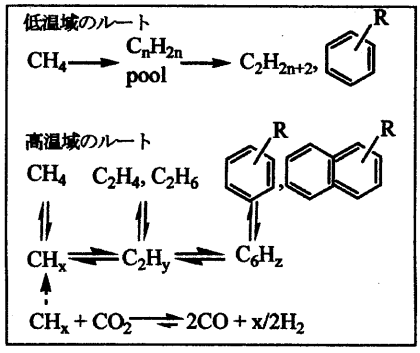


図9 エチレンをMo/HZSM-5に流して測定したTPR/TG/massスペクトル
A) 芳香族生成物, B) 脂肪族生成物と重量変化

くルートを阻害して蓄積炭素量を減らすと同時に，蓄積炭素種のH/C比を上げるとして説明した。

2.4 メタンからベンゼン生成の反応ルート

メタンの芳香族化合物と水素生成反応に関し，直接の証拠は無いものの，メタン→メタンの脱水素化物→エチレン→芳香族化合物という反応ルートが提唱されている。そこで我々は，触媒上にエチレンを流し，温度を上げながら質量分析器（mass）で生成物を分析すると同時に，触媒の重量変化を測定した。その結果を図9に示す[15]。まず，500 K付近でアルカン（71＋72）およびアルケン（69＋70）が生成し（観測されたC_2～C_7のアルカン，アルケンのうち，図にはC_5成分のみを示した。他の成分も同様な位置にピークを示す），次いで600 K付近でトルエン，キシレン（91）を主成分とする芳香族化合物生成のピークが観測される。この際，水素（2）は発生せず，重量変化はごくわずかであった。さらに温度を上げると全ての炭化水素生成が消滅した後，再度900 K以上で重量が増加しながら，水素とベンゼン（78），



スキーム1 芳香族化合物生成ルート

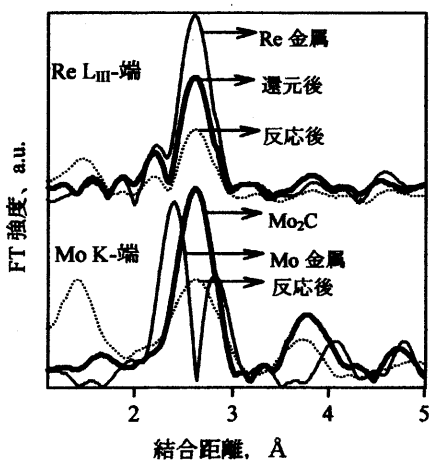
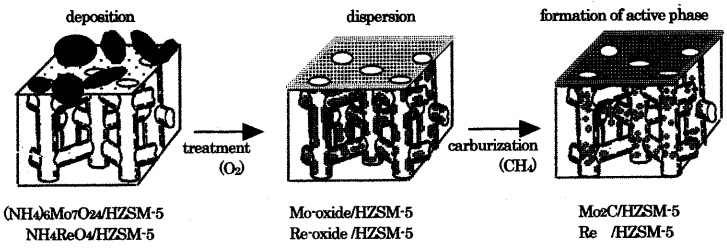


図10 Mo, Re触媒および対応する参照試料のEXAFSスペクトル

ナフタレン（128）が生成した。メタンの反応では，エチレンの高温域に対応する領域においてのみ，重量が増加しながら水素とベンゼンを生成した。これらの結果から，芳香族化合物は，高温域と低温域で全く違う反応ルートを通って生成し，メタンからのベンゼン生成は高温域に対応すると推論した。低温域と高温域の反応ルートについてスキーム1を提案した。

2.5 反応活性種

最近，Moがどのように HZSM-5担体に担持されるかについてESR，^{27}Al-NMR，^{1}H-NMR，TPR，IR，SEMなどの測定結果から議論した幾つかの報告がある[10,16]。まず，著者らは，HZSM-5のSEM像とMo原料をHZSM-5に担持焼成後のSEM像に殆ど違いが認められなかったことから，MoがHZSM-5上に均一に分散していることを示した。また，Mo



スキーム2 メタンの芳香族化反応用触媒の調製

をHZSM-5に担持し加熱した後のIR及び^{1}H-NMR観測の結果から，Si-OH及びAl-OHの全てが減少するが，Moの存在しないHZSM-5を加熱してもごくわずかしか変化せず，MoとOH基との反応が示唆された。さらに，加熱によって生成する水の量は，Moの担持量に比例して量論的に増加し，MoとOH基との反応が結論された。酸性なOH基の大部分が細孔内に存在することから，少なくともMoの一部はZSM-5の細孔内に入り，Alの近傍にある酸性なOH基と反応すると結論できる。

さらに，焼成後のMo/HZSM-5やRe/HZSM-5にメタンを流し873 K以上にすると，COやCO_2が発生し，その終了とともにエチレン，ベンゼンなどの炭化水素の生成が始まる[5,11]。即ち，MoやRe触媒がメタンから高次の炭化水素を生成するには，触媒の還元が不可欠である。還元された触媒がいかなる化合物になっているかを調べるために，著者らは，反応前後の触媒のEXAFS測定を行った。結果を図10に示す。Mo触媒をメタン反応に使用した後のEXAFSスペクトルは，比較試料のβ-Mo_2Cのそれと同一であるがMo金属とは明らかに異なる。即ち，Mo/HZSM-5ではβ-Mo_2Cがメタンの芳香族化反応に活性な化合物である。一方，メタンの反応に使用した後のRe触媒のEXAFSスペクトルは，Re金属および水素還元後のスペクトルと同一であり，金属が活性相であることが結論された。最近，Iglesiaら[15]は，Mo/HZSM-5触媒において酸性なOH基とMoとの反応で出来たMo-Oがメタンと反応してMoカーバイドを生成する際，酸性なOH基が再生するとの議論を展開している。

以上の結果から，触媒調製の各ステップで中心金属がどのように変化し，活性な相を形成していくかをまとめ，スキーム2に示す。まず，高温の焼成処理によりMoあるいはRe酸化物が担体の細孔内／外に高度に分散する。次いで，メタンとの反応でMoの場合はMo_2CをReの場合は金属粒子を形成し，これらと担体のプロトン酸点との協調作用によって，メタンの脱水素縮合反応によるベンゼンなどの芳香族化合物生成を促進すると考えた。

3. 将来の展望

未利用で豊富な炭素資源である天然ガス（メタン）と環境負荷成分であるCO_2を利用して水素とベンゼンやナフタレンなどの化学原料を製造する新しい触媒技術について述べた。現在，本プロセスの実用化に向けた実証試験が，平成12～14年度NEDO地域コンソーシアム事業として取り上げられ，北海道大学，北海道曹達，日本製鋼所，日揮の共同研究として開発が進められている。しかしながら，本プロセスの実用化に向けては，メタン転化率の向上，大きな吸熱反応による触媒層の温度分布の軽減，973 K以上の高温で安定な流動床用触媒の開発，効率的な生成物の分離技術開発，熱資源の有効利用等々障害となる幾つものハードルがある。これらを解決すれば，これからの資源エネルギー問題解決に大きな手掛かり与えることが期待される。

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Synthesis of Benzene from Methane in Catalytic Dehydrocondensation
– Templating Role of Catalyst –

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Dehydrocondensation reaction of methane forming aromatics and hydrogen was reviewed. High activity and high formation selectivity of aromatics in the reaction were realized only on selected catalysts. The necessary factors for central metal and zeolite support of the selected catalysts were described. Catalytic activity kept unchanged for long time-on-stream when CO_2 or CO was added in methane feed at 2~3 atmospheric pressure due to effective removal of coke from catalyst surface by CO or CO_2. Further, the route from methane to aromatics and formation process of active phase of catalyst were discussed.

Keywords: methane, benzene, dehydroaromatization, Mo catalyst, Re catalyst, CO_2

《解 説》

ゼオライト触媒上での炭化水素によるNO選択還元反応：形状支配拡散と吸着支配拡散の影響

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炭化水素によるNO選択還元反応にゼオライト触媒が高い活性を示すが，ゼオライト細孔内の特殊反応場は拡散抵抗というマイナスの効果を持つ場合もある。ディーゼル排ガス浄化のように分子サイズの大きい炭化水素を還元剤に用いた場合，分子サイズとゼオライト細孔径の幾何学的形状に規定された形状支配拡散により拡散速度が著しく低下し，脱硝活性が大きく阻害されることがある。一方で，小分子の場合でも拡散分子と交換カチオンの吸着相互作用が過度に大きくなると，拡散分子が強吸着することにより拡散速度が著しく低下し，脱硝活性の低下を引き起こすことが初めて見出された。このときの拡散は，拡散分子と交換カチオンの吸着相互作用に基づいた吸着支配拡散であり，従来から知られていた形状支配拡散の機構とは全く異なる。このように，ゼオライト細孔内拡散は，従来から知られていた形状支配拡散に代表される物理的な現象だけでなく，交換カチオンと拡散分子の吸着相互作用によっても影響される極めて化学的な現象であることが明らかとなった。

1. はじめに

次世代脱硝技術として期待される炭化水素によるNO選択還元反応（HC-SCR）において，Cu-ZSM-5等の金属イオン交換ゼオライトが高い活性を持つことが知られている[1-5]。金属イオンの種類を変えると触媒活性が著しく変化することから，金属イオンが触媒活性の主要な因子であることは間違いない。さらに，ゼオライト系触媒が酸化物系触媒に比べて著しく高い活性を持つことを考えると，ゼオライト中に保持された特殊な形態の金属イオン，つまり，アニオンが配位していない，あるいは配位不飽和の金属イオンが孤立していることが高活性の主要な原因であろう。また，幅広い温度域で高い活性・選択性を示すことから，ゼオライトのミクロ細孔に起因した高い吸着力もその原因の一つと考えられている。一方で，ミクロ細孔を持つがゆえに生じる様々なマイナスの効果も予想される。特に，本反応のように過酷な反応条件下において有効に機能する高活性ゼオライト触媒を開発する場合，ゼオライト細孔内の拡散抵抗の問題は避けて通れないであろう。ゼオライトのような数オングストロームの細孔内における分子の拡散は超ミクロ孔拡散（configurational diffusionあるいはrestricted diffusion）と言われ，通常の分子拡散やKnudsen拡散の機構とは全く異なる[6]。この超ミクロ孔拡散は，細孔と拡散分子の幾何学的形状に大きく影響を受け，わずかな細孔径の減少や分子サイズの増大によって拡散係数が10桁近くも小さくなってしまうだけでなく化学現象に匹敵する活性化エネルギーを必要とするようになる[7,8]。特定の分子の拡散や特定の反応の進行が立体的因子によって阻害される結果として発現する形状選択性は，この拡散の影響が現れる例のひとつとしてよく知られている[9,10]。しかし，ゼオライト細孔内拡散は幾何学的制限（形状支配拡散）だけで決まるわけではなく，拡散分子と表面の化学的な相互作用に影響を受けることも報告されている。例えば，ZSM-5中の炭化水素の拡散がAl含有量の増大につれて遅くなる現象は，酸点上で拡散分子が滞留するためとされている[11,12]。後述のように，我々は，化学的な相互作用によってゼオライト細孔内の拡散が形状支配

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拡散に匹敵するほど遅くなることを見いだした。本稿では，ゼオライト触媒上でのNO選択還元反応に対する形状支配拡散と吸着支配拡散の影響を紹介する。

2. 反応速度に対する細孔内拡散の影響の評価法（触媒有効係数解析）

みかけの反応速度に対する拡散の影響は触媒有効係数を求めることで評価できる。反応速度に比べて拡散速度が遅く，ゼオライト結晶サイズが大きい（つまり細孔が長い）場合には，反応分子は外表面付近の活性点で消費されてしまい，結晶内部にまで到達しない。その場合，触媒粒子の外表面付近の活性点は触媒反応に使われるが，結晶中心部はまったく寄与しないことになる。触媒反応に使われる割合，つまり，実際の反応速度と拡散速度が十分に速いと仮定した（触媒粒子内部まで反応物濃度が均一と仮定した）場合の速度の比は，触媒有効係数（η）と呼ばれ，触媒粒子が球状で1次反応の場合には，触媒粒子半径（R），真の反応速度定数（k），および有効拡散係数（D_e）を用いて，以下のような比較的簡単な関数で表される[13]。

$$\eta = \frac{1}{\phi}\left(\frac{1}{\tanh(3\phi)} - \frac{1}{3\phi}\right)$$

$$\phi = \frac{R}{3}\sqrt{\frac{k}{D_e}}$$

ここで，ϕはThiele数と呼ばれる。ところが，実際にこの式を用いて触媒有効係数を直接求めることはそう簡単ではない。なぜなら，触媒粒子径（R）は電子顕微鏡や吸着法などにより容易に求めることができるが，真の反応速度定数（k）や有効拡散係数（D_e）を直接求めることはそれほど容易ではないからである。そこで，一般には，粒子径の異なる複数の触媒を用いて見かけの反応速度を測定し，反応速度と粒子径の関係を解析することで触媒有効係数を求める方法がとられる（粒径変化法）[7,14,15]。しかし，依然としていくつかの問題があるように思われる。第1は球形粒子と1次反応の仮定であるが，幸い，反応次数が変わっても，触媒粒子の形状が変わっても，Thiele数を下記のように定義しなおせば，触媒有効係数とThiele数の関係はそれほど大きく変化しない[16]。

$$\phi_S = \frac{V_p}{S_p}\left(\frac{(n+1)kC^{n-1}}{2D_e}\right)^{\frac{1}{2}}$$

ここでV_pとS_pは触媒粒子の容積と外表面積，nは反応次数，Cは触媒外表面における反応物濃度である。ちなみに，球状粒子の場合には，V_p/S_pは$R/3$に等しい。第2の問題は触媒粒子径を変化させることとその測定である。ゼオライト細孔内の拡散を問題とする場合には，上の「粒子」はゼオライト結晶を意味することになるので，ゼオライトの結晶子径を変化させ，その結晶子径（あるいは，V_p/S_p）を測定しなければならない。この場合にも，幸いなことに触媒粒子径は系統的に変化させることは必ずしも要求されていない。大結晶と小結晶の2種類だけでも触媒有効係数を求めることができる。また，窒素吸着等温線のt-plot解析により外表面積を求めれば，V_p/S_pも計算することができる。

3. NO選択還元における形状支配拡散の影響

炭化水素によるNO選択還元法（HC-SCR）は希薄燃焼排ガス中のNO_x除去を想定しているが，中でもディーゼル車の排ガス浄化技術の達成は急を要する課題である。HC-SCR法は排ガス中の未燃炭化水素を還元剤として利用することを想定しているが，ディーゼルエンジン排ガス中の未燃炭化水素濃度はNO_x還元に十分でない。そのため，排ガス中に燃料自体を添加する方法や，炭化水素を触媒上に吸着・濃縮して濃度を高める方法などが考えられている[17]。その場合，還元剤として利用される炭化水素は比較的大きな分子サイズのものが想定されるため，上述のようにゼオライト細孔内拡散の影響は無視できなくなるであろう。このような予想は容易にできるが，実際にどの程度の分子サイズの炭化水素まで還元剤として有効に利用できるのかを明らかにするには，反応速度に対する拡散の影響を明らかにする必要がある。そこで我々は，分子サイズの異なる種々のアルカンと細孔径の異なるゼオライトを用いて，幾何学的形状に起因した拡散速度の違いがNO還元速度に及ぼす影響を定量評価することに試みた。

3.1 触媒有効係数の推算

触媒には結晶サイズの異なるMFI型及びMOR型ゼオライトをCuイオン交換したものを用いた（表1）。

表1 使用触媒の細孔径，結晶サイズおよび化学組成

Sample	Micropore size(Å)	Crystal size(μm)[a]	Si/Al ratio	Cu/Al ratio	Cu exchange level (%)[b]
Cu-MFI(S)	5.1×5.6	0.10	22	0.51	102
Cu-MFI(L)		1.29	20	0.50	100
Cu-MOR(S)	6.5×7.0	0.21	7.5	0.26	52
Cu-MOR(L)		0.71	8.1	0.23	46

[a] Calculated from the external surface area determined by N_2 adsorption isotherms.
[b] Cu/Al×200

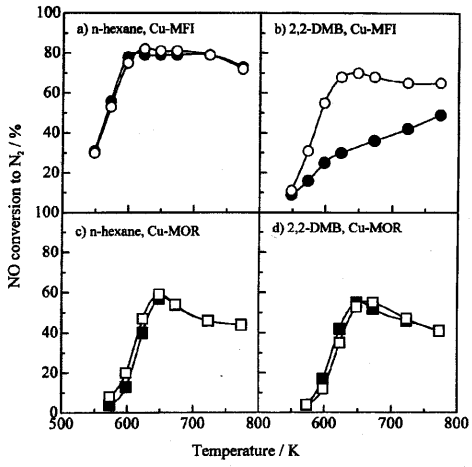

図1 *n*-hexaneおよび2,2-DMBによるNO選択還元反応 (○) Cu-MFI(S); (●) Cu-MFI(L); (□) Cu-MOR(S); (■) Cu-MOR(L)

これらは，ゼオライト結晶のバルク（ICP）においても，外表面層（XPS）においても，Si/Al，Cu/Al比はほぼ同じであり，結晶サイズ以外の化学的性質が一次近似として同じであると見なすことができる。これら触媒上でのC6アルカンによるNO選択還元反応の結果を図1に示した[18,19]。還元剤として*n*-hexaneを用いた場合には，NO転化率は結晶サイズに依存せず，互いに良く一致した（図1a)。一方，2,2-dimethylbutane (2,2-DMB) を用いた場合には，明らかに大結晶Cu-MFI(L) の方がNO転化率は低く抑えられていた（図1b)。ところが，Cu-MOR触媒では，*n*-hexaneと2,2-DMBのどちらを用いた場合でも，NO転化率は結晶サイズに依存しなかった（図1c,d)。同様の結果は*n*-octaneと*i*-octane (2,2,4-trimethylpentane) を用いた場合にも観察された[19]。

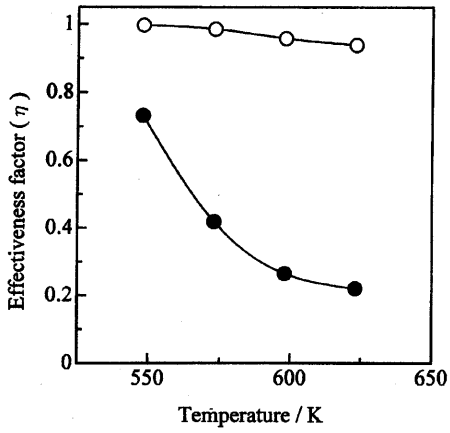

図2 2,2-DMBによるNO選択還元における触媒有効係数の温度依存性。(○) Cu-MFI(S); (●) Cu-MFI(L)

すなわち，Cu-MFIを用いた場合には，*n*-octaneでは結晶サイズの影響は見られないが，*i*-octaneでは明らかに大結晶の方が低い活性を示した。一方で，Cu-MORを用いた場合は結晶サイズの影響は全く観察されなかった。これらの結果は炭化水素の最小分子径とゼオライト細孔径によって説明できる。すなわち，2,2-DMBや*i*-octaneの最小分子径（0.62 nm）はMFIの細孔径（0.51-0.56 nm）より大きいため，細孔内拡散抵抗が大きく，大結晶ほど見かけの反応活性は低くなったと考えられる。これに対して，炭化水素の最小分子径がゼオライト細孔径より小さい場合には，細孔内拡散抵抗もそれほど大きくなく結晶サイズの影響は現れない。

2,2-DMB/MFI系について微分領域で測定した反応速度とゼオライト結晶子径から求めた触媒有効係数の温度依存性を図2に示した[18]。小結晶Cu-MFI(S) の場合には，触媒有効係数はほぼ1に等しく，結晶内部の活性点まで反応に有効に使われていることを示している。一方で，大結晶Cu-MFI(L)の触媒有効係数は1以下となり，特に623 Kでは0.2近くまで低下している。つまり，この温度では大結晶Cu-MFI(L) 中の全触媒活性点のわずか20 %程度しか反応に寄与していないのである。高温ほど触媒有効係数が小さくなるのは反応工学の常識であるが，有効拡散係数の温度依存性より反応速度定数のそれの方が大きいためである。すなわち，高温ほど反応速度定数／有効拡散係数の比が大きくなるので，Thiele数が大きくなり，触媒有効係数は小さくなる。

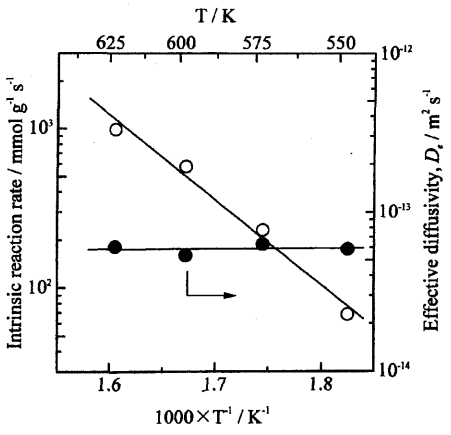

図3 反応速度定数(○)および有効拡散係数(●)のアレニウスプロット

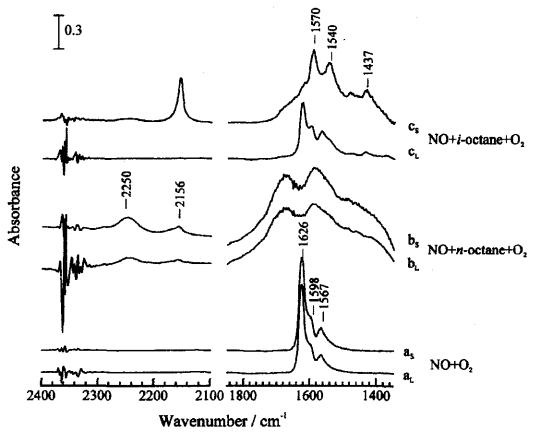

図4 各種ガス流通下の*in-situ* IRスペクトル (523 K)
a) NO + O_2 for 30 min, b) NO + *n*-octane + O_2 for 60 min, c) NO + *i*-octane + O_2 for 60 min. S: Cu-MFI(S), L: Cu-MFI(L)

実際に触媒有効係数の式から求めた反応速度定数と有効拡散係数の温度依存性は図3のようである。ところで，有効拡散係数は気相濃度基準の拡散係数であり，結晶内濃度基準の拡散係数（結晶内における1分子の移動度を表す）に分配係数を乗じたものである[9,11]。図3の場合には有効拡散係数は温度にほとんど依存していないが，これは結晶内拡散係数の温度依存性と分配係数の温度依存性により見かけ上相殺された結果と考えられる。H-MFI上の2,2-DMBの吸着エンタルピーは63 kJ mol^{-1}であると報告されている[11]ので，結晶内拡散の活性化エネルギーもその程度の値を持つことになるが，この値は上述の超ミクロ孔拡散の特徴を反映して，通常の拡散に対する値に比べてかなり大きい点にも注意が必要である。ちなみに，2,2-DMBの最小分子径はMFI細孔径よりも大きいが，この解析結果から，見かけゼオライト細孔径よりもやや大きい分子でも細孔内を(非常に制限されながらも）拡散できることを示しており，炭化水素やゼオライト骨格はある程度フレキシブルであると考えられる。

3.2 *In-situ* IRを用いた形状支配拡散の評価

上述のように，炭化水素の最小分子径がゼオライト細孔径より大きい場合，拡散速度の低下により見かけの反応活性に結晶サイズの影響が現れる。この場合，触媒上の表面吸着種にも結晶サイズの影響が見られるはずである。図4に結晶サイズの異なるCu-MFIに各種ガスを流通した時の定常状態のIRスペクトルを示した[19]。NO + O_2混合ガスを流通した場合，結晶サイズに関わらずNO_2 (1626 cm^{-1}) とNO_3^- (1598, 1567 cm^{-1}) に帰属される吸着種が観察され，それらの強度もほぼ同じであった。同様に，NO + *n*-octane + O_2流通下でも結晶サイズに関わらず表面吸着種はほぼ同じであり，それらは主に吸着炭化水素種や炭化水素の部分酸化物に由来のブロードな吸収が観察された。一方で，NO + *i*-octane + O_2流通下のIRスペクトルは結晶サイズの違いにより明らかに異なることがわかる（spectrum c)。小結晶Cu-MFIでは*i*-octaneの部分酸化物種 (CO, $RCOO^-$) などが主に観察されたが，大結晶Cu-MFIではNO_y種 (NO_3^-, NO_2) が主な吸着種であり，*i*-octane由来の吸着種はほとんど観察されなかった。この吸着種の違いは*i*-octaneの拡散が制限されていることに起因していると考えられる。すなわち，NOやO_2はCu-MFIゼオライトの細孔内を容易に拡散し，結晶内部のCuイオン上でNO_y種を生成することができる。ところが*i*-octaneの拡散は非常に制限されるため，大結晶ほど*i*-octaneが結晶内部に十分拡散できないため，NO_y種が主に観察されたのであろう。

4. NO選択還元における吸着支配拡散の影響

これまで述べてきた形状支配拡散の概念を単純に外挿して，NO選択還元反応の還元剤としてよく用いられているC2～C3程度の炭化水素の場合には，ゼオライト細孔内拡散は影響を持たないと予想する

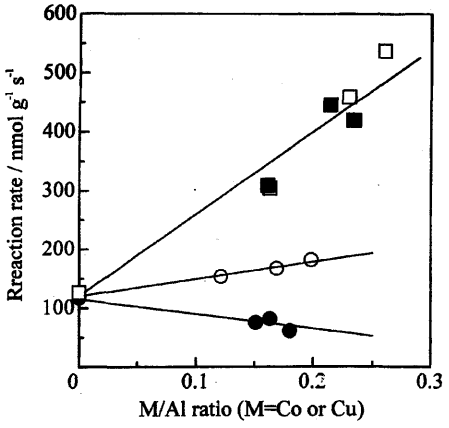

図5 金属イオン交換量に対するNO反応速度（673 K）
(○) Co-MOR(S); (●) Co-MOR(L); (□) Cu-MOR(S); (■) Cu-MOR(L)

のは正しいであろうか？答えは間違いである。我々は，形状支配拡散とは異なる機構によってゼオライト細孔内の拡散が極めて遅くなり，形状支配拡散以上の影響を持つことがありうることを見出した。

4.1 細孔内拡散に対する交換カチオンの影響

最初の例は，Co-MOR上でのプロパンによるNO選択還元反応である[20,21]。図5にNO反応速度に対する金属イオン交換量の影響を示した。H-MOR上（図中のM/Al＝0）では，反応速度はゼオライト結晶子径によらず，細孔内拡散の影響は見られない。Coイオン交換量の変化につれて反応速度は変化したが，その影響はゼオライト結晶子径によって明らかに異なることがわかる。小結晶MOR(S)では，反応速度はCoイオン交換量に対して直線的に増加した。しかし，大結晶MOR(L)では反応速度は逆に低下した。ところが，同じ母体ゼオライトでもCuイオン交換した場合には，反応速度は結晶子径にほとんど依存しない。Co型の方がCu型よりも活性は低いにも関わらず結晶子径の影響を受けやすいのは，Cu型に比べて細孔内拡散が遅いことを示唆していると考えられる。この結果は交換金属イオンの種類によって細孔内拡散の速度が変化することを示している。ゼオライト中に交換された金属イオンの種類や量によってゼオライトの有効細孔径が変化し，それによって拡散速度が変化することが知られている[22]。しかし，Co^{2+}とCu^{2+}のイオン半径にほとんど差がないので，有効細孔径が変化したことでは上の結果を説明できない。このような拡散挙動の異常性は，NOとCoイオンの強い相互作用に起因していると考えられる。Co-MORおよびCu-MOR上でのNO昇温脱離実験（NO-TPD）を行ったところ，NOはCuイオンよりもCoイオン上により強く吸着していることが明らかとなった[21]。このCoイオン上へ強吸着したNO_x種が拡散速度の低下に関係しているようである。ひとつの可能性として，吸着NO_x種によってゼオライトの有効細孔径が小さくなり拡散速度が低下したことが考えられる。もう一つの可能性は，NO分子の拡散を吸着点から吸着点への移動と考えた場合，強吸着によって移動の活性化エネルギーが増大したため拡散速度が低下したことが考えられる。一方で，Cu-MOR上にNOがそれほど強く吸着しないことから，Cu-MOR上で細孔内拡散の影響が見られなかったことも理解できる。

4.2 細孔内拡散に対する炭化水素種の影響

上述のように，ゼオライト細孔内拡散において吸着というパラメーターが重要な支配因子となり得ることを考えると，カチオンと相互作用の大きい分子の場合，拡散速度が小さくなることが予想できる。例えば炭化水素でもアルケンのような不飽和炭化水素を還元剤として用いた場合，それ自体が強吸着することにより細孔内拡散が遅くなることがある[23,24]。実際に，Cu-MFI上でプロパンとエチレンを還元剤に用いて検討した結果，プロパンでは結晶サイズの影響は見られないが，エチレンでは明らかに結晶サイズの影響が見られ，大結晶Cu-MFIの方が小結晶Cu-MFIに比べてNO転化率は低いことがわかった[23]。エチレンを用いた場合の触媒有効係数の温度依存性を図6に示したが，大結晶Cu-MFI(L)の触媒有効係数は1以下となり結晶内部の活性点は有効に使われていないことがわかる。大結晶の温度依存性を見ると，反応温度の増加につれて触媒有効係数が大きくなり，反応温度が高いほど触媒は有効に使われていることを示している。この触媒有効係数の温度依存性は，上述の形状支配拡散の傾向（図2参照）と逆であり，一見すると反応工学の常識に反しているように見える。この触媒有効係数の温度依存性は，細孔内拡散が化学反応より見かけ上，大きな活性化エネルギーを持つとしなければ説明できない。実際に，触媒有効係数とThiele数から求めた真の反応速度定数と有効拡散係数の活性化エネルギーは，それぞれ，

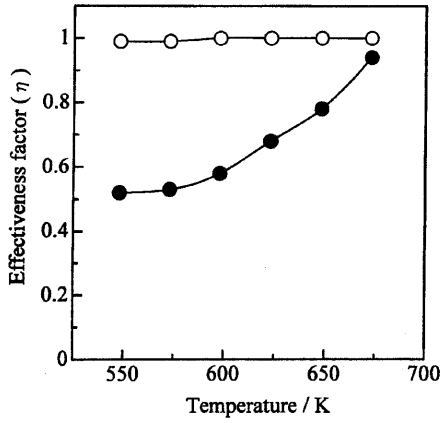

図6 エチレンによるNO選択還元における触媒有効係数の温度依存性。(○) Cu-MFI(S); (●) Cu-MFI(L)

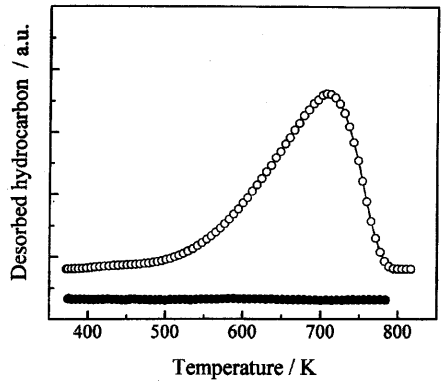

図7 炭化水素の昇温脱離プロファイル
(○：エチレン，●：プロパン)

116 kJ mol^{-1}と150 kJ mol^{-1}であった[23]。ちなみに，このようにして求めた拡散係数の値は，Masudaらの容量法による拡散係数の実測値とよく一致していた[24]。また，Cu-MFI中のエチレンの拡散係数はSilicaliteに比べて10^{-4}～10^{-5}程度小さくなっていることがわかった[24]。ゼオライト細孔内拡散がこのような小さな拡散係数と大きな活性化エネルギーを持つ原因としては，プロパンを用いた場合に拡散抵抗がないこと，エチレン等の分子サイズが細孔径よりかなり小さいことなどから，形状支配拡散は除外される。Cu-MFI上へのプロパンとエチレンの吸着特性を調べるため，炭化水素の昇温脱離実験（HC-TPD）を行ったところ，プロパンはCu-MFIにほとんど吸着しないのに対し，エチレンは高温まで非常に強く吸着することが分かった（図7)。この場合も，4.1で述べたCoとNOの組み合わせの場合と同様に，Cuイオン上へのエチレンの強吸着が拡散係数の低下に関係しているようである。エチレンはπ電子を有するので，遷移金属であるCuイオンと強く相互作用し，Cuイオン上に強吸着して安定化することで拡散速度の低下を招き，見かけの反応速度に拡散の影響が見られたのであろう。

以上のように，金属イオン交換ゼオライト触媒上でのNO選択還元反応においては，ゼオライト細孔内の特殊反応場が拡散抵抗というマイナスの効果を持つ場合があることが明らかとなった。この時の拡散抵抗は，従来から知られていた形状支配拡散に代表される物理的抵抗だけでなく，金属カチオンと拡散分子の吸着相互作用に基づく抵抗も存在する。よって，ゼオライト細孔内の拡散現象を理解する上で，形状支配拡散に影響するゼオライト細孔径や拡散分子サイズに加えて，吸着支配拡散に影響する交換カチオンと拡散分子の化学的相互作用も考慮する必要がある。

5. おわりに

これまでゼオライト細孔内特有の拡散機構として，形状支配拡散がクローズアップされてきた。しかし，本稿で述べたように，吸着支配による拡散もまた形状支配拡散を上回る影響を触媒反応に与えることが明らかとなった。吸着相互作用によっても拡散速度が抑えられることは以前から知られていたが，これほど大きな影響を与えるとは予想されていなかった。拡散現象は，もっぱら化学工学の分野で議論されてきたが，極めて化学的な現象であることも明らかになったわけである。ゼオライト細孔内の分子ダイナミックスがあらたな視点で見直されるきっかけになれば幸いである。

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Selective Catalytic Reduction of NO by Hydrocarbon over Zeolite Catalyst : Influence of Geometry-limited Diffusion and Adsorption-controlled Diffusion

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Although it has been reported that zeolite catalysts have high activity for the selective catalytic reduction of NO by hydrocarbon, unique reaction field of zeolite micropore can also have negative effect of diffusion resistance. In such diesel exhaust with using large molecular size hydrocarbon as a reductant, de-NO_x activity was hindered by the diffusion determined by geometrical configuration between the diffusing molecule and zeolite pore, *i.e.*, geometry-limited diffusion. In the case of small molecule, on the other hand, it was found that the excessive adsorptive interaction between diffusing molecule and exchanged cation led to significant decrease in diffusivity, which resulted in the decrease in de-NO_x activity. In this case, the diffusion was controlled by the adsorptive interaction, *i.e.*, adsorpion-controlled diffusion, whose mechanism was entirely different from the geometry-limited diffusion. Thus, zeolite diffusion was not only a physical phenomenon such as geometry-limited diffusion but also a chemical phenomenon influenced by the interaction between exchanged cation and diffusing molecule.

Keywords: Zeolite, HC-SCR, Diffusion, Geometry, Adsorption

《 レポート 》

ファインケミカルズ合成触媒国際シンポジウム（C&FC2001）に参加して

岐阜大学工学部　窪田好浩

ファインケミカルズ合成触媒国際シンポジウム（C&FC2001）が3月12日～14日の3日間，早稲田大学国際会議場において，触媒学会・ファインケミカルズ合成触媒研究会と早稲田大学の共同主催の元に開催された。主要テーマは，1) 固体触媒，錯体触媒，生体触媒を用いた触媒反応の開発，2) 化学量論反応から触媒量反応への転換，3) 有害物質を使わず安全な物質を使用する合成法，4) 安全かつ効率的に化学反応が起こる反応媒体の開発，5) 新反応プロセス，新合成反応の開発，6) グリーン触媒を用いた効率的なグリーンケミストリーの実現等であった。事前登録参加者数は207名であり，ゼオライト分野からも二十数名の参加があった。

初日，今木実行委員長（三菱化学）のご挨拶，J. C. Warner教授（Univ. of Massachusetts Boston）による製品開発におけるグリーンケミストリーについての概論を皮切りに，Plenary lecture 5件，Invited lecture 11件の講演が3日間を費やして行われた。内容について一つ一つ述べることはここではしないが，最初の講演に象徴されるように，全体としてのトーンは「グリーンケミストリー」を意識したものであり，グリーンケミストリーのシンポジウムであったとしても十分成立する内容であった。これは，もともと触媒を用いたファインケミカルズ合成がより高い原子効率，より小さいE-ファクター値を追究するものであり，そのものがグリーンケミストリーの理念に合致しているからだとも言える。最も多かったのは，Pd錯体を初めとする均一系遷移金属触媒や生体触媒を用いる各種有機合成に関する講演であった。その他，医薬品合成のプロセス開発やコンビナトリアルケミストリーに関する講演等があり，内容的には上記の主要テーマの全てを網羅していた。

ゼオライト分野の研究者にとって馴染みの深い演者として，R. A. Sheldon教授（デルフト工科大学）とW. F. Hölderich教授（アーヘン工科大学）の二人が来日された。Sheldon教授は，ファインケミカルズ合成触媒（ゼオライト触媒を含む）に関する得意のReviewをされ，Hölderich教授はRh錯体固定化MCM-41を用いた不斉水素化反応に関する興味深い成果を発表された。

一般の発表は全てポスター形式で行われた。全部で60件のポスター発表があり，そのうち，広い意味でのゼオライト関連の発表件数は全体の1／4程度であった。ポスター発表者の中で，希望者には“Short Oral Presentation”の時間が特別に与えられ，約7割の人がこれを行った。この口頭発表では日本語・英語の使用が許されていたが，9割以上の発表者が実際には英語で行った。

この会議で，十年近く前に高分子の仕事をしていた頃の知り合いと再会した。彼によれば，この会議は有機合成・触媒化学のいずれにも偏りすぎないところが良いとのことであり，筆者も同感であった。ただ，ゼオライトに関する話題が少ないことは少々寂しかった。とは言え，新開博士（エーザイ）の「プロセスケミストリーは大学で教育することが難しい（教官が詳しくないため）が，将来プロセスケミストを目指すとすれば，実験で観察したこと（例えば反応の色や変化など）から何が起こっているかを見抜く（感じ取る，考える）ことの出来る人が向いている」等というコメントや，学術研究とグリーンケミストリーの関係についてのSheldon教授のコメントなど，質疑応答における各研究者の見解は非常に参考になった。

尾中先生のClosing remarksによれば，本シンポジウムに強い関心をもった女子高校生が遠方からわざわざ見学に来たとのこと。これは情報発信の威力と本シンポジウムの魅力を端的に示すエピソードと言える。シンポジウムを成功に導かれた尾中先生（東大），清水先生（早大）など実行委員の方々に祝福・感謝申し上げるとともに，運営を支えてくれた早稲田大学，東京大学の学生さん達にも感謝したい。

タイトルサービス

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JOURNAL OF POROUS MATERIALS

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お知らせ

第17回ゼオライト研究発表会

第17回ゼオライト研究発表会を下記の要領で島根県松江市の県民会館で開催致します。ゼオライトおよびその類縁物質の基礎と応用について新たな展開を志向します。充実した研究発表と活発な討論の場に，多数の研究者・技術者が参加されることを期待します。この機会に奮ってご参集下さい。

主　催：ゼオライト学会

共催等（順不同）：化学工学会，触媒学会，石油学会，日本イオン交換学会，日本エネルギー学会，日本化学会，日本セラミック協会，日本地質学会，日本粘土学会，有機合成協会（予定）

日　時：平成13年11月21日（水），22日（木）

会　場：島根県民会館（島根県松江市殿町158）JR松江駅からバス約10分，徒歩約20分。出雲空港と米子空港からJR松江駅行きバス運行（共に約45分）

テーマ：ゼオライトおよびその類縁化合物に関連した研究の基礎から応用まで

講演の種類：1）特別講演（2件予定），2）総合研究発表（成果がある程度まとまっている研究を総合したもの。したがって，既発表の研究成果であってもそれらをまとめた内容であればよい。討論を含めて30分程度），3）一般研究発表（未発表の研究成果の発表。討論を含めて20分程度）

発表使用機器：OHP（OHP以外の機器を使って発表される方は，下記問い合わせ先までご連絡下さい）

講演申込締切：7月20日（金）

講演申込：1）講演題目，2）発表者氏名（講演者に○印），3）所属機関，4）講演の種類（総合研究発表か一般研究発表かの区別），5）研究分野（プログラム編成参考用に，つぎの分野のうち一つを選んで下さい。鉱物学，地質学，構造，合成，イオン交換，修飾，吸着，触媒，応用（農業，洗剤など），その他），6）連絡先（郵便番号，住所，氏名，電話番号，FAX番号，e-mailアドレス）を申込用紙に記入し，事務局宛にご郵送願います（FAXまたはe-mailでも可能です）。

登録費：会員（主催並びに共催等の学協会の個人会員，およびゼオライト学会団体会員の法人に属する人を含む）5,000円，学生 2,000円，非会員 8,000円（予稿集代を含む。当日申し受けます。）

予稿原稿締切：9月28日（金）（8月中旬に執筆要領をお送りします。）

懇親会：11月21日（水）講演終了後，サンラポーむらくも（島根県民会館より徒歩3分）にて。会費 5,000円（学生 2,000円）

講演申込先：〒305-8565　茨城県つくば市東1-1
産業技術総合研究所物質プロセス研究部門　清住嘉道，TEL. 0298-61-4632, FAX. 0298-61-4631, e-mail: kiyozumi-y@aist.go.jp

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松江市内の主なホテル：アーバンホテル，ニューアーバンホテル本館，ニューアーバンホテル別館，松江ワシントンホテル，松江東急イン

天然ゼオライト鉱床と石見銀山の探訪

エクスカーションとして，日本最大級の産出量をもつ島根県産ゼオライト鉱床，世界遺産に指定が予定されている石見銀山，最近古代神殿の柱が発掘された出雲大社を見学するツアーを予定しています（最少催行人員：15名）。奮ってご参加ください。

日程：11月23日（金）松江駅（8:30）－天然ゼオライト鉱床－石見銀山－出雲大社－JR出雲市駅（16:40）－出雲空港（17:15）－JR松江駅（18:00）
参加費 5,000円（学生 2,000円）

旅費援助候補者の応募について

本学会では例年と同様に，若手会員諸氏の優れた研究発表を奨励するため，旅費の援助をおこないます。旅費の援助を希望される方は下記の要領でご応募下さい。

— 記 —

応募資格　若手の本学会個人会員または学生会員で講演をおこなう方。援助額　往復旅費実費，ただし，4万円が上限です。採用人数　約5名を予定

応募要領　用紙1枚に氏名，年齢，所属，身分，旅費の概算額，連絡先を記入し，講演申込時（7月20日締切）に上記のゼオライト研究発表会係宛，ご提出下さい。

第17回ゼオライト研究発表会　講演申込書

1) 講 演 題 目	
2) 発 表 者 氏 名 (講演者に○)	
3) 所属機関の略称	
4) 講演の種類 (該当するものに○)	総合研究発表　　一般研究発表
5) 研究分野 (該当するものに○)	鉱物学　地質学　構造　合成 イオン交換　修飾　吸着　触媒 応用　その他
6) 連絡先	住所　〒
	氏名
	TEL.
	FAX.
	E-mail:

＊コピーしてお使い下さい。

2001年度第1回研究会
― 次世代のゲート絶縁膜 ―

主　催：日本表面科学会関西支部
協　賛：ゼオライト学会ほか
日　時：平成13年6月29日（金）13:00～17:30
会　場：松下電器技術館 セミナールーム
〒570-8501 大阪府守口市八雲中町3-1-1
TEL: 06－6906-4801
プログラム：
13:00～14:00　松下技術館見学会
14:00～14:35　高信頼酸窒化膜の形成（三菱電機）梅田浩司，寺本章伸，大野吉和
14:35～15:10　STMによるシリコン酸化膜／基板界面のサブナノメータスケール解析（大阪大学）岩崎 裕
15:10～15:45　Zr系ゲート絶縁膜のMIS構造の形成と評価（東芝）山口 豪
16:00～16:35　Zr，HfおよびLanthanoid酸化物High-kゲート絶縁膜の作製（大阪大学）奥山雅則
16:35～17:25　〈特別講演〉 High-kゲート絶縁膜の現状と課題（松下電子）丹羽正昭
参加費：無料
申込締切：平成13年6月22日（金）
定　員：50名
申込及び問合せ先：
木村健二，〒606－8501 京都市左京区吉田本町 京都大学大学院工学研究科 機械物理工学専攻
TEL/FAX: 075-753-5253
e-mail: kimura@kues.kyoto-u.ac.jp

第14回イオン交換セミナー
「ナノテクノロジーとイオン交換材料」

主　催：日本イオン交換学会
協　賛：ゼオライト学会ほか
日　時：2001年7月19日（木）13:00～17:00
場　所：東京工業大学百年記念館フェライト会議室
東急目黒線，大井町線大岡山駅下車
プログラム：
高分子微粒子の合成と構造形成（東工大）石津浩二
高分子微粒子の製造と最近の動向（総研化学）佐藤雅裕
高分子微粒子を利用した荷電モザイク膜（大日精化）杉戸善文
合成ペプチドイオンチャネルの構造と機能（産総研）樋口真弘
多孔質ケイ酸塩によるイオン交換特性（東学大）國仙久雄
有機－無機複合体からの金属酸化物多孔体の合成とその生成機構（千葉大工）高橋亮治
参加申込締切：7月6日（金）
参加申込方法：1）申込者氏名，2）会員（協賛学会のかたは所属学会名），非会員，学生の区別，3）申込者連絡先（所属部課，所在地，電話番号，FAX番号，電子メールアドレス）をご記入の上，FAXまたは電子メールで下記宛にお申込下さい。
〒194-8543 町田市東玉川学園3-3165 昭和薬科大学分析化学研究室内 日本イオン交換学会事務局
e-mail: n-suzuki@ac.shoyaku.ac.jp
FAX: 042-721-4510
参加費（要旨集を含む）：会員 10,000円，学生1,000円，会員外 15,000円（予約外は2,000円増。ただし学生を除く）
参加費支払：郵便振替，加入者番号 00120-2-155043，加入者名「イオン交換セミナー」（企業の方は参加者の個人名を明記して下さい）

第45回粘土科学討論会のお知らせ

第45回粘土科学討論会を下記の要領にて開催いたします。皆様の参加をお待ち致します。
期　日：平成13年9月13日（木）・14日（金）
主　催：日本粘土学会
共　催：ゼオライト学会ほか
会　場：東洋大学朝霞キャンパス（2号館）
埼玉県朝霞市岡2-11-10
日　程：
9月13日
9：00-12：00　口頭発表（2会場）
13：00-13：45　特別講演
13：50-17：50　須藤俊男先生シンポジウム

18：00～ 懇親会（朝霞キャンパス内）

9月14日

9：00-11：00 口頭発表（2会場）

11：00-12：00 日本粘土学会総会

12：00-15：00 ポスター討論

15：00-17：00 口頭発表

特別講演：生沼 郁（東洋大学経済学部教授）「須藤先生と日本粘土学会の足跡と将来」

須藤俊男先生メモリアルシンポジウム：「21世紀の粘土科学 粘土科学の過去・現在・未来－21世紀への跳躍と夢」

講演申込締切：6月22日（金）

講演要旨締切：7月27日（金）

連絡先：東洋大学経済学部社会経済システム学科 西山 勉，TEL: 048-468-6631（実験室）または048-468-6721（研究室），FAX: 048-468-6790（2号館），e-mail: nishiyam@toyonet.toyo.ac.jp

交通と宿泊：会場までは池袋駅から東武東上線で急行15分，朝霞台駅下車，徒歩10分。またはJR武蔵野線北朝霞駅下車，徒歩10分。宿泊は池袋界隈が便利かと思われます。ホテル名などの案内は省略させていただきます。

日本イオン交換学会・日本溶媒抽出学会連合年会
第17回日本イオン交換研究発表会
第20回溶媒抽出討論会

主 催：日本イオン交換学会，日本溶媒抽出学会

協 賛：ゼオライト学会ほか

日 時：平成13年10月25日（木）～26日（金）

場 所：東北大学工学部 青葉記念会館
〒980-8578 仙台市青葉区荒巻字青葉

懇親会：10月25日（木）青葉記念会館3F食堂

講演申込締切：7月6日（金）郵送，FAX，電子メール

講演要旨締切：9月14日（金）郵送

講演申込方法：1）題目，所属，発表者（講演者に○印），2）申込者氏名，3）申込者連絡先（所属部課，所在地，TEL，FAX，電子メールアドレス），4）100字程度の講演概要，5）英文による題目，氏名，所属，6）発表区分（イオン交換，溶媒抽出のいずれか），7）発表様式（口頭またはポスター）を明記し下記事務所宛お申込み下さい。講演申込者には，執筆要項を送付致します。講演時間は，口頭（12分，質疑3分），ポスター（90分）の予定で，口頭発表はOHPに限ります。なお，口頭発表の希望でも発表件数多数の場合には，ポスター発表に変更させて頂くことがあります。

参加費：一般 6,000円，学生 2,000円（予約申込者は1,000円割引，非会員は予約外扱い）

懇親会費：予約5,000円，当日6,000円

予約申込締切：平成13年10月19日（金）

予約申込方法：1）氏名，2）勤務先名称，3）連絡先所在地，所属部課，TEL，FAX，電子メールアドレス，4）懇親会参加の有無をお書きの上，電子メール，FAXまたは郵便で下記の事務所宛にお送り下さい。

申込先：〒980－8577 仙台市青葉区片平2-1-1 東北大学 多元物質科学研究所，三村 均（TEL: 022-217-5142，FAX: 022-217-5142，e-mail: rengou@iamp.tohoku.ac.jp）

参加費支払方法：郵便振替，加入者番番号 00130-0-119845，加入者名「イオン交換研究発表会係」（企業の方は参加者の個人名を明記して下さい）

URL http://www.anal.chem.tohoku.ac.jp/ionex/ionex.html

3rd International Congress on Environmental Catalysis (3rd ICEC)

December 10-13, 2001
International Conference Center of Waseda University,
Shinjuku, Tokyo, Japan

The "3rd International Conference on Environmental Catalysis" (3rd ICEC) will be held in December 2001, at Waseda University in Tokyo. This conference follows the very successful first meeting in Pisa (1995) and the second one in Miami Beach (1998), with the scope aiming at promoting a global and interdisciplinary approach to catalysis for a better environment and quality of life. This basic theme will be continuing at the 3rd ICEC, with a full 4-day program of oral and poster sessions in a single session format.

TOPICS

- Catalysis for atmospheric environment: mobile engine exhaust, NOX, SO_2, particulates, VOC
- Catalysis for hydrospheric environment: BOD/COD, NH_3/NO_3
- Catalysis for waste detoxification and recycling: thermal/material recycling of plastics, spent catalysts
- Catalysis for global environment: CO_2, fluorocarbons, N_2O
- Catalysis for green and sustainable chemistry
- Catalysis for reduction of hazardous chemicals and endocrine disruptors: chlorinated hydrocarbons
- Catalysis for clean fuel production: desulfurization of petroleum, H_2-production, methane activation, MTBE/DME, biomass conversion

SCIENTIFIC PROGRAM

In the respective areas, there will be presentations by invited speakers and by general papers in oral or poster sessions. Theselection of papers into either oral or poster will be based on extended abstracts.

REGISTRATION

Those who plan to attend the 3rd ICEC are to fill in the Registration Form, which will be attached in the final circular or will be down loaded from here, and return it by October 1, 2001.

Nippon Travel Agency Co., Ltd has been appointed as the official travel agent for the Conference and will handle hotel accommodations.

Please contact:

Nippon Travel Agency Co., Ltd., International Travel Dept.

3rd Fl. Shimbashi Ekimae Bldg. #1, 2-20-15

Shimbashi, Minato-ku, Tokyo 105-8606, Japan

Tel: +81-3-3572-8743 Fax: +81-3-3572-8689

PROCEEDINGS

The Proceedings of the Conference will be published after a scientific review as a special issue of Applied Catalysis B: Environmental, which will include invited lectures and oral papers. Those who wish publish a paper in the proceedings hould submit their manuscripts by the first day of the conference. The "Guide for Authors" can be found on the web site of Elsevier.

LANGUAGE

All abstracts, papers and presentations must be in English.

ORGANIZED BY

Environmental Catalysis Forum of Japan

UNDER THE AUSPICES OF

Catalysis Society of Japan

Research Institute of Innovative Technology for the Earth

The Chemical Society of Japan

The Japan Petroleum Institute

Waseda University

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J. Weitkamp (Germany)
B. Wichterlova (Czech Republic)

KEY DATES

Distribution of final circular: August 1, 2001

Deadline for early registration: October 1, 2001

Full manuscript deadline: December 10, 2001

secretary, 3rd ICEC

Professor Kohichi Segawa

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13th International Zeolite Conference

Montpellier, France
July 8 - 13, 2001

Ogranized
under the Auspices of IZA, GFZ, FEZA

The Organizing Committee and the International Zeolite Association (IZA), with the participation of the French Zeolite Group (GFZ) and the Federation of the European Zeolite Associations (FEZA), address a cordial invitation to participate in the 13th International Zeolite Conference (13th IZC) which will be held from Sunday, July 8, to Friday, July 13, 2001 in Montpellier, France. The Conference will be preceded by a three-day Summer School on Zeolites in Poitiers and followed by a three-day Field Trip to natural zeolite localities in the Massif Central.

ORGANIZING COMMITTEE

GENERAL CHAIRMAN François Fajula

SECRETARY Francesco Di Renzo
ENSCM, 8 rue Ecole Normale,
34296 Montpellier cedex 5, France
Tel. +33-0-467-14-43-23
fax. +33-0-467-14-43-49
e-mail: izc13@argon.enscm.fr

PRE-CONFERENCE SCHOOL

July 5 - 7, 2001

The Pre-Conference School will be held from Thursday, July 5, to Saturday, July 7, 2001, on the Campus of the University of Poitiers (South West of Paris, 1 h 30 by train). Poitiers is famous for his historical heritage, monuments, cellars (wines and spirits : Pineau, Cognac, etc.) and culinary tradition as well as for its architectural and artistic wealth (gallo-roman remains, gothic architecture, roman and medieval art). Its university is one of the oldest in France (1431). Attendees will be housed in residence halls. Fees (3000 FRF, 3300 FRF after April, 15, 2001) include accommodations for 4 nights, meals, travel by train from Poitiers to Montpellier (Sunday, July 8), lecture notes, etc... Attendance will be limited to 100.

TOPICS AND LECTURES

"Process Chemistry", *A. Corma* (Valencia)
Preparation of Zeolite Catalysts, *T. Roberie* (Grace Davison)
Refining Processes : Setting the Scene, *R. Jensen* (UOP)
Catalytic Cracking, *T. Habib* (Grace Davison)
Hydrocracking, *R. van Veen* (Shell)
C4-C6 Alkane Isomerization, *F. Schmidt* (Sud-Chemie)
Base Oil Production and Processing, *M. Daage* (Exxon Mobil)
Aromatics Alkylation. Towards Clean Processes, *F. Beck* (Exxon Mobil)
Paraxylene Preparation. Catalysis Processes, *F. Alario* (IFP)
Separation of Paraxylene, *A. Methivier* (IFP)
Methanol to Olefins (MTO) and Beyond, *P. Barger* (UOP)
Phenol Hydroxylation and Related Oxidations, *G. Bellussi* (ENI)
Fine Chemical Synthesis - Setting the Scene,
J. De Voos, P. Jacobs (KU Leuven)
Aromatics Functionalization, *S. Ratton* (Rhodia)
Fragrance Synthesis, *W. Hoelderich* (Aachen)
Pollution Abatment, *B. Coq* (Montpellier)
Round Table "Future Trends in Zeolite Applications"

FOR ADDITIONAL INFORMATION, please contact **Michel Guisnet** :
Chimie, Université de Poitiers,
40, av. du Recteur Pineau, 86022 Poitiers Cedex, France
Tel. +33-5-49-45-39-05
Fax. +33-5-49-45-37-79-
e-mail: michel.guisnet@univ-poitiers.fr

FIELD TRIP

July 14 - 16, 2001

The field trip will consist in the visit to several sites of geological and mineralogical interest, providing an overview of the hydrothermal occurrences of zeolites in the French Massif Central. The participants will leave from the Conference Center at 17.00 on friday 13 July by bus and will be back at Montpellier at 19.00 on monday 16 July. Hotel accomodations, breakfasts, sandwich lunches, restaurant dinners, guided visits and bus travel are included. The participants' luggage will be taken care of in the bus.

The field trip will include rock-hammering and mineral collecting, as well as the visit to active and disaffected quarries. These activities cannot be considered as completely danger-free, despite the efforts of the organizers to avoid every objective factor of risk. The participants are required to strictly abide safety regulations and to check that all risks are covered by their professional or private insurance. Accompanying persons have to comply with these conditions as normal participants. Some of the sites are only accessible by a short scramble. Please check that your fitness allows you some physical exertion. The participation fees are 2800 French Francs, or 426.86 Euros, per participant.

Most sites cannot accomodate in a safe way a large number of people. As a consequence, the number of participants is limited to 60. You are strongly advised to contact the secretary of the field trip subcommittee (Alain.Tuel@catalyse.univ-lyon1.fr) before registering to be sure that places are still available. In the case you had registered for the field trip after the completion of the participants' list, you will be promptly made aware of your unsuccessful application and will be reimbursed of the field trip fees at your arrival at the conference center.

A document on the geology and mineralogy of the visited sites will be provided to the participants at the conference center. If you are unable to bring your own geologist hammer and safety goggles, please inform the organizers, who will be able to provide these items at a non-profit fare.

SCIENTIFIC PROGRAMME

The scientific programme will include 5 plenary and 6 keynote lectures, 146 oral and about 540 posters presentations, plus a still unknown number of recent research reports. The conference presentations have been organised in 32 technical sessions.

PLENARY LECTURES

on topics of wide interest will be presented by leading experts upon invitation by the Organizing Committee. The plenary lecturers include

PL-1- Monday 9h Ordered mesoporous materials - State of art and prospects (*F. Schüth*)
PL-2- Tuesday 8h30 Clinoptilolite-heulandite: applications and basic research (*T. Armbruster*)
PL-3- Wednesday 8h30 Evolution of extra-large pore materials (*M.E. Davis*)
PL-4- Thursday 8h30 Evolution of refining and petrochemicals. What is the place of zeolites? (*C. Marcilly*)
PL-5- Friday 8h30 Is electron microscope an efficient magnifying glass for micro- and meso- porous materials? (*O. Terasaki and T. Oshuna*)

KEYNOTE LECTURES

Six keynote lecturers have been selected by the Paper Selection Committee.

23-K-01 - Monday 16h20 Delaminated zeolites as active catalysts for processing large molecules (*A. Corma and V. Fornés*)
01-K-01 - Tuesday 9h50 Pentasil zeolites from antartica: from mineralogy to zeolite science and technology (*A. Alberti, G. Cruciani, E. Galli, S. Merlino, R. Millini, S. Quartieri, G. Vezzalini and S. Zanardi*)
19-K-01 - Tuesday 16h20 Use of 1H NMR imaging to study the diffusion and co-diffusion of gaseoushydrocarbons in HZSM-5 catalysts (*P. N'Gokoli-Kekele, M.-A. Springuel-Huet, J.-L. Bonardet, J.-M. Dereppe and J. Fraissard*)
21-K-01 - Wednesday 9h50 Zeolite-based nanocomposites: synthesis, characterization and catalytic applications (*B.V. Romanovsky*)
03-K-01 - Thursday 16h20 Application of combinatorial tools to the discovery and commercialization of microporous solids: facts and fiction (*J. Holmgren, D. Bem, M. Bricker, R. Gillespie, G. Lewis, D. Akporiaye, I. Dahl, A. Karlsson, M. Plassen and R. Wendelbo*)
30-K-01 - Friday 9h50 The local structures of transition metal oxides incorporated in zeolites and their unique photocatalytic properties (*M. Anpo and S. Higashimoto*)

Oral Presentations will be organized in four parallel sessions in adjacent meeting rooms. Special care has been taken to synchronise parallel sessions.

Poster Presentations Four one-day poster sessions will be organized. The posters will be on display in the Conference Main Hall where coffee breaks and exhibitions will be held. Plenty of time and space will be available for discussions of the content of the posters.

TECHNICAL SESSIONS

01 Mineralogy of Natural Zeolite

01-K-01 Pentasil zeolites from antartica: from mineralogy to zeolite science and technology, *A. Alberti, G. Cruciani, E. Galli, S. Merlino, R. Millini, S. Quartieri, G. Vezzalini and S. Zanardi*

01-O-02 Natural zeolites mineralization in the Oligocene-Miocene volcano-sedimentary succession of Central Sardinia (Italy), *P. Cappelletti, G . Cerri, M. de Gennaro, A. Langella, S. Naitza, G. Padalino, M. Palomba and R. Rizzo*

01-O-03 Cation location and its influence on the stability of clinoptilolite, *M.N. Johnson, G. Sankar, C.R.A. Catlow, D. O'Connor, P. Barnes and D.Price*

01-O-04 The structure of Li-phillipsite, *A.F. Gualtieri*

01-O-05 Ion-exchange features of intermediate-silica sedimentary phillipsite, *C. Colella, E. Torracca, A. Colella, B. de Gennaro and D. Caputo and M. de Gennaro*

01-P-06 Zeolites in impact craters, *M.V. Naumov*

01-P-07 Al ordering in a dachiardite framework, *M. Kato and K. Itabashi*

01-P-08 Chemical composition and ion-exchange properties of a natrolite from Zahedan Region, Iran, *A.R. Sardashti, H. Kazemian and M. Akramzadeh Ardakani*

01-P-09 Physical, chemical and structural characterization of the volcanic tuff from the Maramures area, Romania, *R. Pode, G. Burtica, S. Herman, A. Iovi and I. Calb*

01-P-10 Heulandite group zeolites from the Paleogene fresh water lake Blateshnitza Graben, Southwest Bulgaria, *Z. Milakovska, E. Djourova and R. Tzankarska*

01-P-11 Isodimorphism of templates in zeolites. New crystal chemistry of analcime and its analogues, *V.V. Bakakin*

01-P-12 Evaluation of clinoptilolite tuffs from Russia as ion exchangers using NH_4 ions, *I.V. Komarova, N.K. Galkina, V.A. Nikashina, B.G. Anfilov and K.I. Sheptovetzkaya*

01-P-13 Mineralogy, chemistry and ion-exchange properties of the zeolitized tuffs from the Sheinovets caldera, E Rhodopes (South Bulgaria), *R. Ivanova, Y. Yanev, Tz. Iliev, E. Koleva, T. Popova and N. Popov*

01-P-14 Synthesis of titanium, niobium, and tantalum silicalite-1 by microwave heating of the mixed oxide xerogel precursors, *W.S. Ahn, K.Y. Kim, M.H. Kim and Y.S. Uh*

01-P-15 Different silver states stabilized in natural clinoptilolites, *N. Bogdanchikova, B. Concepcion Rosabal, V. Petranovskii, M. Avalos-Borja and G. Rodríguez-Fuentes*

01-P-16 Physical-chemical and adsorptive properties of Armenia natural zeolites, *F. Grigoryan, A. Hambartsumyan, H. Haroyan and A. Karapetyan*

01-P-17 The sorption equilibria in natural zeolite-aqueous solutions systems, *J. Peric, M. Trgo and S. Cerjan-Stefanovic*

02 Zeolite Nucleation and Growth

02-O-01 Small angle X-ray scattering on TPA-Silicalite-1 precursors in clear solutions: influence of silica source and cations, *C.J.Y. Houssin, B.L. Mojet, C.E.A. Kirschhock,V. Buschmann, P.A. Jacobs, J.A. Martens and R.A. van Santen*

02-O-02 Nucleation processes in zeolite synthesis revealed through the use of different temperature-time profiles, *C.S. Cundy, J.O. Forrest and R.J. Plaisted*

02-O-03 High yield synthesis of colloidal crystals of faujasite zeolites, *Qinghua Li, D. Creaser and J. Sterte*

02-O-04 Colloid chemical properties of silicalite-1 nanoslabs, *S. Kremer, C. Kirschhock, P. Rouxhet, P.A. Jacobs and J.A. Martens*

02-O-05 Atomic force microscopy (AFM) used to relate surface topography growth mechanisms in SSZ-42, *M.W. Anderson, N. Hanif, J.R. Agger, C.-Y. Chen and S.I. Zones*

02-P-06 Epitaxial overgrowth of MAZ onto EMT type zeolite crystals, *A.M. Goossens, V. Buschmann and J.A. Martens*

02-P-07 The transformation of zeolite A and X into nitrate cancrinite under low temperature hydrothermal reaction conditions, *J.C. Buhl and C. Taake*

02-P-08 Comparison of crystal linear growth rates for silicalite-1 in thermal and microwave syntheses, *C.S. Cundy and J.O. Forrest*

02-P-09 Effect of initial hydrogel milling on Na-ZSM-5 synthesis, *C. Falamaki, M. Edrissi and M. Sohrabi*

02-P-10 Synthesis and characterization of zeolite ZSM-25, *S.B. Hong, W.C. Paik, W.M. Lee, S.P. Kwon, C.-H. Shin, I.-S. Nam and B.-H. Ha*

02-P-11 A study on the crystallization of a lamellar aluminophosphate APO-M to a three-dimensional aluminophosphate APO-CJ3, *K. Wang, J. Yu, Y. Song, Y. Zou and R. Xu*

02-P-12 Synthesis of nanosized offretite crystals, *J. Hedlund and E. Kurpan*

02-P-13 Silicon oxide plays a driving role in the synthesis of microporous SAPO-11, Z-*Q. Liu and R. Xu*

02-P-14 Synthesis of nanocrystal zeolite Y and its host effect, *H. Yang, R. Li, B. Fan and K. Xie*

02-P-15 Tailoring crystal size and morphology of zeolite ZSM-5, *Ming Liu and S. Xiang*

02-P-16 Modeling of silicalite crystallization from clear solution, *K.A. Carlsson, J. Warzywoda and A. Sacco, Jr.*

02-P-17 Interaction/synergistic effect of Mg^{2+} and Ba^{2+} on the size and morphology of the zeolite L crystals, *S. Ferchiche, J. Warzywoda and A. Sacco, Jr.*

02-P-18 In-situ NMR study of mechanisms of zeolite A formation, *M. Smaihi, S. Kallus and J.D.F. Ramsay*

02-P-19 Effects of synthesis parameters on zeolite L crystallization, *Y.S. Ko, S.H. Chang and W.S. Ahn*

02-P-20 Some aspects of NU-86 zeolite crystallization, *S.V. Dudarev, A.V. Toktarev, G.V. Echevsky, C.L. Kibby and D.J. O'Rear*

02-P-21 Synthesis of zeolite Sr,K-ZK-5, *P.C. Russell, S.L. Stuhler, A.L. Kouli, J. Warzywoda and A. Sacco, Jr.*

02-P-22 Evidence for in-situ directing agent modification in zeolite syntheses, *J.C. Vartuli, G.J. Kennedy, B.A. Yoon and, A. Malek*

02-P-23 The influence of different silica sources on the crystallization kinetics of zeolite Beta, *W. Schmidt, A.V. Toktarev, F. Schüth, K.G. Ione and K. Unger*

02-P-24 Population balance: a powerful tool for the study of critical processes of zeolite crystallization, *B. Subotic, T. Antonic and J. Bronic*

02-P-25 Synthesis of TMA-SOD from a novel type layered silicate by solid state transformation, *Y. Kiyozumi, F. Mizukami, Y. Akiyama, T. Ikeda and T. Nishide*

02-P-26 Direct conversion of bulk-materials into MFI zeolites by a bulk-material dissolution technique, S. *Shimizu and H. Hamada*

02-P-27 Synthesis of a new microporous silicate using DABCO-based structure-directing agent, *Y. Kubota, J. Plévert, T. Honda, P. Wagner, M.E. Davis, T. Okubo, Ya. Goto,, Y. Fukushima and Y. Sugi*

02-P-28 Heteroepitaxial connection of zeolites with different pore structures, *T. Wakihara, J. Plévert, S. Nair, M. Tsapatsis, S. Yamakita, Y. Ogawa, H., Komiyama, M. Yoshimura, M.E. Davis and, T. Okubo*

02-P-29 Study of zeolite A crystallization from clear solution by hydrothermal synthesis and population balance simulation, *J. Bronic, P. Frontera, F. Testa, B. Subotic, R. Aiello and J. B.Nagy*

02-P-30 Synthesis of zeolite SSZ-35 using N-methyl hexahydro-julolidinium salt as a new family of structure-directing agents (SDAs), *Y. Kurata, T.-A. Hanaoka and H. Hamada*

02-P-31 The Fitting equation for zeolite crystallization with seeds, *V. Toktarev and S.V. Dudarev*

02-P-32 Influence of the thermal treatment of the aluminosilicate gel precursor on the zeolite nucleation, *C. Kosanovic, B. Subotic and D. Kralj*

02-P-33 Efficient co-templating roles of amines and amides admixed with alkylammonium salts for the stabilisation of new $AlPO_4$-n topologies, *C. Borges, M.F. Ribeiro, C. Henriques, M.T. Duarte, J.P. Lourenço and Z. Gabelica*

02-P-34 Static synthesis of zeolite MCM-22, *Y.-M. Wang, X.-T. Shu and M.-Y. He*

02-P-35 Synthesis of pure silica Beta by the conventional hydrothermal method, *W. Guo, J. Yao, Y. Luo and Qi. Li*

02-P-36 Hydrothermal transformation of a layered silicate, Na-magadiite, into mordenite zeolite, *T. Selvam and W. Schwieger*

02-P-37 In-situ diagnostic of zeolite crystal growth by real time ultrasound monitoring, *R. Herrmann, W. Grill, T.J. Kim, O. Scharf, R. Schertlen, M. Schmachtl, W. Schwieger, C. Stenzel, H. Toufar and Y. Venot*

02-P-38 Effect of ageing on the decomposition of tetra-alkylammonium ions as studied by microwave heating, *A. Arafat, H. van Bekkum, Th. Maschmeyer, and J.C. Jansen*

02-P-39 Synthesis of siliceous mordenite from system free of amine, *X. Qi, X. Liu and Z. Wang*

02-P-40 High-resolution solid state MAS NMR studies on the role of promoter (phosphate) in the nucleation and crystallization of Silicalite-1 (Si-MFI), *P.R. Rajamohanan, P. Mukherjee, S. Ganapathy and R. Kumar*

02-P-41 The influence of concentration on the structure-directing effects of diethylenetriamine in the synthesis of porosils, *P. Behrens, V.J. Hufnagel and A.M. Schneider*

02-P-42 Synthesis of high-silica MWW zeolite, *L.M. Vtjurina, S.S. Khvoshchev and I.V. Karetina*

03 New Methods of Zeolite Synthesis

03-K-01 Application of combinatorial tools to the discovery and commercialization of microporous solids: facts and fiction, *J. Holmgren, D. Bem, M. Bricker, R., Gillespie, G. Lewis, D. Akporiaye, I. Dahl, A. Karlsson, M. Plassen and R. Wendelbo*

03-O-02 Mesoporous zeolites, *C.J.H. Jacobsen, J. Hou*vicka, A. Carlsson and I. Schmidt*

03-O-03 Synthesis of novel zeolites SSZ-53 and SSZ-55 using organic templating agents derived from nitriles, *S.A. Elomari and S.I. Zones*

03-O-04 Synthesis of IFR-type zeolites optimized for spectroscopic study, *B.S. Duersch and L.W. Beck*

03-O-05 Competitive role of sodium and potassium cations during hydrothermal zeolite crystallization from Na_2O-K_2O-Al_2O_3-SiO_2-H_2O gels, *A.F. Ojo, F.R. Fitch, M. Bülow and M.-L. Lau*

03-P-06 Zeolitization of a spanish bentonite in seawater medium. Effect of alkaline concentration and time, *R. Ruiz, C. Blanco, C. Pesquera and F. González*

03-P-07 Silicalite-1 spheres prepared from preformed resin-silicate composites, *L. Tosheva and J. Sterte*

03-P-08 The synthesis of offretite single crystals using pyrocatechol as complex agent, *F. Gao, G. Zhu, Xiaotian Li, S. Qiu, B. Wei, C. Shao and O. Terasaki*

03-P-09 Synthesis of FER type zeolite in presence of tetrahydrofuran, *G.-Q. Guo, Y.-J. Sun and Y.-C. Long*

03-P-10 Utilization of dry-gel conversion method for the synthesis of gallosilicate zeolites Beta, ZSM-5 and ZSM-12, *R. Bandyopadhyay, Y. Kubota, S.i Nakata and Y. Sugi*

03-P-11 A novel method for the synthesis of cancrinite type zeolites, *C.F. Linares, S. Madriz, M.R. Goldwasser, and C. Urbina de Navarro*

03-P-12 High-throughput strategies for the hydrothermal synthesis of zeolites and related materials, *N. Stock, N. Hilbrandt, K. Choi and T. Bein*

03-P-13 Static zeolite MCM-22 synthesis using two-level factorial design, *J. Warzywoda, S. Dumrul, S. Bazzana and A. Sacco, Jr.*
03-P-14 Influence of nano-particle agglomeration on the catalytic properties of MFI zeolite, *S. Inagaki, I. Matsunaga, E. Kikuchi and M. Matsukata*
03-P-15 Rapid and mass production of porous materials using a continuous microwave equipment, *D.S. Kim, J.M. Kim, J.-S. Chang and S.-E. Park*
03-P-16 Hydrothermal synthesis of vanadium-containing microporous aluminophosphates via the design of experiments approach, *L. Frunza, P. Van Der Voort, E.F. Vansant, R.A. Schoonheyd and B.M. Weckhuysen*
03-P-17 Synthesis of a thin Silicalite-1 membrane, through sintering, for use in a membrane reactor, *E.E. McLeary, A.W. Hoogesteger, R.D. Sanderson and J.C. Jansen*
03-P-18 Mixed alkali templating in the Si/Al = 3 and 10 systems: a combinatorial study, *G.J. Lewis, D.E. Akporiaye, D.S. Bem, C. Bratu, I.M. Dahl, A. Karlsson, R.C. Murray, R.L. Patton, M. Plassen and R. Wendelbo*
03-P-19 Factors affecting composition and morphology of mordenite, *F. Hamidi, M. Pamba, A. Bengueddach, F. Di Renzo and F. Fajula*

04 Isomorphous Substitutions

04-O-01 Direct synthesis of Cu(I)-MFI zeolite in the presence of Cu(II) methylamino complexes as mineralizing and reducing agents, *S. Valange, F. Di Renzo, E. Garrone, F. Geobaldo, B. Onida and Z. Gabelica*
04-O-02 Preparation and catalytic properties of a novel type of zeolites with basic properties, *S. Ernst, M. Hartmann and S. Sauerbeck*
04-O-03 Preparation and characterization of iron-substituted zeolites, *G. Giordano, A. Katovic, A. Fonseca and J.B. Nagy*
04-O-04 Influence of the nature of T atoms on the morphology and crystal size of KZ-2 and ZSM-22 zeolites isomorphously substituted with Al or Fe, *M. Derewinski, M. Kasture, J. Krysciak and M. Stachurska*
04-O-05 Synthesis and characterisation of novel large-pore vanadosilicates AM-13 and AM-14, *P. Brandão, A. Philippou, N. Hanif, J. Rocha and M. Anderson*
04-P-06 Uniform distribution of nickel during the synthesis of Si-ZSM-5 through solid-state transformation, *M. Salou, Y. Kiyozumi, F. Mizukami, S. Niwa, M. Imamura and M. Haneda*
04-P-07 Synthesis, characterization and catalytic activity of FeBEA and FeMFI zeolite obtained by xerogel wetness impregnation, *O.A. Anunziata, L.B. Pierella, E.J. Lede, and F.G. Requejo*
04-P-08 A novel method for the synthesis of chromium aluminosilicate with BEA structure, *X.-H. Tang, L.-R. Pan, J.-Z. Wang and H.-X. Li*
04-P-09 Pure SAPO, CoAPSO and ZnAPSO ATO-like molecular sieves through optimized synthesis procedures, *A. Azzouz, N. Bilba, M. Attou, A. Zvolinschi and S. Asaftei*
04-P-10 Relation between amount of the niobium ammonium complex in the reaction mixture and the crystal size of an Nb- MFI zeolite, *H. Munhoz Jr., S. Rodrigues, P.K. Kiyohara and W. Sano*
04-P-11 Spectroscopy of the formation of microporous transition-metal ion containing aluminophosphates under hydrothermal conditions, *B.M. Weckhuysen, D. Baetens and R.A. Schoonheydt*
04-P-12 Co-templated synthesis of CrAPO-5 with various organic acids, *J. Kornatowski, G. Zadrozna, J.A. Lercher, and M. Rozwadowski*
04-P-13 How to increase the amount of framework Co^{2+} in microporous crystalline aluminophosphates?, *W. Fan, R.A. Schoonheydt and B.M. Weckhuysen*
04-P-14 Preparation of zinc containing zeolite catalysts, A. Katovic, *E. Szymkowiak, G. Giordano, S. Kowalak, A. Fonseca and J. B.Nagy*
04-P-15 Synthesis of Zn and Fe substituted mordenite using citric acid as complexing agent, *M. Dong, J.-G. Wang and Y.-H. Sun*
04-P-16 ET(Zr)S-4 molecular sieve: kinetic and morphological characterization, *D. Vuono, P. De Luca, A. Fonseca, J. B.Nagy and A. Nastro*
04-P-17 Influence of alcali cations on the incorporation of iron into MFI structure in fluoride media, *F. Testa, F. Crea, R. Aiello, K. Lázár, P. Fejes, P. Lentz and J. B.Nagy*
04-P-18 Synthesis and characterization of Co-containing zeolites of MFI structure, *E. Nigro, F. Testa, R. Aiello, P. Lentz, A. Fonseca, A. Oszko, P. Fejes, A. Kukovecz, I. Kiricsi and J. B.Nagy*

05 Synthesis of New Materials

05-O-01 SOMS: Sandia Octahedral Molecular Sieves. A new class of ion exchangers selective for the removal of Sr^{2+} from waste streams, *T.M. Nenoff, M. Nyman, A. Tripathi, J.B. Parise, W.T.A. Harrison and RS. Maxwell*
05-O-02 Hydrothermal synthesis of various titanium phosphates in the presence of organic amine templates, *Yunling Liu, Y. Fu, J. Chen, Y. Zou and W. Pang*
05-O-03 On the role of azamacrocycles and metal cations in the syntheses of metalloaluminophosphates STA-6, -7 and -8, *R. Garcia, E.F. Philp, A.M.Z. Slawin, P.A. Wright and P.A. Cox*
05-O-04 Chiral transference and molecular recognition in novel $Co(en)_3Cl_3$-templated zinc phosphates, *J. Yu, Yu Wang, Z. Shi and R. Xu*
05-O-05 Very open microporous materials: from concept to reality, *A.K. Cheetham, H. Fjellvåg, T.E. Gier, K.O. Kongshaug, K.P. Lillerud and G.D. Stucky*
05-P-06 Synthesis and structures of GIS, ABW and GME beryllophosphate molecular sieves from amine solutions, *H. Zhang, M. Chen, Z. Shi, Y. Zhou, Xin, Xu and D. Zhao*
05-P-07 Microporous gallosilicate TNU materials and their implications for the synthesis of low-silica molecular sieves, *W.C. Paik, M.A. Camblor and S.B. Hong*
05-P-08 Synthesis and characterization of novel nickel phosphates from non-aqueous systems, *Yunling Liu, L. Zhang, P. Zhang, Y. Zou and W. Pang*
05-P-09 Synthesis, characterization and properties of an anionic aluminophosphate molecular sieve with Brönsted acidity, *W. Yan, J. Yu, R. Xu, Y. Han, K. Sugiyama and O. Terasaki*
05-P-10 Synthesis and characterization of an open-framework aluminophosphate $[AlP_2O_6(OH)_2][H_3O]$ containing propeller-like chiral motifs, *W. Yan, J. Yu, Z. Shi and R. Xu*
05-P-11 Synthesis and characterization of an aluminum-substituted manganese phosphate with GIS topology, *H.-M. Yuan, Y.-S. Jiang, W. Chen, J.-S. Chen and R. Xu*
05-P-12 Synthesis and characterisation of novel microporous framework cerium and europium silicates, *D. Ananias, P. Ferreira, A. Ferreira, J. Rocha, J.P. Rainho, C.M. Morais and L.D. Carlos*
05-P-13 Novel microporous framework stannosilicates, *Z. Lin and J. Rocha*
05-P-14 Magadiite intercalated MCM-22, M. Munsignatti, *A.J.S. Mascarenhas, A.L.S. Marques and H.O. Pastore*
05-P-15 Synthesis of aluminum phosphite microporous materials, *N. Li and S. Xiang*
05-P-16 Synthesis, characterization and structural aspects of novel microporous indium, *L.M. King, J. Gisselquist, S.C. Koster, D.S. Bem, R.W. Broach, S.G. Song and R.L. Bedard*
05-P-17 Synthesis and characterization of the silicoaluminophosphate SAPO-47, *L. Xu, Z. Liu, P. Tian, Y. Wei, C. Sun and Shi. Li*
05-P-18 Synthesis, characterization and catalysis of SAPO-56 and MAPSO-56 molecular sieves, *P. Tian, Z. Liu, L. Xu and C. Sun*
05-P-19 Synthesis and structural characterization of a novel microporous zeolitic type aluminium phosphate, *C. Sassoye, S. Girard, C. Mellot-Draznieks, T. Loiseau, C. Hugeunard, F. Taulelle and G. Férey*
05-P-20 Hydrothermal synthesis and crystal structures of two novel open frameworks: $(enH_2)_3[Co_3W_4P_4O_{28}]$ and $(dapH_2)_3[Co_3W_4P_4O_{28}]$, *B. Yan, Y. Xu, N.K. Goh and L.S. Chia*

06 Fundamentals of Micelle Templating

06-O-01 The effect of stoichiometry and synthesis conditions on the properties of mesoporous M41S family silicates, *W.J. Roth and J.C. Vartuli*
06-O-02 What are circular crystals?, *F. Marlow*
06-O-03 Hierarchically mesostructured zeolitic materials with the MFI structure, *D. Trong On, P. Reinert, L. Bonneviot and S. Kaliaguine*
06-O-04 Pore size engineering of MCM-48: The use of different additives as expanders, *M. Mathieu, E. Van Bavel, P. Van Der Voort and E.F. Vansant*
06-P-05 X-ray diffraction analysis of ordered mesoporous silica, *M. Ookawa, Y. Yogoro, T. Yamaguchi and K. Kawamura*
06-P-06 Active MCM-48 supported catalysts: different strategies to increase the structural and chemical stability, *P. Van Der Voort, M. Mathieu and E.F. Vansant*
06-P-07 Strong acidic and high temperature hydrothermally stable mesoporous aluminosilicates with well-ordered hexagonal structure, *Zo. Zhang, Y. Han, R. Wang, S. Qiu, D. Zhao and F.-S. Xiao*
06-P-08 Studies on the synthesis of Al-MCM-41 mesoporous materials, *G.A. Eimer, L.B. Pierella and O.A. Anunziata*
06-P-09 Controlled synthesis of microporous and mesoporous silica-based molecular sieves in the presence of dodecyldimethyl-benzylammonium chloride, *Z.Y. Yuan, W. Zhou, L.M. Peng, J.-Z. Wang and H.X. Li*
06-P-10 Formation of double-mesopore silica and its transformation into MCM-41, X.-Z. *Wang, T. Dou, Y.-Z. Xiao and B. Zhong*
06-P-11 The synthesis and hydrothermal stability of directly usable hexagonal mesoporous silica by efficient primary amine template extraction in acidified water, *K. Cassiers, P. Van Der Voort and E.F. Vansant*
06-P-12 Synthesis and characterization of highly ordered chromium-substituted MCM-48 materials with tailored pore sizes, *C. Pak and G.L. Haller*
06-P-13 Preparation and characterisation of mesoporous silica spheres, *R. Van Grieken, D.P. Serrano, C. Martos, and A.M. Melgares*
06-P-14 Rapid synthesis of high quality MCM-41 silica via ultrasound irradiation, *X.-H. Tang, Yanquing Wang, W. Huang, S. Liu, O. Palchik, E. Sominski, Y. Koltypin and A. Gedanken*
06-P-15 Synthesis of mesoporous aluminosilicate FSM-materials derived from synthetic and natural Saponite, *T. Linssen , M. Barroudi, P. Cool and E.F. Vansant*
06-P-16 ^{129}Xe NMR and adsorption studies of Si-MCM-48 and Al-MCM-48, *G. Øye, M.-A. Springuel-Huet, J. Fraissard, M. Stöcker and J. Sjöblom*
06-P-17 ^{27}Al-NMR studies on Al-MCM-41 molecular sieves synthesized with different Si/Al ratios and different aluminum sources, *W. Böhlmann and D. Michel*
06-P-18 Control of formation of mesoporous SBA-3 and SBA-1 through organic additives, *S. Che and T. Tatsumi*
06-P-19 The influence of Al, La or Ce in the thermal and hydro-thermal properties of MCM-41 mesoporous solids, *R.A.A. Melo and E.A. Urquieta-González*
06-P-20 Controlling the assembly of silica mesoporous materials by varying the decrease in pH, *J. Rathouskŷ, J. Cejka, P.J. Kooyman, M. Slabová and A. Zukal*
06-P-21 Characterisation of MCM-41 aged for different periods, *H. Al-Megren, T.-C Xiao, A.P.E. York, J. Sloan, S. Al-Khowaiter, S.-F. Ji and M.L.H. Green*
06-P-22 Synthesis and characterization of silica and aluminosilica-surfactant nanocomposites, *E. Popovici, A. Visan, D. Filip,G. Burtica, and R. Pode*
06-P-23 Improved thermal stability of mesoporous alumina support of catalysts for the isomerization of light paraffins, *V. González-Peña, C. Márquez-*

Alvarez, E. Sastre and J. Pérez-Pariente

06-P-24 Synthesis of mesoporous molecular sieves MCM-48 under several reaction conditions, *S. Rodrigues da Rocha and L. Domiciano Fernandes*

06-P-25 Parallel synthesis of mesostructured materials, *P. Behrens and C. Tintemann*

06-P-26 Mesostructural transformation in the presence of fluoride anions, *Q.-H. Xia, K. Hidajat and S. Kawi*

06-P-27 Swelled Micelle-Templated Silicas (MTS): structure control and hydrophobic properties, *D. Desplantier-Giscard, A. Galarneau, F. Di Renzo and F. Fajula*

06-P-28 Synthesis of pure and iron-containing mesoporous silica. Effect of washing and removal of template on the porous structure, *L. Pasqua, F. Testa, R. Aiello, F. Di Renzo, and F. Fajula*

07 New Mesoporous Molecular Sieves

07-O-01 Ordered mesoporous carbon molecular sieves by templated synthesis: the structural varieties, *R. Ryoo, S.H. Joo, S. Jun, T. Tsubakiyama, and O. Terasaki*

07-O-02 One-pot synthesis of phenyl functionalized porous silicates with hexagonal and cubic symmetries, *V. Goletto, M. Impéror and F. Babonneau*

07-O-03 State and redox behavior of iron in MCM-41, *G. Pál-Borbély, Á. Szegedi, K. Lázár and, H.K. Beyer*

07-O-04 A comparative study of Cu interaction with niobium- and aluminum-containing MCM-41 molecular sieves, *M. Ziolek, I. Sobczak, I. Nowak, P. Decyk and J. Stoch*

07-O-05 A novel synthesis strategy leading to the formation of stable transition-metal-oxide mesostructures, *X.S. Zhao, J. Drennan and G.Q. Lu*

07-P-06 Vanadosilicate: Cubic mesoporous molecular sieve, *M. Chatterjee, T. Iwasaki, Y. Onodera, H. Hayashi, T. Ebina and T. Nagase*

07-P-07 Synthesis and characterization of mesoporous Cu-silica spheres via a novel co-assemble route, *P. Zhang, N. Bai, X. Meng and W. Pang*

07-P-08 Synthesis and characterization of mesostructured alumina prepared in the presence of dodecylphosphate, *L. Sicard, B. Lebeau, C. Marichal, J. Patarin and F. Kolenda*

07-P-09 Synthesis and characterizations of mesoporous zirconia-based oxide composites, *J. Zha, D. Wu, Y.-H. Sun, Zh. Zhang, H. Zhang and D. Zhao*

07-P-10 Preparation and catalytic property of FeL/Y composite by a new method, *B. Fan, W. Fan and R. Li*

07-P-11 Mesoporous zirconia: an anionic surfactant inorganic composite, precursor of a tridimensional porous material, *G. Pacheco and J.J. Fripiat*

07-P-12 Synthesis of micelle templated TiO_2 mesophases by a sol-gel approach: effect of the surfactant removal, *D.P. Serrano, G. Calleja, R. Sanz and P. Pizarro*

07-P-13 Preparation and characterization of iron oxide nanoparticles in the channels of MCM-41, *C.M. Yang and K.J. Chao*

07-P-14 The evaluation of iron chromophore concentrations from iron containing MCM-41, *C. Nenu, R. Ganea, R Bîrjega, Gr. Pop and M. Pitu*

07-P-15 Synthesis and characterization of the mesoporous material of single crystal particles, *B. Lee, J. N. Kondo, D. Lu and K.i Domen*

07-P-16 Stabilization of uniformly sized and dispersed copper particles in new Cu-Zn-Al mesoporous catalysts, *S. Valange, J. Barrault, A. Derouault and Z. Gabelica*

07-P-17 A direct synthesis route to the mesoporous silicate SBA-2 bearing thiol groups, *I. Díaz, F. Mohino, J. Pérez-Pariente, E. Sastre, P.A. Wright and W. Zhou*

07-P-18 Template synthesis and characterisation of nanoporous alumina with narrow pore size distribution from inorganic salts, *H.Y. Zhu, P. Cool, G.Q. (Max) Lu and, E.F. Vansant*

07-P-19 Preparation and characterization of copper oxide modified MCM-41 molecular sieves, *C. Minchev, R. K(hn, T. Tsoncheva, M. Dimitrov and M. Fr(ba*

07-P-20 Crystalline, mesoporous NiO-ZrO_2-based solid oxide fuel cell catalysts, *P. Ratnasamy, D. Srinivas, H.S. Soni, A.J. Chandwadkar, H.S. Potdar, C.S. Gopinath, and B.S. Rao*

07-P-21 Application of the AASBU method to the prediction of inorganic structures built exclusively of sodalite cages, *S. Girard, P. Pullumbi, C. Mellot-Draznieks, and G. Férey*

07-P-22 Novel synthesis of nanoporous carbons using colloidal templates, *S.B. Yoon, I.S. Shin and J.-S. Yu*

07-P-23 Synthesis and characterization of mesoporous cerium silicate analogues of MCM-41 type molecular sieves, *S. Laha, P. Mukherjee and R. Kumar*

07-P-24 Synthesis of mesoporous materials using filtrate ofalkali treatment of MFI zeolite, *M. Ogura, E. Kikuchi and M. Matsukata*

08 Syntheses with Non-Ionic surfactants

08-O-01 Mesoporous MSU-X silica tuned for filtration and chromatography applications, *C. Boissière, A. Larbot and E. Prouzet*

08-O-02 Highly ordered mesoporous silicas synthesis using deca(oxyethylene) oleylether as surfactant: variation of the weight percentage of surfactant and incorporation of transition metal cations, *G. Herrier and B.-L. Su*

08-O-03 Silica walls of calcined mesostructured SBA-15 materials templated by triblock copolymers, *M. Impéror and A. Davidson*

08-O-04 Template/AlSBA-15 interaction: double resonance NMR study and consequences on structural properties, *J.-B. d'Espinose de la Caillerie, Y. Yue and A. Gédéon*

08-P-05 The double-step synthesis of MSU-X silica: decoupling the assembly mechanism, *E. Prouzet, C. Boissière, N. Hovnanian and A. Larbot*

08-P-06 Steam-stable aluminosilicate MSU-S mesostructures assembled from zeolite seeds, *Yu Liu, W. Zhang and T.J. Pinnavaia*

08-P-07 A study on the mesoporous silica structures templated by triblock copolymers, *C.-P. Kao, H.-P. Lin, M.-C. Chao, H.-S. Sheu and C.-Y. Mou*

08-P-08 Study of methyl modified MSU-X silicas, *Y. Gong, Z. Li, S. Wan, D. Wu, Y.-H. Sun, F. Deng, Q. Luo and Y. Yue*

08-P-09 Study of mesoporous materials with ultra high surface area prepared from alternate surfactants and silicate sources, *J. F. Pérez-Arévalo, J.M. Domínguez, E. Terrés, A. Hernández and M. Miki*

08-P-10 Synthesis and catalytic properties of SO_3H-mesoporous materials from gels containing non-ionic surfactants, *I. Díaz, F. Mohino, E. Sastre and J. Pérez-Pariente*

08-P-11 Secondary hydrolysis process to synthesize highly ordered mesoporous silica from nonionic surfactant with long hydrophilic chain, *J. Fan, C. Yu and D. Zhao*

08-P-12 Mesostructure design using mixture of nonionic amphiphilicblock copolymers, *J.M. Kim, S.-E. Park and G.D. Stucky*

08-P-13 Stability of mesoporous material SBA-15 and its benefit in catalytic performance, *C. Nie, L. Huang, D. Zhao and Q. Li*

08-P-14 Comparative study of the wall properties in highly-ordered silicate and aluminosilicate mesostructured materials of the MCM-41 and SBA-15 types, *L.A. Solovyov, V.B. Fenelonov, A.Yu. Derevyankin, A.N. Shmakov, E. Haddad, A. Gedeon, S.D. Kirik and V.N. Romannikov*

09 Crystal Structure Determination

09-O-01 Localisation of K^+ ions in (Na_4,K)-LSX and K-LSX zeolites by Rietveld analysis and 39K NMR spectroscopy. A new cationic site in the orthorhombic dehydrated K-LSX at room temperature, *J.L. Paillaud, P. Caullet, L. Delmotte, J.C. Mougenel, S. Kayiran and B. Lledos*

09-O-02 NMR crystallography of $AlPO_4$-CJ_2, *F. Taulelle and C. Huguenard*

09-O-03 FOS-5, a novel zeotype with 3D interconnected 12-ring channels, *T. Conradsson, X. Zou and M.S. Dadachov*

09-O-04 Neutron diffraction study of protons in four lanthanum exchanged X and LSX zeolites, *D.H. Olson, B.H. Toby and B.A. Reisner*

09-O-05 Optimized synthesis and structural properties of lithosilicate RUB-29, So-Hyun Park, *J.B. Parise and H. Gies*

09-P-06 Crystal structure of a cadmium sorption complex of dehydrated fully Cd(II)-exchanged zeolite X, *E.Y. Choi, S.H. Lee, Y.W. Han, Y. Kim and K. Seff*

09-P-07 The structure of a copper molybdate and its relation to other natural and synthetic porous materials based on transition metal polyhedra, *L. A. Palacio, A. Echavarría, A. Simon and, C. Saldarriaga*

09-P-08 A 3-D open-framework nickel aluminophosphate $[NiAlP_2O_8]$ $[C_2N_2H_9]$: assembly of 1-D $AlP_2O_8^{3-}$ chains through $[NiO_5N]$ octahedra, *B. Wei, Jihong Yu, Guangshan Zhu, F. Gao, Y. Li, R. Wang, Bo Gao, Xian Xu, S. Qiu and O. Terasaki*

09-P-09 Structural modifications induced by high pressure in scolecite and heulandite: in-situ synchrotron X-ray powder diffraction study, *G. Vezzalini, S. Quartieri, A. Sani and D. Levy*

09-P-10 Preparation, characterization, and crystal structures of fully indium-exchanged zeolite X, *N.H. Heo, S.W. Jung, S.W. Park, J.S. Noh, W.T. Lim, M. Park, and K. Seff*

09-P-11 Structural investigation by powder X-ray diffraction and solid state nuclear magnetic resonance of $AlPO_4$-SOD, M. Roux, *C. Marichal, J.L. Paillaud, L. Vidal, C. Fernandez, C. Baerlocher and J.M. Chezeau*

09-P-12 Layered germanates with 9-membered rings, *X. Zou, T. Conradsson and M.S. Dadachov*

09-P-13 Dehydration dynamics of mordenite by in-situ time resolved synchrotron powder diffraction study: a comparison with electrostatic site energy calculations, *A. Martucci, M. Sacerdoti and G. Cruciani*

09-P-14 Study of water vapor adsorption in the organically-lined channels of AlMepO-??using X-ray powder diffraction, *K. Maeda, L.B. McCusker and C. Baerlocher*

10 Host-Guest Chemistry

10-O-01 Investigation of indium loaded zeolites and additionally promoted catalysts for selective catalytic reduction of NOx by methane, *F.-W. Schütze, H. Berndt, M. Richter, B. Lücke, C. Schmidt, T. Sowade and W. Grünert*

10-O-02 Ion exchange of alkali metals and control of acidic/basic properties of MCM-22 and MCM-36, *J.-O. Barth, R. Schenkel, J. Kornatowski and J.A. Lercher*

10-O-03 Insertion compounds of metal halides with porosils: "Structured Gases", *P. Behrens, M. Hartl, G. Wirnsberger, A. Popitsch and B. Pillep*

10-O-04 Site selective adsorption and catalytic properties of iron in FER and BEA zeolites, *Z. Sobalík, J.E. Sponer, Z. Tvaru*ková, A., Vondrová, S. Kuriyavar and B. Wichterlová*

10-P-05 Non-acidic zinc zeolite systems: preparation methods, formation processes and catalytic properties in dehydrogenation of methanol, *N.Ya. Usachev, E.P. Belanova, A.V. Kazakov, V.P. Kalinin, A.S. Fomin, I.M. Krukovsky, G.V. Antoshin and O.K. Atal'yan*

10-P-06 Incorporation of Ga ions into Y zeolites by reductive solid-state ion exchange, *R.M. Mihályi, H.K. Beyer and M. Keindl*

10-P-07 Heavy metal exchanged zeolites as precursors for high temperature stable phases, *W. Schmidt and C. Weidenthaler*

10-P-08 Preparation and characterization of H-ZSM-5 exchanged with cobalt by solid state ion exchange, *M. Mhamdi, S. Khaddar-Zine, A. Ghorbel, Y. Ben Taarit and C. Naccache*

10-P-09 Cyclic chemical vapour deposition of TEOS on ZSM-5: effect of deposition temperature on shape selective performance, *H. Manstein, K.P. Möller and C.T. O'Connor*

11 Post-synthesis Modification

11-O-01 Gold-based mono- and bimetallic nanoparticles on HY zeolites, *G. Riahi, D. Guillemot, M. Polisset-Thfoin, D. Bonnin and J. Fraissard*

11-O-02 Unravelled from the back: kinetics of alkoxysilane CVD on zeolites and evidence for pore mouth plugging determined from model conversion over stepwise silanised samples, *H.P. Röger, H. Mantein, W. Böhringer, K.P. Möller and C.T. O'Connor*

11-O-03 Templating role of F- towards D4R units : study of the transformation of the fluorogallophosphate Mu-3 into Mu-2, *A. Matijasic, P. Reinert, L. Josien, A. Simon and J. Patarin*

11-O-04 Modification of the Si/Ti ratio in ETS-10, *G. Koermer, A. Thangaraj and S. Kuznicki*

11-O-05 Binuclear oxo-Fe species in Fe/ZSM-5 catalyst prepared by chemical vapour deposition, *P. Marturano, L. Drozdová, A. Kogelbauer and R. Prins*

11-P-06 Dealumination of zeolite KL, *E.E. Knyazeva, V.V. Yuschenko, F. Fajula and I.I. Ivanova*

11-P-07 Formation of acidic hydroxyl groups during preparation of Pt/KL catalysts as studied by1H MAS NMR, *T. Sato, S.-I. Ito, K. Kunimori and S. Hayashi*

11-P-08 Synthesis and characterization of mesopore Y Zeolite, *B. Ma, W.F. Sun, Z.L. Sun and L.R. Chen*

11-P-09 Controlling the pore size of Hß zeolite by improved chemical vapor deposition of $(CH_3)_3Si\text{-}O\text{-}Si(CH_3)_3$, *Y. Chun, X. Ye, Q.H. Xu and A.-Z. Yan*

11-P-10 Influence of pH of the solution on realumination of BEA zeolite, *Y. Oumi, R. Mizuno, K. Azuma, S. Sumiya, S. Nawata, T. Fukushima, T. Uozumi and T. Sano*

11-P-11 Formation of carbon-intercalated molybdenum sulfides, *J.-S. Chen, Y. Wang, Y. Guo, Y. Zou and W. Xu*

11-P-12 Characterization of partly-detemplated $GaPO_4$-LTA, *S.-F. Yu, C.-Y. Xi, H.-M. Yuan and J.-S. Chen*

11-P-13 Another study on the microwave heating of zeolite - without special loading materials, *J. Dong, L. Xie, X. Jing, H. Xu, F. Wu and J. Hao*

11-P-14 Microwave plasma treatment as an effective technique for activation of zeolite catalysts, *I.I. Lishchiner, O.V. Malova and E.G. Krasheninnikov*

11-P-15 Synthesis and characterization of microporous titanium-silicate materials, *S. Mintova, B. Stein, J.M. Reder and T. Bein*

11-P-16 From borosilicate to gallo- and aluminosilicate zeolites: new methods for lattice substitution via post-synthetic treatment, *C.Y. Chen and S.I. Zones*

11-P-17 Studies on the structure of zeolite Y modified by radio-frequency fluorocarbon plasma treatment, *S. Yamazaki, T. Nishimura, K. Furukawa, H. Ijiri and K. Tsutsumi*

11-P-18 Dual-temperature reagent-less ion-exchange separations on zeolites, *V.D. Timofeevskaja, O.T. Gavlina, V.A. Ivanov and V.I. Gorshkov*

11-P-19 Rare earth exchange in small pore zeolites and its effect on their hydrothermal stability, *G. Cao, M.J. Shah and W.A. Wachter*

11-P-20 Modification of mordenite and natural clinoptilolite by copper: role of drying temperature, *I. Rodríguez-Iznaga, V. Petranovskii, G. Rodríguez-Fuentes, N. Bogdanchikova and M. Avalos*

11-P-21 Study on the acidity of modified HY zeolites prepared by combination of chemical dealumination and hydrothermal treatment, *M. Han, L.-P. Zhou, X.-W. Li and L.-Q. She*

11-P-22 Modification of Beta-zeolite by dealumination and realumination, *J.Y. Zhang, L-P. Zhou and X.W. Li*

11-P-23 Ultrastable zeolites Y (USY) modified with phosphorus and boron, *A.V. Abramova, Ye.V. Slivinsky, L.Ye. Kitaev, A.A. Kubasov, H. Lechert, W.D. Basler, V.V. Yushchenk and Z.M. Matieva*

11-P-24 Structural properties and sieving effects of surface modified ZSM-5, *S. Zheng, H. Heydenrych, H.P. Röger, A. Jentys and J.A. Lercher*

11-P-25 The use of binary adsorption studies to investigate the effect of hydrothermal treatment on zeolites Rho and Mordenite, *L.H. Callanan, C.T. O'Connor and E. van Steen*

11-P-26 Acid sites in thermal transformations of Ca-rich clinoptilolite, *G.P. Valueva, I.S. Afanassiev, E.A. Paukshtis, Y.V. Seryotkin, N.K. Moroz and A.A. Budneva*

11-P-27 The effect of calcination on the isomorphously substituted microporous materials using ozone, *D. Mehn, A. Kukovecz, I. Kiricsi, F. Testa, E. Nigro, R. Aiello, G. Daelen, P. Lentz, A. Fonseca and J. B.Nagy*

11-P-28 Alumination of siliceous zeolites, *A. Omegna, M. Haouas, G. Pirngruber and R. Prins*

11-P-29 New hydrophobic Ti-Beta catalyst obtained by silylation and its catalytic performance for olefin epoxidation, *A. Corma, M.E. Dómine, J.A. Gaona, M.T. Navarro, F. Rey and S. Valencia.*

11-P-30 MFI zeolite with uniform mesopores created by alkali treatment, *M. Ogura, E. Kikuchi and M. Matsukata*

12 In-situ Spectroscopy and Catalysis

12-O-01 2D correlation IR spectroscopy of xylene isomerisation on H-MFI zeolite, *F. Thibault-Starzyk, A. Vimont and J.-P. Gilson*

12-O-02 Structure/reactivity correlation in Fe/ZSM5 for deNOx applications. In-situ XAFS characterization and catalysis, *A.A. Battiston, J.H. Bitter and D.C. Koningsberger*

12-O-03 Interaction of diazines with faujasites studied by IR spectroscopy, temperature-programmed desorption, and molecular modeling methods, *J. Döbler, E. Geidel, B. Hunger, K.H.L. Nulens and R.A. Schoonheydt*

12-O-04 DRIFT study of dinitrogen and dihydrogen adsorption on Li- and Na- forms of LSX zeolite, *V.B. Kazansky, A.I. Serykh, E. Tichomirova, V. Yu Borovkov and M. Bulow*

12-P-05 Study of relationship between mordenite acidity and structure with calcination temperature, *Z. Zhu, Q. Chen and We. Chen*

12-P-06 Infrared observation of the stable carbenium ions formed by adsorption of olefins on zeolite Y at low temperatures, *S. Yang, J.N. Kondo and K. Domen*

12-P-07 Characterization of aluminosilicate zeolites by UV-Raman spectroscopy, *Y. Yu, G. Xiong, C. Li and F.-S. Xiao*

12-P-08 Adsorption of furan, 2,5-dihydrofuran and tetrahydrofuran on sodium-ion exchanged faujasites with different Si/Al ratios, *I.A. Beta, H. Böhlig, J. Döbler, H. Jobic, E. Geidel and B. Hunger*

12-P-09 DRIFT and FTIR spectra of N_2 and C_2H_4 adsorbed on CuNaY, *G. Hübner and E. Roduner*

12-P-10 Raman study of the building units in the zeolite structure, *P.P.H.J.M. Knops-Gerrits, X.-Y. Li, N.-T. Yu and P.A. Jacobs*

12-P-11 Positron annihilation study in MCM-41, *H.Y. Zhang, Y.J. He, Y.B. Chen, H.Y. Wang and T. Horiuchi*

12-P-12 NMR studies on the pyrrole adsorption over Na^+, Li^+ exchanged zeolites of type FAU, *M. Sánchez-Sánchez and T. Blasco*

12-P-13 Variable-temperature FTIR study of the equilibrium between C-bonded and O-bonded carbon monoxide in H-ZSM-5, *G. Turnes Palomino, M. Peñarroya Mentruit, A.A. Tsyganenko, E. Escalona, Platero and C. Otero Areán*

12-P-14 Role of the various acid sites in MOR on o-xylene conversion: An in-situ I.R. approach, *O. Marie, F. Thibault-Starzyk, P. Massiani and J.C. Lavalley*

12-P-15 FTIR- Studies on adsorption and decomposition of NO on in-situ synthesized ZSM-5/cordierite catalysts, *N. Guan, X. Shan, X. Zeng, S. Xiang, A. Trunschke and M. Baerns*

12-P-16 Acid sites in dealuminated mordenite, *V.L. Zholobenko and G.P. Mitchell*

12-P-17 Infrared study of iron-exchanged Y zeolite and its HDS activity, *M. Nagai, O. Uchino and S. Omi*

13 Frameworks and Acid Sites

13-O-01 Where are the acid sites in zeolites? A novel NMR approach to measure B/Al ordering around structure directing agents, *H. Koller, M. Kalwei, C. Fild, R.F. Lobo, M.A. Camblor, L.A. Villaescusa and L. van Wüllen*

13-O-02 The effect of the nature of heteroatoms (Al, Fe, B) on their distribution in the ZSM-5 structure, *J. Dedecek, M. Tudor and J. Cejka*

13-O-03 Toward the quantification of aluminum in zeolites using high-resolution solid-state NMR, *C. Fernandez, A.-A. Quoineaud, V. Montouillout, S. Gautier and S. Lacombe*

13-O-04 Acidity of ITQ-2 zeolite as studied by FT-IR spectroscopy of adsorbed molecules in comparison with that of MCM-22, *B. Onida, F. Geobaldo, L. Borello and E. Garrone*

13-P-05 In-situ FTIR studies of the acidity of $H_3PW_{12}O_{40}$ and its porous salts. Interaction with H_2O, NH_3 and pyridine, *N. Essayem, A. Holmqvist, G. Sapaly, J.C. Védrine and Y. Ben Tâarit*

13-P-06 Dynamics of p-nitroaniline in the micropore of zeolite ZSM-5 studied by solid-state NMR, *S. Hayashi and Y. Komori*

13-P-07 The use of microcalorimetry to study the effects of post-synthesis treatments on the modification of the acidity of several HY-type zeolites, *A. Auroux and M.L. Occelli*

13-P-08 ESR investigations of the catalytic properties of Lewis acid sites in H-mordenite, *T.M. Leu and E. Roduner*

13-P-09 External surface acidity of modified zeolites: ESR via adsorption of stable nitroxyl radicals and IR spectroscopy, *A.B. Ayupov, G.V. Echevsky, E.A. Paukshtis, D.J. O'Rear and C.L. Kibby*

13-P-10 Very strong acid site in HZSM-5 formed during the template removal step; its control, structure and catalytic activity, *A. Kohara, N. Katada and M. Niwa*

13-P-11 Correlation between 11B NMR isotropic chemical shifts and structural parameters in borates and boro-silicates, *J. Plévert, F. Di Renzo and F. Fajula*

13-P-12 Investigation of the paramagnetic effect of oxygen in the ^{23}Na MAS NMR and ^{23}Na MQMAS NMR spectra of LiNaX, *R.J. Accardi, M. Kalwei and R.F. Lobo*

13-P-13 Characterization of acidic sites in HY and LaY zeolites by laser-induced fluorescence of adsorbed quinoline, *A. Lassoued, J. Thoret, P. Batamack, A. Gédéon and J. Fraissard*

13-P-14 Dynamic behaviour of acetonitrile molecules adsorbed in $ALPO_4$-5 and SAPO-5 studied by solid NMR method, *S. Ishimaru, M. Ichikawa, K. Gotoh and R. Ikeda*

13-P-15 Determination of the Si/Al ratio of faujasite-type zeolites, *C.H. Rüscher, J.-C. Buhl and W. Lutz*

13-P-16 Theoretical investigation of the chemical shift anisotropy of toluene adsorbed on zeolite X, *A. Simperler, A. Philippou, D.-P. Luigi, R.G. Bell and M. W. Anderson*

13-P-17 Acid properties of dexydroxylated ferrierites studied by IR spectroscopy, *J. Datka, B. Gil and K. Góra-Marek*

13-P-18 Aluminium species in activated zeolites: solid-state NMR spectroscopy of the active sites, *B.H. Wouters, T.-H. Chen and P.J. Grobet*

13-P-19 Aluminium distribution in high silica pentasil ring zeolites, *B. Wichterlová, J. Dedecek, Z. Sobalík and J. Cejka*

13-P-20 Effects of hydration on $AlPO_4$-14 and $AlPO_4$-18 structures: ^{31}P MAS and ^{27}Al 3Q-MAS NMR study, *C.V. Satyanarayana, R. Gupta, K. Damodaran, S. Sivasanker and S. Ganapathy*

13-P-21 Comparative study of the acidity of the structurally related faujasite type zeolites: FAU, EMT and ZSM-20, *H. Kosslick, R. Fricke, H. Miessner, D.L. Hoang and W. Storeck*
13-P-22 The effect of flexible lattice aluminum in zeolites during the nitration of aromatics, *M. Haouas, A. Kogelbauer and R. Prins*
13-P-23 Characterization of acidic sites in zeolites by heteronuclear double resonance solid state NMR, *S.B. Waghmode, A. Abraham, S. Sivasanker, J.P. Amoureux and S. Ganapathy*
13-P-24 Measurement of MQMAS heteronuclear correlation spectra in microporous aluminophosphates, *C. Fernandez and M. Pruski*
13-P-25 FTIR studies of the interaction of aromatic and branched aliphatic compounds with internal, external and extraframework sites of MFI-type zeolite materials, *T. Armaroli, A. Gutiérrez Alejandre, M. Bevilacqua, M. Trombetta, F. Milella, J. Ramírez and G. Busca*

14 Frameworks, Cations, Clusters

14-O-01 Imaging the mesopores in zeolite Y using three-dimensional transmission electron microscopy, *A.H. Janssen, A.J. Koster and K.P. de Jong*
14-O-02 ^{17}O NMR studies of the structure and basic properties of zeolites, *D. Freude, T. Loeser and U. Pingel*
14-O-03 Theoretical interpretation of UV-VIS spectra of Cu- and Ag-species in zeolites: structure vs. transition energies, *P. Nachtigall, M. Davidová, M. Silhan and D. Nachtigallová*
14-O-04 Silver ions and quantum-sized silver sulfide clusters in zeolite A, *D. Brühwiler, C. Leiggener and G. Calzaferri*
14-O-05 Elucidating the nature and reactivity of cobalt ions in CoAPOs. A combined FTIR and EPR study of NO and NO_2 adsorbed at 77K and 298K, *E. Gianotti, M.C. Paganini, G. Martra, E. Giamello, S. Coluccia and L. Marchese*
14-P-06 Relaxation processes of Na ion in dehydrated Na_{12}-A zeolite, *T. Ohgushi and K. Ishimaru*
14-P-07 Adsorption of DTBN at monovalent cations in zeolite y as studied by electron spin resonance spectroscopy, *M. Gutjahr, W. Böhlmann, R. Böttcher and A. Pöppl*
14-P-08 Nature of the active sites of Mo-containing zeolites. XANES studies at Mo K and LIII -edges, *F.G. Requejo, E.J. Lede, L.B. Pierella and O.A. Anunziata*
14-P-09 EPR studies on nitrogen monoxide in zeolites, *H. Yahiro, N.P. Benetis, A. Lund and M. Shiotani*
14-P-10 Evidence of partially broken bridging hydroxyls in molecular sieves from1H MAS spin echo NMR spectroscopy, *T.-H. Chen, B.H. Wouters and P.J. Grobet*
14-P-11 Structure change of molecular sieve SAPO-37 at high temperature studied by ^{27}Al MQ MAS NMR, *T.-H. Chen, B. Wouters and P. Grobet*
14-P-12 Effects of molecular confinement on structure and catalytic behaviour of metal phthalocyanine complexes encapsulated in zeolite-Y, *S. Seelan, D. Srinivas, M.S. Agashe, N.E. Jacob and S. Sivasanker*
14-P-13 Investigations on isomorphous substitution and catalytically active centres in MeAPO-31 (Me = Mn, Co, Zn, Ti), *N. Novak Tusar, A. Ristic, A. Ghanbari-Siahkali, J. Dwyer, G. Mali, I. Arcon and V. Kaucic*
14-P-14 A comparative study of Ti^{4+} sites in titanium silicalite (TS-1) synthetized by different methods, *N.G. Gallegos, A.M. Alvarez, J.F. Bengoa, M.V. Cagnoli, S.G. Marchetti and A.A. Yeramian*
14-P-15 Behaviour of Fe(III) ions in Y zeolites in the presence of Cu(II) and Ag(I) ions: an ESR study, *A.L. Kustov, E.E. Knyazeva, E.A. Zhilinskaya, A. Aboukais and B.V. Romanovsky*
14-P-16 FT-Raman spectroscopic studies of host-guest interactions in zeolites, *Y. Huang, J.H. Leech and R.R. Poissant*
14-P-17 High-temperature MAS NMR investigation of the mobility of cations and guest compounds in zeolites X and Y, *M. Hunger, A. Buchholz and U. Schenk*
14-P-18 Generation of long-lived electron-hole pairs through sorption of biphenyl into acidic ZSM-5 zeolites, *I. Gener, A. Moissette, H. Vezin, J. Patarin, and C. Brémard*
14-P-19 Defects study in microporous materials by HRSEM, HRTEM and diffraction techniques, *G. Gonzalez, Z. Lopez and R. Reichelt*
14-P-20 The effect of the framework structure on the chemical properties of the vanadium oxide species incorporated within zeolites and their photocatalytic reactivity, *S. Higashimoto, M. Matsuoka, M. Che and M. Anpo*
14-P-21 Characterization of aluminium and iron sites in MCM-22, *J. Cejka, J. Dedecek, J. Kotrla, M. Tudor, N. *ilková and S. Ernst*
14-P-22 Valency and coordination states of iron in FeAPO-11. An in-situ Mössbauer study, *K. Lázár, N. *ilková and J. Cejka*
14-P-23 Comparative properties of modified HEMT and HY zeolites from the FTIR study of CO adsorption: effect of the dealumination and amorphous debris on the Brönsted acidity, *O. Cairon and T. Chevreau*
14-P-24 Raman spectroscopic study of 2,2'-bipyridine sorbed into ZSM5, *A. Moissette, C. Brémard, I. Gener and N. Louchart*
14-P-25 Fractals of silica aggregates, *Z. Li, D. Wu, Y.-H. Sun, J. Wang, Yi Liu and B. Dong*
14-P-26 Structure of Mo species incorporated into SBA-1 and SBA-3 studied by XAFS and UV-VIS spectroscopies, *H. Yoshitake, S.H. Lim, S. Che and T. Tatsumi*
14-P-27 Quantification of electric-field gradients in the supercage of Y zeolites by comparing the chemical shifts of ^{131}Xe (I = 3/2) and ^{129}Xe (I = 1/2), *Y. Millot, P.P. Man, M.-A. Springuel-Huet and J. Fraissard*
14-P-28 Iron species present in Fe/ZSM-5 catalysts prepared by ion exchange in aqueous medium or in the solid state, *M.S. Batista, M.A.M. Torres, E. Baggio-Saitovich and E.A. Urquieta-González*
14-P-29 Laser ablation mass spectrometry : a technique for observing zeolite occluded molecules, *S. Jeong, K.J. Fisher, G.D. Willett and R.F. Howe*
14-P-30 Characterisation of TS-1 active sites by adsorption of organic probes, *C. Flego, A. Carati and M.G. Clerici*
14-P-31 NIR FT-Raman spectroscopy on molecular sieves, *E. Löffler and M. Bergmann*
14-P-32 Characterization of Zn and Fe substituted mordenite by XAFS, *M. Dong, J.-G. Wang and Y.-H. Sun*
14-P-33 Identification of vanadium species in VAPO and VAPSO aluminophosphate by UV resonance raman spectroscopy, *Jia. Yu, Z. Liu, Q. Xin and C. Li*
14-P-34 On the interaction of H_2O with TS-1: a spectroscopic and ab-initio study, *A. Damin, G. Ricchiardi, S. Bordiga, F. Bonino, A. Zecchina, F. Ricci, G. Spanò, F. Villain and C. Lamberti*
14-P-35 Spectroscopic study of the nature of vanadyl groups: influence of the support (SiO_2 and Alß and Siß zeolites), *S. Dzwigaj, M. Matsuoka, M. Anpo and M. Che*
14-P-36 Characterization of Ni, Pt zeolite catalysts by TEM and EDX, *M.H. Jordão and D. Cardoso*
14-P-37 NMR and ESR investigations of alkali metal particles in NaY zeolite, *F. Rachdi and L.C. de Ménorval*
14-P-38 Topochemical changes in large MFI-type crystals upon thermal treatment in oxidizing and non-oxidizing atmosphere, *O. Pachtová, B. Bernaue, J.-A. Dalmon, S. Miachon, I. Jirka, A. Zikánová and M. Kocirík*
14-P-39 Structure of Fe(III) sites in iron substituted aluminophos-phates: a computational and X-ray spectroscopic investigation, *C. Zenonos, A. Beale, G. Sankar, D.W. Lewis, J.M. Thomas and C.R.A. Catlow*
14-P-40 Possible formation of $Cu^{+2}(CO)_2(H_2O)n$ complexes in a ZSM-5 zeolite prepared by direct synthesis: evidence for the occurrence of Cu^+-Cu^+ pairs?, *F. Geobaldo, B. Onida, M. Rocchia, S. Valange, Z. Gabelica and E. Garrone*

15 Modelling and Theoretical Studies A

15-O-01 Proton jumps in dehydrated acidic zeolite catalysts. Rate predictions based on ab-initio calculations, *M. Sierka and J. Sauer*
15-O-02 Ab-initio simulation of dynamical processes in zeolites, *L. Benco, T. Demuth, J.Hafner, F. Hutschka and H. Toulhoat*
15-O-03 A theoretical study of the methylation of toluene by methanol over acid mordenite, *A. Vos, X. Rozanska, R. Schoonheydt, R. van Santen, F. Hutschka and J. Hafner*
15-O-04 Coverage effects on adsorption of water in faujasite: An ab-initio cluster and embedded cluster study, *J. Limtrakul, S. Nokbin, P.Chuichay, P. Khongpracha, S. Jungsuttiwong and T.N. Truong*
15-O-05 The Beckmann rearrangement catalyzed by silicalite: a spectroscopic and computational study, *G.A. Fois, G. Ricchiardi, S. Bordiga, C. Busco, L. Dalloro, G. Spanò and A. Zecchina*
15-P-06 A reactivity index study to choose the best template for zeolite synthesis, *A. Chatterjee and T. Iwasaki*
15-P-07 Effects of ion-exchanged alkali metal cations on the photolysis of alkyl ketones included within ZSM-5 zeolite cavities: A study of ab-initio molecular orbital calculations, *H. Yamashita, S. Takada, M. Nishimura, H. Bessho and M. Anpo*
15-P-08 Encapsulated guest atoms within the basic beta cage of sodalitic zeolite. A theoretical ab-initio study, *N.U. Zhanpeisov and M. Anpo*
15-P-09 n-Hexane aromatization over Pt-alkaline zeolites: ab-initio calculations on the influence of the exchanged cations and zeolite type (L, and Y) on electronic properties of Pt, *S.B. Waghmode, P. Bharathi, R. Vetrivel and S. Sivasanker*
15-P-10 A theoretical study of adsorption of carbon monoxide on Ag-ZSM-5 zeolite, *S. Jungsuttiwong, P. Khongpracha, T.N. Truong and J. Limtrakul*
15-P-11 A theoretical/spectroscopic study of the coordination of transition metal ions in zeolites, *A. Delabie, M.H. Groothaert, R.A. Schoonheydt and K. Pierloot*
15-P-12 Ab-initio study of the adsorption and reactions of hydrocarbons in mordenite, *T. Demuth, L. Benco, J. Hafner, H. Toulhoat and F. Hutschka*
15-P-13 Properties of Cu^{2+} and Cu^+ cations in MFI framework: DFT and IR studies, E. Broclawik, *J. Datka, B. Gil and P. Kozyra*
15-P-14 Nonempirical (ab-initio) and semiempirical calculations of the elementary fragments of zeolites. Permeability of rings zeolite fragments, *A.V. Gabdrakipov, L.D. Volkova, N.A. Zakarina and V.Z. Gabdrakipov*
15-P-15 Modelling light alkane transformation over HZSM-5 zeolite, *X. Wang, F. Lemos and F.R. Ribeiro*
15-P-16 Activity-acidity relationship in Y zeolite: an experimental and quantum-chemical study, *X. Wang, M.A.N.D.A. Lemos, F. Lemos, C. Costa and F.R. Ribeiro*
15-P-17 DFT study of structure changes in hydrated $AlPO_4$-n: The case of $AlPO_4$-34, *G. Poulet, A. Tuel and P. Sautet*
15-P-18 A mechanistic exploration of alkene epoxidation mediated by H_2O_2 within porous titanosilicate catalysts, *C.M. Barker, N. Kaltsoyannis and C.R.A. Catlow*
15-P-19 Transition-state shape-selectivity insights from a Fukui function overlap method, *L.A. Clark and R.Q. Snurr*
15-P-20 A DFT study of the isomerization reactions of aromatics catalyzed by acidic zeolites, *X. Rozanska, R.A. van Santen, F. Hutschka, and J. Hafner*
15-P-21 Ab-initio investigation of non-framework aluminum species in zeolites, *D. Lopes Bhering, C.J.A. Mota and A. Ramirez-Solís*
15-P-22 A DFT study of the cracking reaction of thiophene activated by zeotype

catalysts: role of the basic Lewis site, *X. Rozanska, R.A. van Santen and F. Hutschka*

15-P-23 Modelling transition metal cations in zeolites: how do they interact with the framework?, *D. Berthomieu, A. Goursot, J-M. Ducéré, G. Delahay, B. Coq and A. Martinez*

15-P-24 Theoretical prediction of IR spectra of guest molecules in zeolites: the stretching frequency of CO adsorbed at various cationic sites in ZSM-5, *T.A. Wesolowski, A. Goursot and J. Weber*

15-P-25 Development of a tight-binding treatment for zeolites, *M. Elstner, A. Goursot, Z. Hajnal, T. Heine and J. Weber*

15-P-26 1-D growth of selenium wires in silicalite-1 zeolite, *C. Bichara and R.J.-M. Pellenq*

15-P-27 Cumulative coordinates for approximations of atomic multipole moments in cationic forms of aluminosilicates, *A.V. Larin and D.P. Vercauteren*

15-P-28 Computer simulations of water in zeolites, *C. Bussaia, R. Haberlandt, S. Hannongbuai and S. Jost*

16 Modelling and Theoretical Studies B

16-O-01 Simulating shape selectivity in alkane hydroconversion by zeolites, *M. Schenk, T.L.M. Maesen and B. Smit*

16-O-02 Molecular dynamics of the faujasite (111) surface, *B. Slater and C.R.A. Catlow*

16-O-03 Adsorption of xylene isomers and water in faujasites. A molecular simulation study, *S. Buttefey, A. Boutin and A.H. Fuchs*

16-O-04 Reaction dynamics in acidic zeolites: room temperature tunneling effects, *J.T. Fermann and S.M. Auerbach*

16-O-05 Molecular modeling of multicomponent diffusion in zeolites and zeolite membranes, *M.J. Sanborn, A. Gupta, L.A. Clark and R.Q. Snurr*

16-P-06 Location of triethylmethylammonium ions in MFI by combining molecular modeling and X-ray diffraction, *R. Millini*

16-P-07 A hypothetical zeolite structure MCR16: topological design and template choice, *B. Li, P. Sun, Q. Jin and D. Ding*

16-P-08 Computational analysis of the shape-selective isopropylation of biphenyl over large pore zeolites, *J. Joffre, D. Mravec and P. Moreau*

16-P-09 De novo simulation and spectroscopic study of iron speciation in ZSM-5 and CIT-5, *P.-P.H.J.M. Knops-Gerrits and W.A. Goddard III*

16-P-10 Exact statistical mechanical treatment of a lattice model of hydrocarbon adsorption on zeolites, *G. Manos, L.J. Dunne, Z. Du and M.F. Chaplin*

16-P-11 Computational studies of the structure of Na- and H-Mordenite, *A.E. Gray, A. O'Brien and D.W. Lewis*

16-P-12 Monte Carlo simulation of isobutane in silicalite, *D. Paschek and R. Krishna*

16-P-13 Characterisation of hypothetical zeolite frameworks, *M.D. Foster, R.G. Bell and J. Klinowski*

16-P-14 Cation mobility and the sorption of chloroform in zeolite NaY: a molecular dynamics study, *N.A. Ramsahye and R.G. Bell*

16-P-15 Computational studies of the calcination of fluorinated gallophosphates: exploration of their template-free calcined forms, *S. Girard, J.D. Gale, C. Mellot-Draznieks and G. Férey*

16-P-16 Molecular simulation studies on the effectiveness of template type on TS-1 crystal morphology, *H. Zhou, H. He and Z. Jing*

16-P-17 A molecular dynamic approach on the selective confor-mational change of ethylene glycol in sodalite cage, *M. Sato*

16-P-18 The mutual influence of dynamic processes acting in different time scales, *S. Fritzsche, R. Haberlandt, A. Schüring and M. Wolfsberg*

16-P-19 Lattice-dynamical calculations for zeolites of natrolite group, *S.V. Goryainov and M.B. Smirnov*

16-P-20 Kinetic modelling of the dynamic interaction between NO and N_2O over Cu-ZSM5, *R. Pirone, P. Ciambelli, A. Di Benedetto, B. Palella and G. Russo*

17 Principles of Adsorption

17-O-01 Liquid-solid and solid-solid phase transitions of oxygen in a single cylindrical pore, *K. Morishige and Y.Ogisu*

17-O-02 Structural study of benzene, tetrachloroethene and trichloroethene sorbed phases in silicalite-1, *N. Floquet, J.P. Coulomb, G. Weber, O. Bertrand and J.P. Bellat*

17-O-03 Molecular ordering of the adsorbed phase within the microporous model aluminophosphate $AlPO_4$-11 at cryogenic temperatures, *N. Dufau, N. Floquet, J-P. Coulomb, P.L. Llewellyn and J. Rouquerol*

17-O-04 Adsorption properties of a supercritical fluid on mesoporous molecular sieves under high pressure, *Ya. Goto, N. Setoyama, Y. Fukushima, T. Okubo, Yu. Goto, Y. Imada, Y. Kubota and Y. Sugi*

17-P-05 Confinement in model host materials: experimental study of quasi-(1d) systems, *J.P. Coulomb, N. Floquet, C. Martin, Y. Grillet and J. Patarin*

17-P-06 Studies on desorption behavior of organics on siliceous ferrierite, *B. Qian, Y. Zeng and Y.-C. Long*

17-P-07 Investigation of hydrocarbon adsorption on large and extra-large pore zeolites, *C.Y. Chen and S.I. Zones*

17-P-08 Different chemisorption methods applied to zeolite supported Pt-catalysts, *J.C. Groen, J. Pérez-Ramírez and L.A.A. Peffer*

17-P-09 Structure vs. adsorption properties of 5A zeolites, *H. Paoli, T. Bataille, B. Rebours, A. Méthivier and H. Jobic*

17-P-10 Hydrogen adsorption in lithium exchanged Na A zeolites, *S. Kayiran, F. Darkrim and A. Gicquel*

17-P-11 Macroscopic and microscopic investigations of the interaction of a chloroalkene on a MFI zeolite, *V. François, S. Maure, F. Bouvier, G. Weber, O. Bertrand, J.P. Bellat and C. Paulin*

17-P-12 Sorption and pore condensation behavior of pure fluids in mesoporous MCM-48 silica, MCM-41 silica and controlled pore glass, *M. Thommes, R. Köhn and M. Fröba*

17-P-13 Pore size analysis with H_2O adsorption measurement of organically modified MCM-41 type materials, *N. Igarashi, K. Nakai, K. Hashimoto and T. Tatsumi*

17-P-14 Adsorption of carbon dioxide by X zeolites exchanged with Zn^{2+} and Cu^{2+}: isosteric heat and adsorption isotherms, *A. Khelifa, Z. Derriche and A. Bengueddach*

17-P-15 A combination of high resolution manometry, gravimetry and microcalorimetry to study the co-adsorption of Ar/N_2 mixtures on 5A and ^{13}X zeolites, *S. Moret, F. Rouquerol, J. Rouquerol and P.L. Llewellyn*

17-P-16 Gas adsorption microcalorimetry on zeolites under supercritical conditions up to 15 bars, *T. Poyet, F. Rouquerol, J. Rouquerol and P.L. Llewellyn*

18 Adsorption and Separation Process

18-O-01 An experimental adsorbent screening study for CO_2 removal from flue gas, *P.J.E. Harlick, H. Halsall-Whitney and F. Handan Tezel*

18-O-02 Amino acids in BEA type channels, *C. Buttersack and A. Perlberg*

18-O-03 Kinetic separation of binary mixtures of carbon dioxide and C_2 hydrocarbons on modified LTA-type zeolites, *C.J. Guo, D. Shen and M. Bülow*

18-O-04 A novel adsorbent for the separation of propane/propene mixtures, *W. Zhu, F. Kapteijn and J. A. Moulijn*

18-O-05 Iodide removal using zeolite-based reactive adsorption, *S. Kulprathipanja and B. Spehlmann*

18-P-06 Evaluation of mesoporous silicas as stationary phases for high performance liquid chromatography (HPLC), *L. Sierra, B. Lopez, A. Ramirez and J.-L. Guth*

18-P-07 Adsorption of N-nitrosamines by zeolites in solutions, *Ying Wang, J.H. Zhu, D. Yan, W.Y. Huang and L.L. Ma*

18-P-08 An adsorption-desorption process for separation of C_8 aromatics, *G.-Q. Guo and Y.-C. Long*

18-P-09 Preparation of Na-A zeolite capillary columns by in-situ synthesis, *D. Kou, Z. Li, J. Wu, Ming Liu and S. Xiang*

18-P-10 Adsorption of CO_2, SO_2 and NH_3 on zeolitic materials synthesized from fly ash, *S. Hernández, R. Juan, X. Querol, N. Moreno, P. Ferrer and J.M. Andrés*

18-P-11 Dibenzothiophene adsorption over zeolites with faujasite structure, *J.L. Sotelo, M.A. Uguina, M.D. Romero, J.M. Gómez, V.I. Águeda and M.A. Ortiz*

18-P-12 Pressure swing adsorption of ethyl acetate on silica MCM-41, *S. Namba, D. Yomoda, J. Aoyagi, K. Minagawa, T. Kugita and J. Izumi*

18-P-13 P occlusion in LTA: an approach for enhancing N_2 adsorption properties, *L. Johnson and M. Miller*

18-P-14 Sulfur guard bed material from local bentonite deposits, *S. Mikhail and T. Zaki*

18-P-15 Simulation for removal of binary solvent vapor by adsorption onto high silica zeolite, *K. Chihara, T. Saito, H. Suzuki, H. Yamaguchi and Y. Takeuchi*

19 Diffusion: Fundamental Approach

19-K-01 Use of1H NMR imaging to study the diffusion and co-diffusion of gaseous hydrocarbons in HZSM-5 catalysts, *P. N'Gokoli-Kekele, M.-A. Springuel-Huet, J.-L. Bonardet, J.-M. Dereppe and J. Fraissard*

19-O-02 Studies of adsorption, diffusion and molecular simulation of cyclic hydrocarbons in MFI zeolites, *L. Song, Z.L. Sun and L.V.C. Rees*

19-O-03 The effect of silanisation on the intracrystalline diffusivity of ZSM-5, *W.L. Duncan and K. P. Möller*

19-O-04 Interference microscopy as a tool of choice for investigating the role of crystal morphology in diffusion studies, *O. Geier, S. Vasenkov, E. Lehmann, J. Kärger, R.A. Rakoczy and J. Weitkamp*

19-O-05 Estimation of the interphase thickness and permeability in polymer-zeolite mixed matrix membranes, *A. Erdem-Senatalar, M. Tatlier and S.B. Tantekin-Ersolmaz*

19-P-06 Modeling of sulfur dioxide breakthrough curves from ternary wet mixtures on MOR type zeolite, *M. Mello, M. Eic, S. Hocevar and U. Lavrencic-Stangar*

19-P-07 The diffusion and sorption dynamics of acetylene in zeolites, *Gy. Onyestyák, J. Valyonand and L.V.C. Rees*

19-P-08 Transient uptake measurements using an oscillating microbalance: effect of acid leaching on the diffusivity of n-hexane in Pt/H-Mordenite, *S. van Donk, A. Broersma, O.L.J. Gijzeman, J.H. Bitter and K.P. de Jong*

19-P-09 Use of ^{129}Xe NMR spectroscopy to study gaseous hydrocarbon diffusion in a fixed bed of HZSM-5 zeolite, *M.-A. Springuel-Huet, P. N'Gokoli-Kekele, C. Mignot, J.-L. Bonardet and J. Fraissard*

19-P-10 Adsorption and diffusion of alkanes and their mixtures in silicalite studied with positron emission profiling technique, *A.O. Koriabkina, D. Schuring, A.M. de Jong and R.A. van Santen*

20 Zeolite Membranes and Films

20-O-01 Polyamines as strong covalent linkers for the assembly of mono and double layers of zeolite crystals on glass, *K. Ha, Y.S. Chun, A. Kulak, Y.S. Park, Y.-J. Lee and K.B. Yoon*

20-O-02 The use of seeds in the synthesis of mono-and bi-layered zeolite membranes, *L. Gora, G. Clet, J.C. Jansen and Th. Maschmeyer*

20-O-03 Growth of oriented mordenite membranes on porous (-Al_2O_3 supports, *G. Li, X. Lin, E. Kikuchi and M. Matsukata*

20-O-04 Depth-sensitive structural study of silicalite-1 films with grazing

incidence X-ray diffraction, *S. Mintova, T.H. Metzger and T. Bein*
20-O-05 Regeneration of supercritical carbon dioxide by alumina supported MFI zeolite and mesoporous silica membranes, *K.J. Chao, C.H. Kao, Y.W. Chiu, X.R. Lin and C.S. Tan*
20-P-06 Dehydrogenation of ethylbenzene to styrene using ZSM-5 type zeolite membranes as reactors, *X.-F. Zhang, Y.-S. Li, J.-Q. Wang, H.-O. Liu and C.-H. Liu*
20-P-07 Preparation of high-permeance ZSM-5 tubular membranes by varying-temperature synthesis, *Y.-S. Li, Xio. Zhang, J.-G. Wang and S. Guo*
20-P-08 Synthesis of FAU type films on steel supports using a seeding method, *Z. Wang, J. Hedlund and J. Sterte*
20-P-09 Structured zeolite ZSM-5 coatings on ceramic packing materials, *O. Öhrman, U. Nordgren, J. Hedlund, D. Creaser and J. Sterte*
20-P-10 Effects of synthesis parameters on intra-pore zeolite formation in zeolite A membranes, *M. Lassinantti, J. Hedlund and J. Sterte*
20-P-11 Pure-silica zeolite low-k dielectric thin films by spin-on process, *Zhen. Wang, H. Wang, A. Mitra, L. Huang and Yu. Yan*
20-P-12 Preparation of silicalite-1 and beta zeolite/ceramic composite membranes and removal of trace phenol and benzene from water through them, *Xiansen Li and S. Xiang*
20-P-13 Factors affecting film thickness in the preparation of supported ZSM-5 zeolite, *E.I. Basaldella, A. Kikot, J.F. Bengoa and J.C. Tara*
20-P-14 Growing zeolite films onto gold surfaces, *E I. Basaldella, A. Kikot, J.O. Zerbino and J.C. Tara*
20-P-15 Diffusivities of zeolite coatings, *M. Tatlier, S.B. Tantekin-Ersolmaz and A. Erdem-Senatalar*
20-P-16 Crystal growth mechanism of LTA and FAU and densification process of zeolite film by seed growth, *I. Kumakiri, Y. Sasaki, W. Shimidzu, T. Yamagushi and S.-I. Nakao*
20-P-17 In-situ synthesis of ZSM-5 on aluminum surfaces, *F. Scheffler and W. Schwieger*
20-P-18 Conceptual process design of an all zeolite membrane reactor for the hydroisomerization of C_5/C_6, E.E. McLeary, *R.D. Sanderson, C. Luteijn, E.J.W. Buijsse, L. Gora, Th. Maschmeyer and J.C. Jansen*

21 Nanocomposite Fundamentals and Applications

21-K-01 Zeolite-based nanocomposites: synthesis, characterization and catalytic applications, *B.V. Romanovsky*
21-O-02 Methods of synthesis for the encapsulation of dye molecules in molecular sieves, *M. Wark, M. Ganschow, Y. Rohlfing, G. Schulz-Ekloff and D. Wöhrle*
21-O-03 MCM-41 silica monoliths and diluted magnetic semi-conductors: a promising union for fabricating nanosized quantum wires, *F. Brieler, M. Brehm, L. Chen, P.J. Klar, W. Heimbrodt and M. Fröba*
21-O-04 Potential microlasers based on $AlPO_4$-5/DCM composites, *Ö. Weiß, F. Schüth, L. Benmohammadi and F. Laeri*
21-O-05 Light-emitting BN, Si, and SiC nanoparticles encapsulated in molecular sieves, *Xiaotian Li, C. Shao, F. Gao, S. Qiu, F.-S. Xiao and O. Terasaki*
21-P-06 Fabrication of hollow fibers and spheres composed of zeolites by layer-by-layer adsorption method, *Y. Tang, Y.-J. Wang, X.-D. Wang, W.-L. Yang and Z. Gao*
21-P-07 The zeolitisation of diatoms to create hierarchical pore structures, *S.M. Holmes, R.J. Plaisted, P. Crow, P. Foran, C.S. Cundy and M.W. Anderson*
21-P-08 Generating the narrowest single-walled carbon nanotubes in the channels of $AlPO_4$-5 single crystals, *G.D. Li, Z.K. Tang, N. Wang, K.H. Wong and J.S. Chen*
21-P-09 Zeolite- an effective nucleating agent of $Na_2HPO_4 \cdot 12H_2O$, *J. Dong, X. Jing and Yu. Zhang*
21-P-10 Synthesis and characterization of SnO_2 nano particles in zeolite hosts, *Yi. Zhang, Xu. Wang and Xi. Wang*
21-P-11 Encapsulation of $Mn(bipy)_2$ into the zeolite Y prepared via different routes, *B. Fan, W. Cheng and R. Li*
21-P-12 Preparation of zeolite Beta/polystyrene beads and the corresponding hollow spheres, *V. Valtchev and S. Sferdjella*
21-P-13 Synthesis, characterization and catalysis of manganese(II) complexes encapsulated in NaX and NaY zeolites, *J.M. Silva, R. Ferreira, C. Freire, B. de Castro and J. L. Figueiredo*
21-P-14 Guest-host interactions in systems containing liquid crystals confined to molecular sieves, *S. Frunza, L. Frunza, A. Schönhals, H.-L. Zubowa, H. Kosslick and R. Fricke*
21-P-15 Zeolite Beta ordered macroporous structures with improved mechanical strength and controlled mesoporosity, *V. Valtchev, S. Sferdjella and H. Kessler*
21-P-16 Synthesis of zeolites with organic lattice, *K. Yamamoto, Y. Takahashi and T. Tatsumi*
21-P-17 Crystallization mechanism of AlMePO-(, *Y. Qi, G. Wang and Z. Liu*
21-P-18 Crystal structure and magnetic properties of rubidium clusters in zeolite LTA, *T. Ikeda and T. Kodaira*

22 Advanced Materials

22-O-01 The effect of the location of framework negative charge on the ordering of templates in zeolite IFR, *R.E. Morris and L.A. Villaescusa*
22-O-02 A new family of microporous vanadium phosphates and related compounds with organic coordination, *S. Feng, Z. Shi, L. Zhang, H. Zhao, D. Zhang and Z. Dai*
22-O-03 Catalytic properties of novel nickel(II) phosphate with nanoporous structure, *J.-S. Chang, D.S. Kim, S.-E. Park, P.M. Forster, A.K. Cheetham and G. Ferey*
22-O-04 Characterization of corrosion-resistant zeolite coatings on Al alloys, *H. Wang, Zhen. Wang, X. Cheng, A. Mitra, L. Huang and Y. Yan*
22-O-05 Template synthesis and catalysis of bimetallic platinum-rhodium and -palladium nanowires in mesoporous materials, *A. Fukuoka, Y. Sakamoto, S. Inagaki, N. Sugimoto, Y. Fukushima and M. Ichikawa*
22-P-06 Tailored generation of titanium oxide species within porous Si-MCM-41, *P. Prochnow, G. Schulz-Ekloff, M. Wark, J.K. Thomas, A. Zukal and J. Rathousky*
22-P-07 Optical switching with photochromic dye molecules encapsulated in the pores of molecular sieves by in-situ synthesis, *C. Schomburg, D. Wöhrle, G. Schulz-Ekloff and M. Wark*
22-P-08 Formation of carbon nanotubes on various molecular sieves supported metal oxides, *A.M. Zhang, Q.H. Xu, J.J. Zhao and J.M. Cao*
22-P-09 Encapsulation of Tb[(ClBOEP)$_4$P](acac) in Si-MCM-41 by the method of ship-in-bottle and its luminescent properties at 77 K, *Q. Xu, Z. Zhao, L. Li, G. Liu, H. Ding, J. Yu and R. Xu*
22-P-10 A new adsorbent with magnetic properties based on natural clinoptilolite, V. Pode, V. Georgescu, V. Dalea, R. Pode, and E. Popovici
22-P-11 Preparation of microcalorimetric gas sensors with CoAPO-5, *S. Mintova, J. Visser and T. Bein*
22-P-12 Study of cation-exchange properties of an organozeolite, *V.A. Nikashina, E.M. Kats, I.V. Komarova, N.K. Galkina and K.I. Sheptovetskaja*
22-P-13 Advanced electrode materials based on mesoporous aluminum-stabilized anatase, *A. Attia, S.H. Elder, R. Jirásek, L. Kavan, P. Krtil, J. Rathouskŧ and A. Zukal*
22-P-14 Dye-zeolite assemblies for optical sensing applications, *J.L. Meinershagen and T. Bein*
22-P-15 A new sorbent based on clinoptilolite-containing tuff modified by polyethylene, *I.N. Meshkova, V.A. Nikashina, T.M. Ushakova, V.G. Grinev, N.Yu. Kovaleva, A.A. Zaborskii, T.A. Ladygina and L.A. Novokshonova*
22-P-16 Molecular sieves from pillaring of some romanian bentonite, *E. Popovici, I. Bedelean, D. Pop, G. Singurel, D. Macocinschi and H. Bedelean*
22-P-17 Electronic states and arrangements of AgI and CuI clusters incorporated into zeolite LTA, T. Kodaira and T. Ikeda
22-P-18 PbI_2 nanoclusters in zeolite LTL: host-guest chemistry and optical properties, *G. Telbiz, O. Shvets, V. Vozny and M. Brodyn*
22-P-19 Application of the molecular sieves as matrices for the pigments, *S. Kowalak, A. Jankowska, N. Pietrzak and M. Strózyk*
22-P-20 Laser dye doped mesoporous silica fibers: host-guest interaction and fluorescence properties, *G. Telbiz, O. Shvets, S. Boron, V. Vozny, M. Brodyn and G. Stucky*
22-P-21 Spectroscopic properties of dye-loaded mesoporous silicas of the structural type MCM-41, *B. Onida, B. Bonelli, M. Lucco-Borlera, L. Flora, C. Otero Areán and E. Garrone*

23 Micro- and Mesoporous Materials in Fine Chemistry

23-K-01 Delaminated zeolites as active catalysts for processing large molecules, *A. Corma and V. Fornés*
23-O-02 Pd-zeolites as catalysts for the Heck reaction: a screening of reaction parameters affecting catalyst heterogeneity, *M. Dams, D.E. De Vos, L. Drijkoningen and P.A. Jacobs*
23-O-03 Beckmann rearrangement of cyclohexanone oxime over mesoporous MCM-41 and MCM-48 type materials, *R. Gläser, H. Kath and J. Weitkamp*
23-O-04 Knoevenagel condensation between ethylcyanoacetate and benzaldehyde over base catalysts immobilized on mesoporous materials, *Y. Choi, K.-S. Kim, J.-H. Kim and G. Seo*
23-O-05 One-step synthesis of MIBK from acetone over Pt/X catalysts, *L.V. Mattos, F.B. Noronha and J.L.F. Monteiro*
23-P-06 Selective hydroxyethylation of furfuryl alcohol with aqueous acetaldehyde in the presence of H-form zeolites, *A. Finiels, W. Balmer and C. Moreau*
23-P-07 Selective synthesis of monooctylamines by ammonia alkylation with octanol using NaY, ZSM-5, SAPO-5, SAPO-11, SAPO-31, SAPO-34, *S. Amokrane, R. Rebai, S. Lebaili, D. Nibou and G. Marcon*
23-P-08 Conversion of monoethanolamine in other organic nitrogen compounds on H-mordenite and H-clinoptilolite, *G. Torosyan, S. Sargsyan and A. Grigoryan*
23-P-09 The influence of ammonia adsorption on Y Zeolite and natural clinoptilolite activity in ethanol transformation, *L. Akhalbedashvili, A. Mskhiladze and Sh. Sidamonidze*
23-P-10 Enantioselective synthesis and separation of terminal epoxides and diols using a catalytic membrane system containing chiral Co(III) salen, *S.-D. Choi and G.-J. Kim*
23-P-11 Asymmetric trimethylsilylcyanation of benzaldehyde catalyzed by chiral Ti(IV) salen complexes immobilized on MCM-41, *J.-H. Kim and G.-J. Kim*
23-P-12 Mechanistic study of aniline methylation over acidic and basic zeolites Y, *I.I. Ivanova, E.B. Pomakhina, A.I. Rebrov, Yu.G. Kolyagin, M. Hunger and J. Weitkamp*
23-P-13 Heterogeneous base catalysis: characterization of zeolites and mixed oxides using nitromethane as a NMR probe molecule and activity in the Michaël condensation of nitromethane and cyclohex-2-en-1-one, *E. Lima, L.-C. de Ménorval, M. Laspéras, J.-F. Eckhard, D. Tichit, P. Graffin and F. Fajula*
23-P-14 Synthesis of (-pinene derivatives using redox-mesoporous molecular sieves, *Y.-W. Suh, T.-M. Son, N.-K. Kim, W.-S. Ahn and H.-K. Rhee*
23-P-15 Ring opening reactions of methyloxirane over DZSM-5 and DAlMCM-41 molecular sieves - A mechanistic study, *A. Fási, I. Pálinkó, Á. Gömöry and I. Kiricsi*
23-P-16 Hydrodechlorination of1, 2,4-trichlorobenzene on Ni/Al-MCM-41

catalysts, *Y. Cesteros, P. Salagre, F. Medina, J.E. Sueiras and G.L. Haller*
23-P-17 Adsorption of cytochrome c onto ordered mesoporous silicates, *J. Deere, E. Magner, J.G. Wall and B.K. Hodnett*
23-P-18 Vapor phase Beckmann rearrangement of cyclohexanone oxime over tantalum pillared magadiite, *S.J. Kim, M.H. Kim, Y. Ko, G. Seo and Y.S. Uh*
23-P-19 Hydration of ?-pinene over heteropolyacids encaged in USY zeolites, *J. Vital, A.M. Ramos, I.F. Silva, J.E. Castanheiro, M.N. Blanco, C. Caceres, P. Vasquez, L. Pizzio and H. Thomas*
23-P-20 Selective adsorption of trans unsaturated fatty acid compounds in MFI type zeolites, *S. Paulussen, M. Goddeeris and P.A. Jacobs*
23-P-21 Novel delaminated zeolites are more active acid catalysts than conventional zeolites and mesoporous Al/MCM-41 for the synthesis of fine chemicals, *M.J. Climent, A. Corma, V. Fornés, H. García, S. Iborra, J. Miralles and I. Rodríguez*
23-P-22 The design of zeolites catalysts for the synthesis of orange blossom and apple fragrances, *M.J. Climent, A. Corma and A. Velty*
23-P-23 Catalytic in-situ infrared spectroscopic study of n-butyraldehyde aldol condensation, *U. Rymsa, M. Hunger and J. Weitkamp*
23-P-24 Oxyhalogenation of aromatic compounds in presence of KCl or KBr and H_2O_2 over zeolites, *N. Narender, P. Srinivasu, S.J. Kulkarni and K.V. Raghavan*
23-P-25 Synthesis and characterization of mesoporous Pt-MCM-41 and its application in enantioselective hydrogenation of 1-phenyl-1,2-propanedione, *E. Toukoniitty, B. Sevcíková, N. Kumar, P. Mäki-Arvela, T. Salmi, J. Väyrynen, T. Ollonqvist, E. Laine, P.J. Kooyman and D. Yu. Murzin*
23-P-26 Isomerization of p-eugenol on palladium-containing zeolites, *Ts.M. Ramishvili, M.K. Charkviani and L.D. Kashia*
23-P-27 The use of MCM-22 as catalyst for the Beckmann-rearrangement of cyclohexanone oxime to (-caprolactam, *G. Dahlhoff, U. Barsnick, W. Eickelberg and W.F. Hölderich*
23-P-28 Nickel supported on zirconium doped mesoporous silica as catalysts for the gas phase hydrogenation of acetonitrile, *P. Braos-García, L. Díaz, P. Maireles-Torres, E. Rodríguez-Castellón and A. Jiménez-López*
23-P-29 Synthesis of fine chemicals intermediates over basic zeolites, *C.O. Veloso, A.C. Pinto, E.N. Santos and J.L.F. Monteiro*
23-P-30 Selective chlorination of diphenylmethane over zeolite K-L, *A.P. Singh and S.M. Kale*
23-P-31 Butylation of phenol on medium pore AlPO4-11, -31 and -41 structures: effect of silicon incorporation, *C.V. Satyanarayana, U. Sridevi and B.S. Rao*
23-P-32 The catalytic synthesis of the glycidol from the glycerol carbonate in presence of zeolite A, *J.W. Yoo and Z. Mouloungui*
23-P-33 Transfer hydrogenation of unsaturated ketones MCM-41, *J. Wahlen, D.E. De Vos, M. De bruyn, P.J. Grobet and P.A. Jacobs*

24 New Routes to Hydrocarbon Activation

24-O-01 Dehydroisomerization of n-butane to isobutene over Pd modified silicoaluminophosphate molecular sieves, *Y. Wei, G. Wang, Z. Liu, C. Sun and L. Xu*
24-O-02 Conversion of methane over Ag-Y in the presence of ethene, *T. Baba, H. Sawada, Y. Abe and Y. Ono*
24-O-03 Peculiarities in the hydroconversion of n-hexadecane over bifunctional catalysts, *L. Perrotin, A. Finiels, F. Fajula and T. Cholley*
24-O-04 H(catalyzed heterogeneous aziridination of olefins, *B. Chanda, R. Vyas, A.V. Bedekar, B.B. Kasture and V.N. Joshi*
24-O-05 MCM-41 as support for metallocene catalysts - ethylene polymerization, *C.A. Henriques, M.F.V. Marques, S. Valange, Z. Gabelica and J.L.F. Monteiro*
24-P-06 Study of coke and deactivation over H-Beta zeolite, *Z. Zhu, T. Ruan, Q. Chen, W. Chen and D. Kong*
24-P-07 Photoionization of N-alkylphenothiazines in transition-metal-ion modified mesoporous silica SBA-15 molecular sieves, *Z. Luan and L. Kevan*
24-P-08 Potential use of AlMCM-41 for activation of metallocene catalyst, *T. Sano, T. Niimi, T. Miyazaki, S. Tsubaki, Y. Oumi and T. Uozumi*
24-P-09 Activation of butanes with olefins carbenium cations over zeolite catalysts, *S.E. Dolinsky and V.A. Plakhotnik*
24-P-10 Immobilization and mobilization of surface species during transformation of ethylene over HZSM-5 catalysts, *Zikánová, M. Kocirík, M. Derewinski, P. Sarv, J. Dubskt, P. Hudec and A. Smiesková*
24-P-11 Zeolite-L as support of Fe microcystals for the Fischer-Tropsch synthesis, *M.V. Cagnoli, N.G. Gallegos, A.M. Alvarez, J.F. Bengoa, A.A. Yeramian and S.G. Marchetti*
24-P-12 Nb- and Ti-containing silica-based mesoporous molecular sieves as catalysts for photocatalytic oxidation of methane, *J. Xin, X. Chen, J. Suo, Xia. Zhang, L. Yan and Shuben Li*
24-P-13 Catalytic properties of micelle templated microporous and mesoporous materials for the conversion of low-density polyethylene, *J. Aguado, D.P. Serrano, R. Van Grieken, J.M. Escola and E. Garagorri*
24-P-14 Epoxidation of propylene in fixed bed reactor using supported titanium silicalite catalyst, *X.S. Wang, Gang Li, H.S. Yan and X.W. Guo*
24-P-15 Acetylene and alkene oligomerization on ETS-10 having induced Brønsted acidity, *A. Zecchina, C. Pazè, C. Otero Areán, G. Turnes Palomino, F.X. Llabrési Xamena and S. Bordiga*
24-P-16 Isomerization of n-butane over small crystals of H-Beta and Pt-H-Beta zeolite catalysts, *N. Kumar, M. Vaini, V. Nieminen, R. Byggningsbacka, L.-E. Lindfors, T. Salmi, D. Yu. Murzin and E. Laine*
24-P-17 Ethylene oligomerization with nickel-containing NaX zeolite, *M.O. de Souza, F.M.T. Mendes, R.F. de, Souza, J.H. Z. dos Santos, L. Caumo, V. Conz, F. Majolo and L. V. Barbosa*
24-P-18 Studies of the methanol to hydrocarbons reaction using isotopic labeling: mounting evidence for a hydrocarbon pool mechanism, *S. Kolboe*
24-P-19 Formation and reactivity of alkoxy species through the reaction of alkylhalides with metal-exchanged zeolites, *R.J. Corrêa and C.J.A. Mota*
24-P-20 The use of ITQ-7 as catalyst for alkylation of isobutane with 2-butene, Corma, *M.J. Díaz-Cabañas, C. Martínez and S. Valencia*
24-P-21 Butene isomerization over ferrierite and SUZ-4 zeolite, *V.L. Zholobenko and C.L.T. Stevens*
24-P-22 Volatile products of the conversion of cyclohexene over Al-MCM-41, *M. Rozwadowski, M. Lezanska, J. Wloch, K. Erdmann, G. Zadrozna and J. Kornatowski*
24-P-23 Cu-Y zeolite catalysts for methanol and ethanol steam reforming, *M. Laniecki*
24-P-24 Hexenes obtaining on the nickel - ion exchanged zeolites, *M.K. Munshieva*
24-P-25 Catalytic sites of mesoporous silica in degradation of polyethylene, *A. Satsuma, T. Ebigase, Y. Inaki, H. Yoshida, S. Kobayashi, Md.A. Uddin, Y. Sakata and T. Hattori*
24-P-26 The nature of medium acidity in [CuO/ZnO/ZrO_2]SAPO-34 hybrid catalyst for CO_2 hydrogenation: the study of the interactions between metal oxides and acid sites in zeolite, *S.-K. Ihm, S.-W. Baek, Y.-K. Park and K.-C. Park*
24-P-27 Reaction pathways for the aromatization of cyclohexane and cyclohexene on Zn/H-ZSM-5 zeolites, Urda, *G. Telbiz and I. Sandulescu*
24-P-28 Coke species and coking mechanism of SAPO-34 in MTO process, *Y. Qi, G. Wang, Z. Liu, L. Xu, X. Gao and W. Cui*
24-P-29 Pt-2,2'bipyridine complex encapsulated in Y zeolite - catalysts for ethylene selective dimerization, *R. Zavoianu and E. Angelescu*
24-P-30 Aromatics formation from C_4-C_4= technical fraction over zinc- and zinc/copper-containing ZSM-5 zeolites, *N. Bilba, Gh. Iofcea, I. Asaftei, D.M., Padurariu and C.C. Pavel*
24-P-31 Aromatization of mixed-C_4 hydrocarbons over the HZSM-5 catalysts modified by Zn and Ni cations, *L. Wei, J.Z. Gui, H.S. Ding, X.T. Zhang, H.Y. Li, L. Song, Z.L. Sun and L.V.C. Rees*

25 Conversion of Aromatics

25-O-01 Shape-selective methylation of 4-methybiphenyl into 4,4'-dimethybiphenyl over modified ZSM-5 catalysts, *J.-P. Shen, L. Sun and C. Song*
25-O-02 Facile Friedel-Craft's alkylation of phenol with 4-hydroxybutan-2-one over (and Y zeolites to produce raspberry ketone, *K.K. Cheralathan, I.S. Kumar, B. Arabindoo, M. Palanichamy and V. Murugesan*
25-O-03 Selective alkylation of naphthalene to 2,6-dimethyl-naphthalene catalyzed by MTW zeolite, *G. Pazzuconi, G. Terzoni, C.Perego and G. Bellussi*
25-O-04 Transalkylation reaction of phenol with trimethylbenzenes over Y and EMT zeolites, *V. Hulea, I. Fechete, P. Caullet, H. Kessler, T. Hulea, C. Chelaru, C. Guimon and E. Dumitriu*
25-O-05 Benzene alkylation with alkanes over modified MFI catalysts, *A.V. Smirnov, E.V. Mazin, O.A. Ponomoreva, E.E. Knyazeva, S.N. Nesterenko and I.I. Ivanova*
25-P-06 Isopropylation of napthalene over large pore zeolites, *R.K. Ahedi, S. Tawada, Y. Kubota and Y. Sugi*
25-P-07 Shape-selective tert-butylation of biphenyl over HM, HY and H(zeolites in the liquid phase, *D. Mravec, J. Horniakov(, M. Kr(lik, M. Hronec, J. Joffre and P. Moreau*
25-P-08 1-Acetyl-2-methoxynaphthalene isomerization over zeolites. Effect of pore structure, *V. Moreau, E. Fromentin, P. Magnoux and M. Guisnet*
25-P-09 Alkylation of phenol with propylene over solid acid catalysts, *B. Wang, C.W. Lee, T.-X. Cai and S.-E. Park*
25-P-10 Transalkylation of trimethylbenzene with toluene over large pore zeolites, J. *Cejka, A. Krejcí and J. Hanika*
25-P-11 Physicochemical characterization and catalytic activity of Al-HMS for N-methylation of aniline, *J.M. Campelo, A. Garcia, D. Luna, J.M. Marinas, A.A. Romero and J.J. Toledano*
25-P-12 Catalytic activity of secondary aluminated mesoporous molecular sieve AlMCM-41 in the Friedel-Crafts reaction of bulky aromatic compounds, *H. Hamdan , A.B. Kim and M.N. Mohd Muhid*
25-P-13 Naphthalene alkylation with methanol employing solid catalysts, *J. Aguilar-P, A. Corma, J.A. de los Reyes, L. Noreña, G. Muñoz, J.M. Sanchez, Torales and I. Hernández*
25-P-14 Alkylation of biphenyl and naphthalene with propene. Is zeolite Beta a shape-selective catalyst?, *D.M. Roberge and W.F. Hölderich*
25-P-15 Alkylation of benzene by propane with participation of space divided centres, *S.I. Abasov, R.R. Zarbaliyev, G.G. Abbasova, D.B. Tagiyev and M.I. Rustamov*
25-P-16 Alkylation of isopropylnaphthalene over heteropoly acid catalysts supported on mesoporous materials, *M.-W. Kim, W.-G. Kim, J.-H. Kim, Y. Sugi and G. Seo*
25-P-17 Highly selective isopropylation of xylenes catalyzed by zeolite Beta, *C.R. Patra, S. Kartikeyan and R. Kumar*

26 Catalysis for Oil Refining

26-O-01 The isomerization selectivity in FCC process, *L.-J. Yan, M.-Y. He, J. Fu and J. Long*
26-O-02 Design of zeolite catalyst for paraffin isomerisation, *J. Houzvicka, C.J.H. Jacobsen and I. Schmidt*
26-O-03 Cyclohexane ring opening on metal-zeolite catalysts, *T.V. Vasina, O.V. Masloboishchikova, E.G. Khelkovskaya-Sergeeva, L.M. Kustov and P. Zeuthen*

26-O-04 Selective ring opening of naphthenic molecules, *M. Daage, G.B. Mc Vicker, M.S. Touvelle, C.W. Hudson, D.P. Klein, B.R. Cook, J.G. Chen, S. Hantzer, D.E.W. Vaughan and E.S. Ellis*
26-O-05 Reforming of FCC heavy gasoline and LCO with novel borosilicate zeolite catalysts, *C.Y. Chen and S.I. Zones*
26-P-06 Hydroisomerization of n-decane in the presence of sulfur. Effect of metal-acid balance and metal location, *L.B. Galperin, S.A. Bradley and T.M. Mezza*
26-P-07 Hydrodesulfurization of benzothiophene over noble metals supported on mesoporous silica MCM-41, *M. Sugioka, A. Seino, T. Aizawa, J.K.A. Dapaah, Y. Uemichi and S. Namba*
26-P-08 Catalytic functionalities of USY zeolite supported hydrotreating catalysts, *K.S. Rawat, M.S. Rana and G. Murali Dhar*
26-P-09 Highly active, selective and stable ferrierite-based catalysts for the skeletal isomerization of n-C_5-C_7, *C.P. Nicolaides, J. Makkonen and M. Tiitta*
26-P-10 Producing synthetic steamcracker feed from cycloalkanes (or aromatics) on various zeolite catalysts, *A. Raichle, H. Scharl, Y. Traa and J. Weitkamp*
26-P-11 n-Heptane hydroconversion and methylcyclohexane cracking as model reactions to investigate the pore topology of Nu-88 zeolite, *S. Lacombe, A. Patrigeon and E. Benazzi*
26-P-12 New Mo and NiMo hydrodesulfurization catalysts supported on Al-MCM-41. Effect of the support Si/Al molar ratio, *T. Klimova, M. Calderón and J. Ramírez*
26-P-13 Hydrogenation and ring opening of mono- and diaromatics for Diesel upgrading on Pt/Beta catalysts, *M.A. Arribas, J.J. Mahiques and A. Martínez*
26-P-14 Hydro denitrogenation activity of NiO - MoO_3 catalysts supported on various mesoporous alumino silicates, *K. Shanthi, N.R. Sasi Rekha, R. Moheswari and T. Sivakumar*
26-P-15 Model hydrocracking catalysts combining NiMo sulfide and large-pore zeolite: effect of the zeolite nature on the location of NiMo sulfide in relation with catalytic properties, *J. Leglise, D. Cornet, M. Baâlala, C. Potvin and J.-M. Manoli*
26-P-16 Effect of zeolite acidity characteristics on the deactivation behavior of bifunctional large-pore zeolite catalysts during cyclopentane hydroconversion, *S. Gopal and P.G. Smirniotis*
26-P-17 Characterization and catalytic activities of MCM-41 supported WS_2 hydrotreating catalysts, *T. Chiranjeevi, P. Kumar, M.S. Rana, G. Murali Dhar and T.S.R. Prasada Rao*
26-P-18 Isomerization and hydrocracking of n-heptane and n-decane over bifunctional mesoporous molecular sieves, *C. Bischof and M. Hartmann*
26-P-19 Isomerization of cyclohexane, n-hexane and their mixtures on zeolite catalyst, *A. Holló, J. Hancsók and D. Kalló*
26-P-20 Application of adsorption Dubinin-Radushkevich equation for study of n-pentane and m-xylene conversion catalysts microporous structure, *S.B. Agayeva, B.A. Dadashev, S.I. Abasov and D.B. Tagiyev*
26-P-21 Hydroisomerization of n-hexadecane over Pt/Al-MCM-41 catalysts: two different Al incorporation methods, *K.-C. Park and S.-K. Ihm*
26-P-22 Zr-containing hexagonal mesoporous silicas as supports for hydrotreating catalysts, *N.G. Kostova, A.A. Spojakina, L.A. Petrov, O. Solcova and K. Jiratova*
26-P-23 New catalysts for isomerization of long-chain n-paraffins, *M.I. Levinbuk, L.M. Kustov, T.V. Vasina, O.V. Masloboishchikova, M.L. Pavlov, I.E. Gorbatkina and V.A. Khavkin*

27 Selective Oxidation and Sulfur Resistance

27-O-01 Singular catalytic properties of Ti-MWW in the selective oxidation of alkenes, *P. Wu, T. Komatsu, T. Yashima and T. Tatsumi*
27-O-02 Epoxidation of propylene over TS-1 containing trace aluminum, *X. Guo, Xi. Wang, Min Liu, Gang Li, Yo. Chen and J. Xiu*
27-O-03 One step benzene oxidation to phenol using N_2O over acid zeolites, *G. Juttu and R.F. Lobo*
27-O-04 Dual pathways for benzene hydrogenation on Pt/mordenites: implication for sulfur tolerance, *L. Simon, J.G. van Ommen, A. Jentys and J.A. Lercher*
27-O-05 Sulfur resistance of PtPd catalysts: preparation, characteri-zation and catalytic testing, *K. Thomas, C. Binet, T. Chevreau, D. Cornet and J.-P. Gilson*
27-P-06 Microporous metallosilicates for the oxidation of hydrocarbons: preparation, characterization and catalytic activity, *U. Arnold, R.S. da Cruz, D. Mandelli and U. Schuchardt*
27-P-07 High catalytic activity of Fe(III)-substituted alumino-phosphate molecular sieves (FeAPO) in oxidation of aromatic compounds, *X. Meng, Y. Yu, L. Zhao, J. Sun, K. Lin, M. Yang, D. Jiang, S. Qiu and F.-S. Xiao*
27-P-08 Selective oxidation of propyl alcohols over zeolites modified with cations of the transition metals, *A.M. Aliyev, D.B. Tagiyev, S.M. Medzhidova, S.S. Fatullayeva, A.R. Kuliyev, T.N. Shakhtakhtinsky, G.A. Ali-zade and K.I. Matiyev*
27-P-09 Niobium leaching from the catalysts applied in the sulfoxidation of thioethers with hydrogen peroxide, *M. Ziolek, A. Lewandowska, M. Renn, I. Nowak, P. Decyk and J. Kujawa*
27-P-10 Biomimetic oxygen transfer by Co and Cu complexes immobilized in porous matrices, *K. Hernadi, I. Pálinkó, E. Böngyik and I. Kiricsi*
27-P-11 Titanium molecular sieves convert hydrogen peroxide into $1O_2$, *F.M. van Laar, D.E. De Vos, P. Grobet, J.-M. Aubry, L. Fiermans and P.A. Jacobs*
27-P-12 Propane oxidation on Cu/ZSM-5 catalyst: the effect of copper and aluminum content in the reducibility and in the activity of Cu active species, *M.S. Batista and E.A. Urquieta-González*
27-P-13 Oxidizing conversion of isobutanol on zeolites, *S. Zulfugarova*
27-P-14 Photocatalytic production of H_2O_2 over heterogenized quinone in zeolite, *J.S. Hwang, C.W. Lee, H.S. Chai and S.-E. Park*
27-P-15 Liquid-phase oxidation of cyclohexane in the presence of chromium and iron ETS-10 materials, *A. Valente, P. Brandão, Z. Lin, F. Gonçalves, I. Portugal, M.W. Anderson and J. Rocha*
27-P-16 Effect of oxygen concentration on catalyst deactivation rate in vapor phase Beckmann rearrangement over acid catalysts, *T. Takahashi and T. Kai*
27-P-17 On the role of the titanium active site in the phenol/anisole hydroxylation over titanium substituted crystalline silicates, *U. Wilkenhöner, D.W. Gammon and E. van Steen*

28 Confinement and Physical Chemistry for Catalysis

28-O-01 Reactivity enhancement by molecular traffic control - a consequence of released single-file constraints, *P. Bräuer and J. Kärger*
28-O-02 Aromatization of n-hexane over ZnNi/HZSM-5 catalyst induced by microwave irradiation, *J.Z. Gui, H.S. Ding, N.N. Liu, Y.R. Gao, Z.L. Cheng, X.T. Zhang, B. Ma, L. Song, Z.L. Sun and L.V.C. Rees*
28-O-03 Artificial photosynthesis using zeolites, *N. Castagnola and P.K. Dutta*
28-O-04 Synthesis of macrocycles using molecular sieve catalysts, *M. Radha Kishan, N. Srinivas, S.J. Kulkarni, M. Ramakrishna Prasad*
28-O-05 Effect of single-file diffusion on the hydroisomerization of 2,2-dimethylbutane on platinum loaded H-mordenite, *F.J.M.M. de Gauw, J. van Grondelle, R.A. van Santen G. Kamalakar and K.V. Raghavan*
28-P-06 Use of coke-selectivated H-ZSM-5 in xylene isomerization, *F. Bauer and A. Freyer*
28-P-07 Photocatalytic reactions on chromium containing mesoporous molecular sieves under visible light irradiation: decomposition of NO and partial oxidation of propane, *H. Yamashita, K. Yoshizawa, M. Ariyuki, S. Higashimoto and M. Anpo*
28-P-08 Enhancing the shape selectivity of nanocrystalline HZSM-5 zeolite via comprehensive modifications, *H.C. Guo, X.S. Wang and G.R. Wang*
28-P-09 Nature of shape-selective catalysis in the ethylation and the isopropylation of biphenyl over H-mordenites, *Y. Sugi, S. Tawada, T. Sugimura, Y. Imada, Y. Kubota, T. Hanaoka and T. Matsuzaki*
28-P-10 Adsorption of selected amino acids from aqueous solutions on mesoporous molecular sieves, *S. Ernst, M. Hartmann and S. Munsch*
28-P-11 Influence of OH groups of Beta zeolites on the synthesis of MTBE, *F. Collignon and G. Poncelet*
28-P-12 About a possibilities of effectiveness increasing of porous catalyst granules with controlled activity profile, *V.V. Andreev*
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07-R-08- An XPS Study of Ordered (Ti-MCM-41, [Ti]-MCM-41) and Amorphous (MST) Mesoporous Silica-Titania, *G. Moretti, A.M. Salvi, M.R. Guascito, G. Ferraris, M. Guidotti and R. Psaro*

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07-R-10- Preparation of Thermally Stable Meso-Organized Pure Titania Thin Film by Dip-Coating, *D. Grosso, G.J. A.A. de Soler-Illia, E.L. Crepaldi, F. Babonneau and C. Sanchez*

07-R-11- Synthesis and Characterization of Ce-Containing MCM-41, *S.-H. Chien and C.-L. Chen*

07-R-12- Synthesis and Characterization of DAM-1 Molecular Sieves, *K.J. Balkus, Jr., D. Coutinho and Y. Ma*

8 Synthesis with non-ionic surfactants

08-R-01- Block Copolymers as Templates for Mesoporous SiO_2 Synthesis, *K. Flodstrom, V. Alfredsson, P. Kipkemboy and A. Fogden*

08-R-02- Synthesis and Characterization of Thin Films of TiO_2 and SiO_2 with Cubic, Hexagonal and Lamellar Mesostructures, *K.L. Frindell, P. Alberius-Henning, R.C. Hayward, E.J. Kramer, B.F. Chmelka and G.D. Stucky*

9 Crystal structure determination

09-R-01- Rehydration Mechanisms in Zeolites: Reversibility of T-O-T Breaking and of Tetrahedral Cation Migration in Brewsterite, *A. Alberti, G. Vezzalini, S. Quartieri, G. Cruciani and S. Bordiga*

09-R-02- Localization of Ruthenium-Red in Zeolite NaY, *C.E.A. Kirschhock, K. Possemiers and P.A. Jacobs*

09-R-03- True Structure of Trigonal Bipyramidal SiO_4F- Species in Siliceous Zeolites, *M.P. Attfield, C.R.A. Catlow and A.A. Sokol*

09-R-04- Thermal Behaviour and Dehydration Mechanism of Bikitaite, *G. Vezzalini, O. Ferro, S. Quartieri, A.F. Gualtieri, G. Cruciani, E. Fois, C. Ceriani and A. Gamba*

09-R-05- The New Web Database of Zeolite Structures of the IZA Structure Commission www.iza-structure.org /databases, *C. Baerlocher and L.B. McCusker*

09-R-06- Can you Write the Crystal Chemical Formula for your Favorite Zeolite? Some Guidelines to the New IUPAC Recommendations, *L.B. McCusker, F. Liebau and G. Engelhardt*
09-R-07- The Crystal Structure of as Synthesized TPA-Silicalite-1: Fluoride Counter Anions Localization and Twin Analysis, *E. Aubert, F. Porcher, M. Souhassou, Y. Dusausoy and C. Lecomte*
09-R-08- Synthesis and Structure Refinement of Zinc Gallophosphate with Laumontite Topology, *N. Z. Logar, M. Helliwell, M. Mrak, A. Ristic and V. Kaucic*

10 Host-guest chemistry

10-R-01- Adsorbed Amount Dependence of Behavior of p-Nitroaniline in the Micropore of ZSM-5, *Y. Komori and S. Hayashi*
10-R-02- Immobilization of Chloroperoxidase inside Mesocellular Foam, *Y.-H. Han, N. Maier, G.D. Stucky and A. Butler*
10-R-03- $[PdCl_2(NH_2(CH_2)_{12}CH_3)_2]$ Heterogeneized on Zeolites as Hydrogenation Catalyst, *J.A. Diaz-Aunon, J. Garcia-Martinez, M.C. Roman-Martinez and C. Salinas-Martinez de Lecea*
10-R-04- Synthesis and Characterization of Insertion Compounds of Indium Halides in Siliceous and Alumosilicate MFI Structures, *M. Hartl, C. Schmidt, E. Löffler, P. Behrens and W. Grünert*
10-R-05- Synthesis and Characterization of Azidoiron(III) Cyclam Complex inside Zeolite-Y, *X. Hu and K. Meyer*

11 Post-synthesis modification

11-R-01- Study of Dealumination of Small Crystal Y Zeolite, *P. Wang, J. Du, Z. Da and M. He*
11-R-02- Removal of Tetramethylammonium Ions from Zeolite Alpha, *O. Kresnawahjuesa, D.H. Olson, R.J. Gorte and G.H. Kühl*
11-R-03- Preparation of Mullite Micro-Vessels by a Combined Treament of Zeolites, *C. Kosanovic, B. Subotic and A. Ristic*
11-R-04- Strong Brönsted Acidity and High Catalytic Activity for Alkane Cracking over Na_2H_2-EDTA-Treated Ultra Stable Y Zeolite, *Y. Kageyama, N. Katada and M. Niwa*
11-R-05- Effect of Reduction and Oxidation Process on Ga/ZSM5 Catalysts, *M. Garcia, P. Magusin, E. Abbenhuis, J. van Wolput, P. Thüne and R.A. van Santen*

12 In-situ spectroscopy and catalysis

12-R-01- In Situ FT-IR Study of But-1-ene Reaction over H-FER: Evidence for Channel Blocking by Slowing Diffusing Hydrocarbon Species, *F.C. Meunier, L. Domokos, A. Kinage, K. Seshan and J.A. Lercher*
12-R-02- Conversion of n-Butene-1 on the Zeolite H-Ferrierite. An in Situ 13C MAS NMR Study, *A.G. Stepanov, M.V. Luzgin, H. Ernst and D. Freude*
12-R-03- Electron Energy Loss Spectroscopy: the Location, Amount and Nature of Carbonaceous Deposits on Zeolite Crystals, *S van Donk, O. Stéphan, F.M.F. de Groot, J.H. Bitter and K.P. de Jong*

13 Frameworks and acid sites

13-R-01- Surface Acidity Diagnosis of Small Pore Size (3.5nm) AlSBA-15 Mesoporous Materials by Laser-Induced Fluorescence (LIF) Spectroscopy, *E. Haddad, A. Lassoued, J.L. Bonardet, J. Fraissard and A. Gédéon*
13-R-02- Motions of the Tetrapropylammonium Structure-Directing Template in a Purely Siliceous MFI Zeolite Studied by Multinuclear Solid State NMR, *A.R. Lewis, B. Meurer, R.D. Gougeon, N. Berdonnier, M. Reinholdt and J.M. Chézeau*
13-R-03- Characterization of Internal and External Acidity of H-ZSM5 Zeolites, *Q. Zhao, W.-H. Chen and S.-B. Liu*
13-R-04- Acidity of the Aluminosilicate Mu-14 (structure code IT), A. Simon, *R.D. Gougeon, J. L. Paillaud and V. Valtchev*
13-R-05- Probing the Mobility of iso-Butyl Acohol inside Zeolite H-ZSM5 by Deuterium Solid State NMR Spectroscopy, Alexander, *G. Stepanov, M.M. Alkaev and A. Shubin*
13-R-06- Disorder in ETS-10, *R.F. Howe and P.D. Southon*
13-R-07- Dealumination Pattern of REUSY Zeolite Contained in Fluid Cracking Catalysts, *F. Hernandez-Beltran, J.C. Moreno-Mayorga, M.L. Guzman-Castillo, J. Navarrete-Bolanos, M., Gonzalez-Gonzalez and B.E. Handy*
13-R-08- Selective Dealumination of Zeolite Beta, *P. Sarv, M. Derewinski and I. Heinmaa*

14 Frameworks, cations, clusters

14-R-01- Physical Characterization of Some MnS Clusters Encapsulated in Natural Zeolites, *F. Iacomi and A. Vasile*
14-R-02- New Opportunities in the Characterization of Paramagnetic Transition Metal Centers in Zeolites Offered by High Field Electron-Nuclear Double Resonance Spectroscopy, *D. Goldfarb, P. Manikandan, B.M. Weckhuysen, R. Grommen, D. Arieli and D.W.E. Vaughan*
14-R-03- Location of Chemisorbed Methylium Ions in Zeolites by Neutron Diffraction and13C MAS NMR, *S. Vratislav, M. Dlouha and V. Bosacek*
14-R-04- Insights into the Structure of Bimetallic Clusters Supported on Mesoporous Silica, *S.T. Bromley, G. Sankar, T. Maschmeyer, J.M. Thomas and C.R.A. Catlow*
14-R-05- Structural Defects and Stability of FER Synthesized with Different Structure Directing Agents, *M. Hironaka, K. Itabashi and M. Nakano*
14-R-06- The Location of the Fluoride Ion in Tetrapropylammonium Fluoride Silicalite-1 Determined by $^1H/^{19}F/^{29}Si$ Triple Resonance CP, REDOR and TEDOR NMR Experiments, *C.A. Fyfe, D.H. Brouwer, A.R. Lewis and J.-M. Chézeau*
14-R-07- Estimation of Extra-Framework Aluminum in AlPO-5 and SAPO-5 Microporous Aluminophosphates by Diffuse Reflectance Spectroscopy, *M.A. Zanjanchi and F. Khadem-Nahvi*
14-R-08- Probing the Connectivity in Intermediate Gel Phases of Molecular Sieve Synthesis by Solid State NMR, *Y. Huang and D. Machado*
14-R-09- Simulation of the ^{29}Si MAS NMR of Silicalite, *E. Martinez Moralez, J.J. Fripiat and L. Javier Alvarez*
14-R-10- CW and Pulsed EPR and Magnetic Measurements on Pt/KL and Pt-Fe/KL, *T. Schmauke, R.-A. Eichel, J. Zheng, E. Roduner and A. Schweiger*
14-R-11- Characterization of Defects and Surface Structures in Zeolites by High Resolution Microscopy Techniques, *G. Gonzalez and R. Riechelt*
14-R-12- Continuous Flow NMR with Hyperpolarized Xenon: Recent Applications, *A. Gédéon, J. Logan, T. Meersmann, J. Fraissard and A. Pines*

16 Modelling and theoretical studies (A, B)

16-R-01- Mathematical Model of Temperature's Anomaly at Coadsorption of Binary Mixture, *N.A. Samoilov*
16-R-02- Theroretical Study on the Stability and Geometry of the a and b Cationic Sites in Mg(II)-Ferrierite, *J.E. Sponer, Z. Sobalik and B. Wichterlova*
16-R-03- Monte Carlo Simulations of Dielectric Relaxation in Na-Mordenites with Various Si/Al Ratios, *G. Maurin, P. Senet, S. Devautour, F. Henn, V.E. van Doren and J.C. Giuntini*
16-R-04- Diffusion of Simple Sorbates in Silicalite: Effect of Anisotropic Frameworks and Geometrical Correlation, *S. Kar and C. Chakravarty*
16-R-05- A Simulation Study on the Topotactic Transformation from $AlPO_4$-21 to $AlPO_4$-25, *J. Li, J. Yu and R. Xu*
16-R-06- Molecular Modelling Studies of Aluminophosphonate Structures, *P. Grewal, P.A. Cox, J.D. Gale and P.A. Wright*
16-R-07- Modelling Hydrated Zeolites: Al Ordering in Goosecreekite, *D.W. Lewis, A.R. Ruiz-Salvador, N. Almora-Barrios, A. Gomez and M. Mistry*
16-R-08- Simulation of Positronium in Silica Sodalite, *P. Hastings, A.L.R. Bug and P. Sterne*
16-R-09- MD Simulation of as-Synthesized $AlPO_4$-34 Triclinic Molecular Sieve, *M. Praprotnik, M. Hodoscek, D. Janezic and S. Hocevar*
16-R-10- Molecular Dynamic Simulations of One-Dimensional Water Molecule Chains in Zeolites: The case of Bikitaite, *P. Demontis, G. Stara and G.B. Suffritti*

17 Principles of adsorption

17-R-01- Thermodynamic and Kinetic Study of Adsorption of Linear and Ramified C_5 Paraffins on Silicalite, *E. Lemaire, J.-M. Simon, J.-P. Bellat, C. Paulin and A. Méthivier*
17-R-02- Localization of Water Molecules and Sodium Ions in Na-Mordenite by Thermally Stimulated Current Measurement, *S. Devautour, G. Maurin, A. Abdoulaye, F. Henn and J.C. Giuntini*

18 Diffusion: fundamentals approach

18-R-01- The New Type of Adsorbers - Sectioning Apparatuses, *N.A. Samoilov*
18-R-02- Azeotropic Adsorption of Organic Solvent Vapor Mixture on High Silica Zeolite, *K. Chihara, H. Suzuki, T. Saito, R. Kamiyama, Y. Takeuchi, C. Mellot and A.K. Cheetham*
18-R-03- Entropy-Based Separation of Mixtures of Linear and Branched Alkanes in the 5-7 Carbon Atom Range by Sorption on Silicalite-1, *S. Calero, B. Smit and R. Krishna*
18-R-04- Insertion Compounds of Anthracenes with Zeosils, *R. Jäger and P. Behrens*
18-R-05- Influence of the K Content of a 4(-Zeolite in N_2/CH_4 Separation, *D. Richardeau, G. Joly, P. Magnoux,, S. Mignard, M. Guisnet and M. Thomas*
18-R-06- Natural Adsorbents Characterization by Gas Chromatography, *G. Aguilar A. and A.M. Maubert F.*
18-R-07- Modified Mesoporous Silicates and Aluminosilicates for the Adsorption and Decomposition of Toxic Gases, *M. Naderi, J.L. Pickett, M.J. Chinn and D.R. Brown*
18-R-08- Mesoporous Molecular Sieves as Adsorbents for Bioseparations, *J.M. Kisler, G.W. Stevens and A.J. O'Connor*

20 Zeolite membranes and films

20-R-01- A Novel Method for Evaluating the Adhesion Strength of Zeolite Membrane, *Y. Zhang, G. Lu, Zh. Xu and Q. Chen*
20-R-02- Hydrothermal Synthesis of Mordenite Membrane on Porous Substrate, *Y. Zhang, Zh. Xu and Q. Chen*
20-R-03- Low Dielectric Constant Mesoporous Silica Films Prepared from TEOS, *B.-Z. Wan, C.-Y. Ting and D.-F. Oyan*
20-R-04- Modified ZSM-5 Zeolite Membranes: Preparation and Separation Performance, *V.A. Tuan, C.J. Gump, R.D. Noble and J.L. Falconer*
20-R-05- The Formation Process of Pore Channels Oriented Perpendicular to the Surfaces of Mesoporous Films, *Y. Sasaki, Y. Ichihara and T. Nishikawa*
20-R-06- Mixed Surfactant Route for the Preparation of Mesoporous Silica Films, *N. Petkov, S. Mintova and T. Bein*
20-R-07- Tubular MFI Membranes by In-Situ Nucleation and Secondary Growth, *C. Algieri, G. Golemme, S. Kallus and J.D.F. Ramsay*
20-R-08- Preparation and Charaterization of Lamellar Mesostructured Semiconductor-Organic Films, *N. Bai, H. Chen, J. Chen, Y. Liu, Y. Guo, D. Li and W. Pang*
20-R-09- Na-A Crystallisation on Porous alpha-Al_2O_3, *H. Richter and I. Voigt*
20-R-10- Microwave Synthesis of Zeolite and Zeo-like Composite Membranes: MFI/(-Al_2O_3 and Sodalite/(-Al_2O_3, *A. Julbe, F. Cazevieille, G. Volle and C. Guizard*

21 Nanocomposite fundamentals and applications

21-R-01- Adsorption of Heavy Metal Anions on Protonated Diamino-Functionalized Mesoporous Silica, *T. Yokoi, T. Tatsumi and H. Yoshitake*

22 Advanced materials

22-R-01- Redox Behaviour of SnO_2 Nanoparticles Encapsulated in the Pores

of Zeolite NaY and Mesoporous Si-MCM-41 towards Reductive Gas Atmospheres, *Y. Altindag, M. Warnken, N.I. Jaeger and M. Wark*
22-R-02- ENSCM: Enantiomeric New Self-Constructed Materials, *C. Bied, J.J.E. Moreau, L. Vellutini and M. Wong Chi Man*
22-R-03- Antiblocking Agent for Polyolefin Films Derived from Zeolite Pc, *S. Kojima, H. Nakata and K. Sugano*
22-R-04- "Tectons" as Zeolite-like Supramolecular Materials, *C. Trolliet, O. Saied, J.D. Wuest and A. Tuel*
22-R-05- Sorption Material for Dosimetry of Radionuclides in Environmental Water, *A.Yu. Andryushchenko, A.B. Blank and N.I. Shevtsov*
22-R-06- Incorporation of Aluminium into Mesoporous Silica Films, *J. Mori, T. Yoshikawa, K. Kuroda and M., Ogawa*
22-R-07- Hybrid Inorganic-Organic Frameworks Exhibiting Exceptionally High Thermal Stability, *P.M. Forster and A.K. Cheetham*
22-R-08- Organic-Containing Mesoporous Material with a Periodic Pore-Wall Structure, *S. Inagaki, S. Guan, T. Ohsuna and O. Terasaki*
22-R-09- Covalently Bound Porphyrin-based Complexes in Mesoporous Films, *J.M. Reder and T. Bein*
22-R-10- High Surface Area SiC via Carbothermal Reduction of a Carbon-Containing Mesoporous MCM-48 Silica Phase, *J. Parmentier, J. Patarin, J. Dentzer and C. Vix-Guterl*
22-R-11- Synthesis of Bare, Nanoscale, Metallic Wires from within a Zeolite Crystal, M.J. Edmondson, *W Zhou, S.A. Sieber, I.P. Jones, I. Gameson, P.A. Anderson and P.P. Edwards*
22-R-12- Preparation and Application of MCM-22 Ship-in-a-bottle Complexes, *G. Gbery, A. Zsigmond, P. Pantano, and K.J. Balkus, Jr.*
22-R-13- The Application of DAM-1 Molecular Sieves in Optical Sensors, *D. Coutinho, C. Meek, P. Pantano and K.J. Balkus, Jr.*

23 Micro and mesoporous materials in fine chemistry

23-R-01- Epoxidation of (,(-Unsaturated Ketone over Various Titano-Silicates, *M. Sasidharan, P. Wu and T. Tatsumi*
23-R-02- Vapor-Phase Conversion of Citronellal over Zeolites and MCM-41 Mesoporous Molecular Sieves, *D.-L. Shieh and A.-N. Ko*
23-R-03- Liquid-Phase Beckmann Rearrangement Catalyzed by Large-Pore Zeolites and Mesoporous Molecular Sieves, *Chawalit N., Wu P. and T. Tatsumi*
23-R-04- Photoepoxidation of Propene with Molecular Oxygen on Ti-Zeolites and their Modifications by a Metal Ion-Implantation, *H. Yamashita, K. Ikeue, K. Kida, Y. Kanazawa and M. Anpo*
23-R-05- Mo/Zr Mixed Oxide Supported on MCM-41 as Strong Acid Catalysts, *T. Li and S. Cheng*
23-R-06- Selective Cleavage of Hydroxyl Protecting Groups Using Mesoporous Silicas, *A. Itoh, T. Kodama and Y. Masaki*

24 New routes to hydrocarbon activation

24-R-01- Complex Study of Zeolites in Methyl Benzenes Isomerisation, *L.A. Surina and S.A. Surina*
24-R-02- Catalase and Monooxygenase Activity of Ferropro-toporphyrin Immobilized on Zeolite NaM, *T.M. Nagiev, S.Z. Zulfugarova, M.T. Abbasova, Ch.A. Mustafaeva and A.A. Abbasov*
24-R-03- Catalytic Synergy Effect of Mo-V Incorporated SBA-1 Cubic Mesoporous Materials in the Selective Oxidation of Methane, *L.-X. Dai, K. Tabata, E. Suzuki and T. Tatsumi*
24-R-04- Photoinduced non Oxidative Coupling of Methane over H-Zeolite around Room Temperature, *Y. Kato, H. Yoshida, A. Satsuma and T. Hattori*
24-R-05- Influence of Heteropoly Acid and USY-Zeolite on Activity of Nickel Catalysts Containing Ni,H-ZSM-5 Zeolite, *J.R. Grzechowiak and A. Masalska*

25 Conversion of aromatics

25-R-01- Catalytic Performance of Mesoporous MCM-41 during Cracking Reaction, *W.H. Chen, Q. Zhao, H.P. Lin, Y.S. Yang, C.Y. Mou and S. B. Liu*
25-R-02- Influence of the Properties of Zeolite Beta on its para-Selectivity in the Nitration of Toluene and Nitrotoluene, *S. Bernasconi, G. Pirngruber, A. Kogelbauer and R. Prins*
25-R-03- Explanation by Molecular Modelling of the Particular Behavior of the H-MCM-22 Zeolite during m-Xylene Transformation, *S. Laforge, S. Rui, D. Martin and M. Guisnet*
25-R-04- Hydroconversion of1-Methylnaphthalene over Pt/USY and Pt-Pd/USY Catalysts in the Presence, or not, of Thiophene, *C. Petitto, G. Giordano, F. Fajula and C. Moreau*
25-R-05- Calorimetric Study of the Friedel-Crafts Acetylation of Anisole by Acetic Anhydride on Zeolite HBEA, *H. Lachas, E.G. Derouane and D. Bethell*

26 Catalysis for oil refining

26-R-01- Effect of Zeolite Additives on the Formation of Al-Ni-Mo Catalysts in Oil Fractions Hydroprocesses, *R.R. Aliev, S.A. Surin and L.A. Surina*
26-R-02- MCM-41 as a Support for CoMoS HDS Catalysts, *P.J. Kooyman, P. Waller, A.D. van, Langeveld, C. Song, K.M. Reddy and J.A.R. van Veen*
26-R-03- Catalytic and Selective Activity of MEAPO-36 in the Hydrocracking of Gaz Oil, *C.W. Ingram and K.Y. Ghebreyessus*
26-R-04- The Conversion of Methanol to Hydrocarbons over Dealuminated Zeolite H-BETA, *M. Björgen and S. Kolboe*
26-R-05- Basic Cs-modified Pt/MCM-41: Activity in n-Hexane Conversion, *J. Blanchard, M. Breysse, K. Fajerwerg, P. Massiani, M.F. Ribeiro and J. M. Silva*
26-R-06- Conversion of Methanol and 1-Butene over an H-ZSM-5 Zeolite Catalyst: Use of Isotopic Labeling for Mechanistic Studies, *S. Svelle and S. Kolboe*

27 Selective oxidation and sulfur resistance

27-R-01- Oxidation with H_2O_2 over Ti-Containing Molecular Sieves - A New Method for Removing Organo Sulfur Compounds from Fuels, *V. Hulea, F. Fajula and J. Bousquet*
27-R-02- Fe-ZSM-5 Catalysts for the N_2O Oxidation of Benzene to Phenol, *R. Monaci, E. Rombi, D. Meloni, V. Solinas, I. Rossetti and L. Forni*
27-R-03- Photooxidation of1-Butene on Cationic Forms of Zeolites, *S.L.Y. Tang, D.J. McGarvey and V.L. Zholobenko*
27-R-04- Oxidation of Alcohols Catalyzed by Chromium-Exchanged Zeolite Y, *A. Arafat*

28 Confinement and physical chemistry for catalysis

28-R-01- Paramagnetic Silver Clusters in Sodalites, *J. Michalik, H. Yamada and J. Perlinska*
28-R-02- Stabilization of Pt Nanoparticules in Basic Pt/CsBeta: a XAS Study, *P. Massiani, F. Villain, M.F. Ribeiro, J.M. Silva and M. Taibi*
28-R-03- The Reactivity of Molecules Trapped within the SAPO-34 Cavities in the Methanol-to-Hydrocarbons Reaction, *B. Arstad and S. Kolboe*
28-R-04- Selective Synthesis of Hexamethylbenzene from Methanol over Zeolite Catalysts, *G. Torosyan, H. Sargsyan, Kh. Harutiunyan, St. Babayan and V. Marukhyan*

29 New approaches to catalyst preparation

29-R-01- Metallic Nanoparticles from Heterometallic Co-Ru Carbonyl Clusters in MCM-41, *F. Schweyer, P. Braunstein, C. Estournès, J. Guille, H. Kessler, J.-L. Paillaud and J. Rosé*
29-R-02- Overgrowth of Mesoporous Molecular Sieve Material over ZSM-5, *F.A. Twaiq and S. Bhatia*
29-R-03- Sulfated Mesoporous Zirconia, an Active Catalyst for n-Butane Isomerization, *X. Yang and F. Jentoft*

30 Environmental catalysis

30-R-01- Bleaching of Aqueous Methylene Blue with Mesoporous Silicates Synthesized by Rapid Room Temperature Method, *I. Moriguchi, M. Honda and Y. Teraoka*
30-R-02- Effect of Copper Precursors in Solid-State Ion Exchange with MFI Zeolite on NO Decomposition Activity, *H. Furukawa, I. Moriguchi and Y. Teraoka*
30-R-03- Metal Exchanged Zeolites as a New Desulfurization Agent in Pipeline Natural Gas at Room Temperature, *S. Satokawa and Y. Kobayashi*
30-R-04- Methane Reforming by Carbon Dioxide on Nickel Exchanged ZSM-5, USY and Mordenite Zeolites, *D. Halliche, O. Cherifi and A. Auroux*
30-R-05- Selective Reduction of NO to Nitrogen in the Presence of Oxygen, *T. Furusawa, K. Seshan, S.E. Maisuls, J.A. Lercher, L. Lefferts and K. Aika*
30-R-06- Oxidative Photoreaction with FSM-16, *A. Itoh, T. Kodama, S. Inagaki and Y. Masaki*
30-R-07- Redox Properties of Silver Incorporated Zeolites, *A.J. Evans, R.M. Ormerod and V.L. Zholobenko*
30-R-08- Synthesis, Characterization and Photocatalytic Property of Ti-Beta Zeolite, *S.-H. Chien, C.-H. Lu, H.C. Chiu and Y-S. Yang*
30-R-09- Catalytic Decomposition of N_2O over Steamed Fe/ZSM-5, *Z. Qingjun, B.L. Mojet, J. van Grondelle and R.A. van Santen*
30-R-10- High-Field ESR Spectroscopy of Cu(I)-NO Complexes in Cu-ZSM-5, *A. Pöppl and M. Hartmann*

31 Environment-friendly applications of zeolites

31-R-01- (K, Na, NH4)-ZSM-5- Adsorption of Harmful Substances, *A. Cizmek*
31-R-02- Use of the Khongurin-Zeolite in the Prolongation of Disinfectants, *N.P. Tarabukina and M.P. Neustroev*
31-R-03- Application of Natural Zeolite (Khongurin) in the Breeding and for Cattle Fattening in Yakutia, *N.S. Permyakov*
31-R-04- Use of Natural Zeolites in Medical and Biological Studies, *R.I. Aizman, A.D. Gerasev, S.N. Lukanina, G.A. Svyatash, L.E. Panin and T.I. Ryabichenko*
31-R-05- Zeolites as Adsorbents for VOC Monitoring, *S.S. Rayalu, A. Shrivastava, S.U. Meshram, P. Kumar, N.K. Labhsetwar and M.Z. Hasan*
31-R-06- Removal of Lead and Nickel Ions Using Synthetic Zeolite 4A, *B. Cekova, V. Jankovski and I. Temelkov*
31-R-07- Fly-Ash Waste Conversion to Zeolitic Industrial Sorbent Commodity Having Environmental Uses, *J.M. McKernan and K.H. Mathers*
31-R-08- Ammonia Removal from Gases by Adsorption on Clinoptilolite, *M. Kocirik, K. Ciahotny, L. Melenova, O. Pachtova and H. Jirglova*
31-R-09- Zeolite Modification and Application Study for Decontamination of Nuclear Liquid Waste (part 1), *R. Gevorkyan, H. Yeritsyan, Y. Keheyan, H. Sarkisyan, A. Sahakyan and A. Hovhannisyan*
31-R-10- Zeolite Modification and Application Study for Decontamination of Nuclear Liquid Waste (part 2), *R. Gevorkyan, H. Yeritsyan, Y. Keheyan, H. Sarkisyan, C. Presutti, G. Christidis, D. Moraetis, N. Kekelidze and L. Akhalbeldashvili*
31-R-11- Integrated Utilization of Natural Zeolites in Wastewater Depuration and Agriculture: Application of an Italian Chabazite-Rich Tuff, *E. Marchi and E. Passaglia*

32 Zeolite minerals and health

32-R-01- Influence of the Countercations (Na^+, K^+, Ca^{2+} and Mg^{2+}) of a Natural Zeolite-Clinoptilolite on the Removal of Ammonium, T. *Filipan, L. Curkovic, A. Farkas and A. Pisarovic*
32-R-02- New Applications of Zeolites in the Life Sciences: Tobacco Smoke, *W.M. Meier*

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編集後記

最近「ナノテクノロジー」というキーワードがマスコミに取り上げられる機会が増えてきました。ゼオライトを研究に対象にしている我々はもちろん，化学に携わる方々の多くは，「ナノ」より小さい原子・分子をいじっているという自負をお持ちと思います。そのため，「何を今さら」と考えられている方も少なくないのではないでしょうか？もっとも，「ナノ」が話題になると，研究のチャンスがいろいろと増えてくるので，口にこそあまり出しませんが（！）

そこで少々，何故「ナノテク」なのかを私なりに考えてみました。細孔の世界でも，マクロポアは，粒子を固めたり，いろいろな方法で隙間をつくることで，作られています。一方ゼオライトのミクロポアの世界は，分子，イオンが組み合わさり，孔が形成されています。しかしながら，非シリカ系ではもう少し大きなものが存在するものの，シリカ系では，最大でも14員環で1次元のUTD-1とCIT-5しか合成されてきていません。歴史的な順番は逆になりますが，分子集合体をもちいたメソポーラスシリカ の合成が如何に画期的であったかを改めて思い知らされます。メソポーラスシリカに より，メソとミクロの間はかなり繋がってきています。更にメソポアの大きい方へは，共重合体を用いたアプローチが開拓されました。遠からず，メソとマクロの間も 繋がるのではないでしょうか？

以上を思いめぐらせた結果，「ナノテクノロジーの開花により，原子・分子レベルからのBuild-upとバルクからのBreak-downの接点がいよいよ埋まることで，あらゆる階層の構造制御が可能になり，その結果として，より高度な機能の利用が可能になる」という結論を導いたのですが，読者諸氏のご意見はいかがでしょうか。

(T. O.)

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