Heterogeneous catalyzed aryl–nitrogen bond formations using a valine derivative bridged metal–organic coordination polymer†

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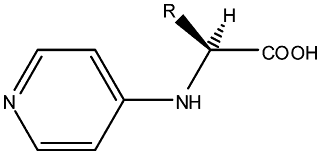
A new 1D macrocyclic copper(II) coordination polymer, [Cu**L**$_2$] (**1**, **L** = *N*-(4-pyridyl)-D,L-valine), based on a valine derived ligand was synthesized and characterized by single-crystal X-ray diffraction studies. The catalytic experiments showed that compound **1** was an efficient catalyst for the cross-coupling reaction of arylboronic acids with imidazole in the absence of additional additives. The crystal structure analysis of **1** suggested that the vacant axial coordination position of the copper(II) atoms played a very important role in the efficient catalytic transformation process.

Introduction

In the past few years, there has been a great increase in the interest and generation of functional metal–organic frameworks (MOFs), owing to their various topological architectures and applicable properties in diverse fields such as catalysis, gas storage, separation and ion-exchange, *etc.*[1–6] Since amino acid residues provide functional properties and highly selective substrate-binding abilities in many extended biological structures, the preparation of bio-analogous materials comprising amino acid residues as building blocks has attracted a great deal of attention.[7,8] There is much current interest in the crystal engineering of coordination networks by combining amino acids and metal nodes with suitable geometries, which can help develop new technologically useful materials for a variety of applications.[9]

We are particularly interested in the synthesis of coordination solids based on multitopic linkers derived from amino acids and the exploration their catalytic properties in heterogeneous processes. According to the coordination preferences of transition metal atoms, we have modified the amino acids with pyridyl groups to generate a series of effective bridging linkers (Scheme 1). Basically, the carboxylic acid and pyridyl groups within each ligand can link up metal nodes for the construction of extended solid-state framework structures, while the remaining unit within the ligand can be used for attachment of functional groups. Herein we wish to report the synthesis, crystal structure and highly efficient catalytic properties of a new 1D coordination polymer, [Cu**L**$_2$] (**1**, **L** = *N*-(4-pyridyl)-D,L-valine).



Scheme 1

Experimental details

Materials and methods

The *N*-(4-pyridyl)-D,L-valine (**L**) ligand was synthesized according to the literature method,[10] while all the other chemicals were purchased and used without further purification. The IR spectrum was recorded using KBr pellets on a FTS-40 spectrophotometer. Powder X-ray diffraction data (PXRD) were recorded on a RIGAKU D/MAX 2550/PC using Cu Kα (λ = 1.5406 Å) radiation. Thermogravimetric analysis (TGA) was carried out in air on a NETZSCH STA 409 PC/PG instrument at a heating rate of 4 °C min^{-1}. ^{1}H NMR spectra were recorded on a 400 MHz spectrometer in $CDCl_3$ solution and the chemical shifts were reported relative to the internal standard TMS (0 ppm).

Synthesis of [CuL$_2$] (1). Heating a mixture of $Cu(NO_3)_2\cdot 3H_2O$ (12.1 mg, 0.05 mmol) and **L** (9.7 mg, 0.05 mmol) in a mixed solvent of EtOH (3 mL), H_2O (1.5 mL) and DMF (0.1 mL) in a capped vial at 90 °C afforded purple crystals after five days, which were filtered, washed with EtOH and Et_2O, and dried at room temperature (yield: 52% based on **L**). Anal. calcd for $C_{20}H_{26}CuN_4O_4$ (%): H, 5.83; C, 53.44; N, 12.47. Found: H, 5.76; C, 52.96; N, 12.51. IR (KBr pellet, ν/cm^{-1}): 1622 s, 1534 s, 1468 w, 1411 w, 1400 s, 1344 m, 1299 m, 1252 m, 1209 s, 1162 m, 1091 m, 1065 m, 1024 s, 819 s, 747 m, 665 w, 597 w, 530 m.

Representative procedure for the Ar–N coupling of arylboronic acids with imidazole using solid catalyst 1. A mixture of 0.12 mmol phenylboronic acid, 0.1 mmol of imidazole and 0.01 mmol of solid **1** in 2 mL MeOH was vigorously stirred in air at room temperature for 12 h. The reaction mixture was filtered, washed, evaporated and extracted with CH_2Cl_2. The organic phase was combined, dried over anhydrous $MgSO_4$, filtered and evaporated. Subsequently, the organic phase was separated and passed through a short silica gel column to remove the surplus phenylboronic acid (hexane : ethyl acetate, 50 : 50 volume ratio). An aliquot was analyzed by gas chromatography (GC) to give the conversion value, calculated by using a pure sample as standard. ^{1}H NMR (500 MHz, $CDCl_3$): δ 7.84 (s, 1H), 7.43–7.47 (m, 2H), 7.32–7.37 (m, 3H), 7.26 (s, 1H), 7.19 (s, 1H) ppm; MS (ESI) *m*/*z*: 145.1 ([*M* + H]$^+$).

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X-Ray data collection and structure determination

Data collection and the determination of the unit cell for compound **1** were performed on a Rigaku R-AXIS RAPID. The data were collected using graphite-monochromated Mo Kα radiation ($\lambda = 0.71073$ Å) at 293 K. Empirical absorption corrections were applied using the **ABSCOR** program.[11] The structure was solved by direct methods and refined using a full-matrix least-squares method with the **SHELXTL-97** program package.[12] All non-hydrogen atoms were located successfully from Fourier maps and were refined anisotropically. The positional disorder in compound **1** was evidenced by the presence of two positions for each copper atom despite collecting the crystal data several times. We have tried using low symmetry space groups, such as monoclinic space groups *Cc* and *C*2, and triclinic space groups *P*1 and $P\bar{1}$, to attempt to solve the positional disorder problem. However, the problem remained. Thus, we attempted to solve the crystal structure by refining each copper atom as occupying two disordered positions. Initially, the occupancies were refined using free variables, but in the final cycles of refinement, these occupancies were held as fixed values near the refined values of 75% for Cu(1) and 25% for Cu(2). Since copper atoms are sited on a two-fold axis, the occupancy for Cu(1) should be 0.375, while Cu(2) is 0.125. Although the final results ($R1 = 0.0907$, w$R2 = 0.1294$) are slightly large, the molecular structure is well behaved. The poor crystal data suggested that only the gross connectivity of the structure was obtained. Hence, we will not discuss the structural characteristics in detail. CCDC 695224.† The data collection parameters, crystallographic data and final agreement factors are collected in Table 1.

Results and discussion

Purple crystals of **1** were synthesized by the reaction of $Cu(NO_3)_2 \cdot 3H_2O$ with the **L** ligand, in a mixed solvent of H_2O, EtOH and DMF, at 90 °C for five days. Single-crystal X-ray diffraction analysis had revealed that compound **1** crystallizes in the monoclinic space group *C*2/*c*. There is half a Cu(II) atom and one **L** ligand in the asymmetric unit (Fig. 1). Each copper(II) atom is tetra-coordinated to two carboxyl oxygen atoms (Cu(1)–O = 1.961(7) Å) and two pyridyl groups (Cu(1)–N = 1.948(7) Å) of four **L** ligands in a distorted square-planar geometry, with axial positions left uncoordinated. Thus, two **L** ligands are bidentate, *trans*-linking two Cu(II) centers to form an 18-membered macrocyclic ring 8.517 Å in diameter that lies in the *ac* plane, which is further extended into a one-dimensional polymeric chain running along the (101) direction (Fig. 2).

Table 1 Crystal data and structural refinements for compound **1**

Compound	[Cu**L**$_2$]
Formula	$C_{20}H_{26}CuN_4O_4$
Formula weight	449.99
Crystal color	Purple
Crystal system	Monoclinic
Space group	*C*2/*c*
Unit cell dimensions	$a = 15.833(3)$ Å $b = 9.330(2)$ Å $c = 15.152(3)$ Å $\beta = 114.57(3)°$
Volume/Å^3	2035.4(7)
Z	4
Calculated density/g cm^{-3}	1.468
F(000)	940
Absorption coefficient/mm^{-1}	1.107
θ for data collection/°	3.13 to 23.00
Reflections collected	5910 ($R_{int} = 0.1518$)
Data/parameters	1344/138
Goodness-of-fit on F^2	1.062
$R1$ (w$R2$) [$I > 2\sigma(I)$]	0.0907 (0.1294)
Largest diff. peak and hole/e Å^{-3}	0.297 and −0.254

$R1 = \sum(|F_o| - |F_c|)/\sum|F_o|$, w$R2 = [\sum w(F_o^2 - F_c^2)^2/\sum w(F_o^2)^2]^{0.5}$.

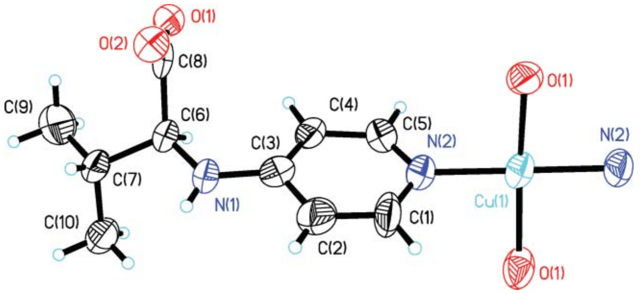


Fig. 1 ORTEP representation of the symmetry expanded local structure for **1** (30% probability ellipsoids).

It is worth noting that there is no significant supermolecular interaction other than van der Waals contacts between the 1D macrocycle chains (Fig. 3). By careful inspection of the crystal structure, we found that there are only weak hydrogen bonds between the parallel polymeric chains, which arise from the amino groups and deprotonated carboxyl oxygen atoms (N–O = 2.918 Å).

The TGA graph showed that there was no weight loss until 160 °C for compound **1** (Fig. 4). Above 160 °C, the product began to lose coordinated **L** ligands, resulting in decomposition, which

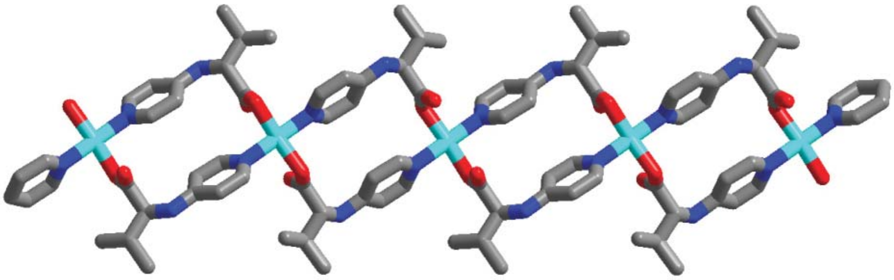


Fig. 2 The 1D polymeric macrocyclic chain in **1** (cyan, red, blue and deep grey colors represent Cu, O, N and C atoms, respectively).

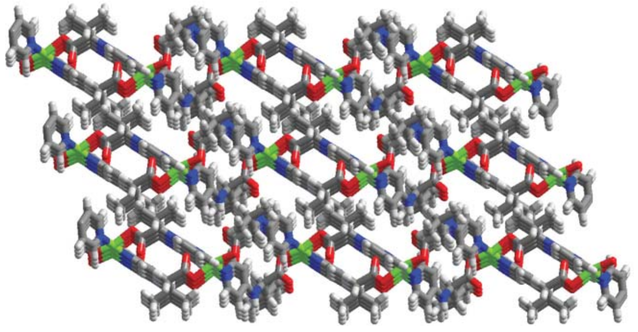


Fig. 3 A view of the packing diagram of **1** (green, red, blue, deep grey and light grey colors represent Cu, O, N, C and H atoms, respectively).

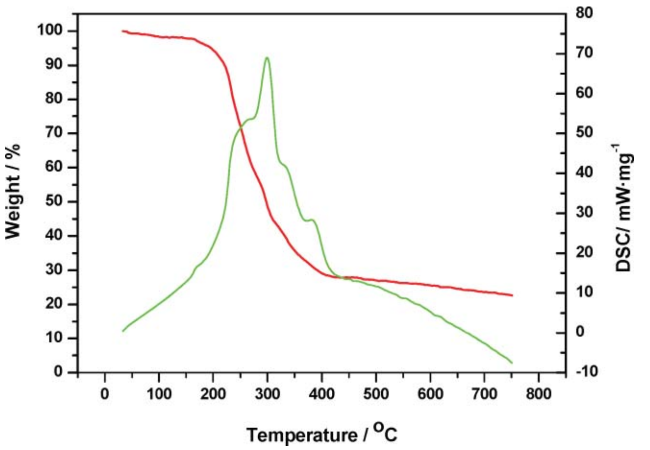


Fig. 4 TGA plot of **1**.

was consistent with the X-ray crystal structural analysis result. The PXRD of a bulk sample of **1** showed a sharp diffraction pattern similar to that of the simulated, which confirmed the purity of the sample (Fig. 5). The PXRD spectrum of a sample of **1** that had been ground and heated at 120 °C for 5 h in air showed a sharp diffraction pattern similar to that of the unheated sample. This result indicates that the neutral framework of **1** was maintained and stable up to 120 °C, indicating it can be used for further catalytic application studies.

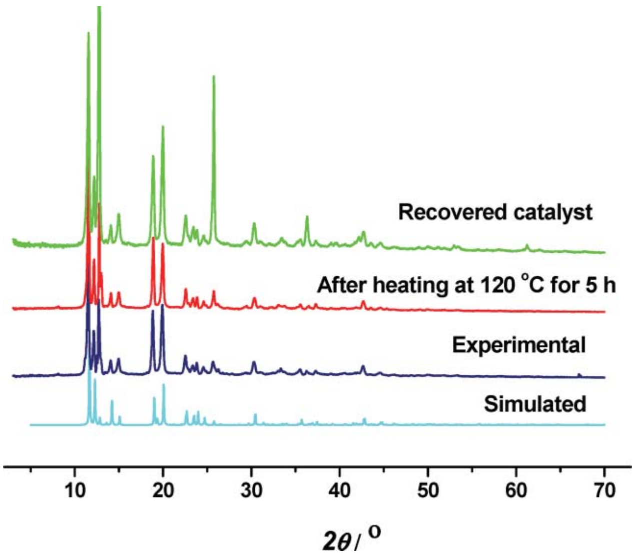
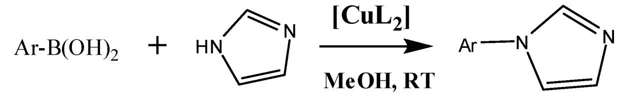


Fig. 5 Powder X-ray diffraction patterns for compound **1**.

Since the copper(II) atom is in a distorted square-planar coordination geometry and the axial positions are vacant in **1**, we speculated that the copper atoms can coordinate to suitable organic substrates, utilizing the axial coordination sites to generate catalytic intermediates. These interesting structural features promoted us to study the catalytic activities of compound **1**.

The development of efficient methods for the synthesis of *N*-arylimidazoles under mild conditions has recently gained considerable attention in synthetic chemistry because of the compounds' biological activities.[13] And, as it is well established that, in homogeneous and heterogeneous systems, Cu(II) complexes are active catalysts for a range of cross-coupling reactions of arylboronic acids with organic amines,[14–17] we attempted to employ our complex in the cross-coupling reaction of arylboronic acids with imidazole to verify our hypothesis. The treatment in air of **1** with phenylboronic acid and imidazole in methanol for 12 h generated the anticipated *N*-phenylimidazole in very high yield (Table 2, entry 1). Subsequently, a number of structurally and electronically diverse substrates were employed for the catalytic coupling reaction (Table 2, entries 2 to 6). When *p*-tolylboronic acid was used, the corresponding *N*-arylation product was obtained in good yield (Table 2, entry 2). Upon treatment of *p*-chlorophenylboronic acid with imidazole, a chlorinated *N*-phenylimidazole was formed in 94% yield (Table 2, entry 4). However, the cross-coupling yield was lower for *o*-cyanophenylboronic acid (Table 2, entry 6). When the cross-coupling reaction time was prolonged, the conversion was slightly improved (Table 2, entry 3). The catalyst can be separated by simple filtration from the reaction mixture, which was then reused in the successive run. The slightly decreased yield (85%) may be due to the small amount of catalyst lost through filtration. PXRD for the recovered material showed that the structural integrity of the catalyst was maintained during the catalytic reaction (Fig. 5). The average particle size of recovered solid **1** was 45 nm in diameter, deduced from Scherrer's formula using the (11$\bar{4}$) diffraction peak. We can not observe trace cross-coupling product from a mixture of phenylboronic acid and imidazole in the presence of the filtrate of **1** in methanol under

Table 2 The cross-coupling of arylboronic acids with imidazole and compound **1** catalyst[a]



Entry	Ar	Time/h	Yield (%)[b]
1	Ph	12	97
2	4-CH_3–Ph	12	85
3	4-CH_3–Ph	24	94
4	4-Cl–Ph	12	94
5	3-CH_3–Ph	12	84
6	4-CN–Ph	12	55

[a] Arylboronic acid (0.12 mmol), imidazole (0.1 mmol) and solid **1** (0.01 mmol) were stirred in MeOH (2 mL) at room temperature. [b] Yields (%) were determined by GC on a *SE-54* column.

otherwise identical conditions. These experiments demonstrated the heterogeneous nature of the present catalytic system. However, as there is no channel or cavity in solid **1**, the catalytic reaction might take place on the solid surface. Most interestingly, the surface promoted catalytic reaction can make the transformation process highly efficient without any additional additives.

There have been many reports on attempts to optimize the copper-promoted *N*-arylation of arylboronic acids with imidazole to furnish *N*-arylimidazoles.[15–17] Comparing with these reports, the catalytic activity of **1** used at room temperature was better than that of homogenous and heterogeneous catalysts used under similar reaction conditions, and it even rivals them when used at evaluated reaction temperatures. The striking catalytic activity of **1** prompted us to investigate further its catalytic properties. We tried different solvents, such as DMF, THF, CH_2Cl_2 and H_2O. However, almost no product can be observed when using any of these, except for trace amounts of the desired product obtained when using DMF. Obviously, it can be concluded that the solvent effect was one of the most important factors in the coupling of arylboronic acids with imidazole in the case of compound.**1** Together with the crystal structure analysis result, we believe that the distinct catalytic activity of **1** can be attributed to the coordinatively vacant axial positions of copper atoms, which generated dramatically different results compared to the aforementioned systems.

Collman *et al.* have proposed that the coupling reaction mechanism to involve binuclear Cu(II) catalyzed C–N bond formation.[15] However, our structurally characterized catalyst suggests a mononuclear process because all of the copper(II) atoms are fixed by the coordinating ligands during the catalytic process.

Conclusions

In conclusion, we have successfully synthesized a new bridged copper(II) coordination polymer with valine derived ligands, which acted as an efficient catalyst for the cross-coupling of arylboronic acids with imidazole in methanol. The attractive features of the catalytic system are: (1) no need for additional additives, (2) can be used at room temperature, (3) affords good to excellent yields, (4) reusability of the catalyst. The crystal structure analysis, together with the catalytic results, suggests that the coordinatively vacant axial positions in the copper(II) atoms plays a very important role in the efficient catalytic activity of **1**. Most significantly of all, the structurally characterized catalyst is very important for further study of the catalytic mechanism.

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References

1 (*a*) S. Lee, A. B. Mallik, Z. Xu, E. B. Lobkovsky and L. Tran, *Acc. Chem. Res.*, 2005, **38**, 251; (*b*) K. S. Suslick, P. Bhyrappa, J.-H. Chou, M. E. Kosal, S. Nakagaki, D. W. Smithenry and S. R. Wilson, *Acc. Chem. Res.*, 2005, **38**, 283; (*c*) P. Feng, X. Bu and N. Zheng, *Acc. Chem. Res.*, 2005, **38**, 293; (*d*) B. Kesanli and W. Lin, *Coord. Chem. Rev.*, 2003, **246**, 305; (*e*) G. Férey, C. Mellot-Draznieks, C. Serre and F. Millange, *Acc. Chem. Res.*, 2005, **38**, 217; (*f*) C. N. R. Rao, S. Natarajan and R. Vaidhyanathan, *Angew. Chem., Int. Ed.*, 2004, **43**, 1466; (*g*) I. Goldberg, *Chem. Commun.*, 2005, 1243; (*h*) J. L. C. Rowsell and O. M. Yaghi, *Angew. Chem., Int. Ed.*, 2005, **44**, 4670; (*i*) R. J. Hill, D.-L. Long, N. R. Champness, P. Hubberstey and M. Schröder, *Acc. Chem. Res.*, 2005, **38**, 335; (*j*) N. W. Ockwig, O. Delgado-Friedrichs, M. O'Keeffe and O. M. Yaghi, *Acc. Chem. Res.*, 2005, **38**, 176; (*k*) D. Bradshaw, J. B. Claridge, E. J. Cussen, T. J. Prior and M. J. Rosseinsky, *Acc. Chem. Res.*, 2005, **38**, 273; (*l*) Special issue on "Reticular Chemistry: Design, Synthesis, Properties and Applications of Metal–Organic Polyhedra and Frameworks", beginning with: T. D. Hamilton, G. S. Papaefstathiou and L. R. MacGillivray, *J. Solid State Chem.*, 2005, **178**, 2409.

2 (*a*) S. L. James, *Chem. Soc. Rev.*, 2003, **32**, 276; (*b*) C. Janiak, *Dalton Trans.*, 2003, 2781; (*c*) M. Oh, G. B. Carpenter and D. A. Sweigart, *Acc. Chem. Res.*, 2004, **37**, 1; (*d*) S. Kitagawa, R. Kitaura and S.-i. Noro, *Angew. Chem., Int. Ed.*, 2004, **43**, 2334.

3 (*a*) J. S. Seo, D. Wand, H. Lee, S. I. Jun, J. Oh, Y. Jeon and K. Kim, *Nature*, 2000, **404**, 982; (*b*) L. Pan, H. Liu, X. Lei, X. Huang, D. H. Olson, N. J. Turro and J. Li, *Angew. Chem., Int. Ed.*, 2003, **42**, 542; (*c*) C.-D. Wu, A. Hu, L. Zhang and W. Lin, *J. Am. Chem. Soc.*, 2005, **127**, 8940; (*d*) S.-H. Cho, B. Ma, S. B. T. Nguyen, J. T. Hupp and T. E. Albrecht-Schmitt, *Chem. Commun.*, 2006, 2563; (*e*) C.-D. Wu and W. Lin, *Angew. Chem., Int. Ed.*, 2007, **46**, 1075.

4 (*a*) K. Seki and W. Mori, *J. Phys. Chem. B*, 2002, **106**, 1380; (*b*) N. L. Rosi, J. Eckert, M. Eddaoudi, D. T. Vodak, J. Kim, M. O'Keeffe and O. M. Yaghi, *Science*, 2003, **300**, 1127; (*c*) G. Férey, M. Latroche, C. Serre, F. Millange, T. Loiseau and A. Percheron-Guégan, *Chem. Commun.*, 2003, 2976; (*d*) B. Kesanli, Y. Cui, M. Smith, E. Bittner, B. Bockrath and W. Lin, *Angew. Chem., Int. Ed.*, 2005, **44**, 72.

5 (*a*) K. Uemura, S. Kitagawa, M. Kondo, K. Fukui, R. Kitaura, H.-C. Chang and T. Mizutani, *Chem.–Eur. J.*, 2002, **8**, 3586; (*b*) M. P. Suh, J. W. Ko and H. J. Choi, *J. Am. Chem. Soc.*, 2002, **124**, 10976; (*c*) D. Bradshaw, T. J. Prior, E. J. Cussen, J. B. Claridge and M. J. Rosseinsky, *J. Am. Chem. Soc.*, 2004, **126**, 6106.

6 (*a*) B. F. Hoskins and R. Robson, *J. Am. Chem. Soc.*, 1990, **112**, 1546; (*b*) K. S. Min and M. P. Suh, *J. Am. Chem. Soc.*, 2000, **122**, 6834.

7 (*a*) Z. Jiang, Z. Liang, X. Wu and Y. Lu, *Chem. Commun.*, 2006, 2801; (*b*) M. Klussmann, H. Iwamura, S. P. Mathew, D. H. Wells Jr., U. Pandya, A. Armstrong and D. G. Blackmond, *Nature*, 2006, **441**, 621; (*c*) B. List, *Synlett*, 2001, 1675.

8 (*a*) B. List, *Acc. Chem. Res.*, 2004, **37**, 548; (*b*) W. Notz, F. Tanaka and C. F. Barbas, III, *Acc. Chem. Res.*, 2004, **37**, 580.

9 Some recent examples: (*a*) E. V. Anokhina and A. J. Jacobson, *J. Am. Chem. Soc.*, 2004, **126**, 3044; (*b*) J. J. Vittal, X. Wang and J. D. Ranford, *Inorg. Chem.*, 2003, **42**, 3390; (*c*) N. Guillou, C. Livage, M. Drillon and G. Férey, *Angew. Chem., Int. Ed.*, 2003, **42**, 5314; (*d*) R. Matsuda, R. Kitaura, S. Kitagawa, Y. Kubota, R. V. Belosludov, T. C. Kobayashi, H. Sakamoto, T. Chiba, M. Takata, Y. Kawazoe and Y. Mita, *Nature*, 2005, **436**, 238; (*e*) R. Vaidhyanathan, D. Bradshaw, J.-N. Rebilly, J. P. Barrio, J. A. Gould, N. G. Berry and M. J. Rosseinsky, *Angew. Chem., Int. Ed.*, 2006, **45**, 6495; (*f*) Y. Zhang, M. K. Saha and I. Bernal, *CrystEngComm*, 2003, **5**, 34; (*g*) B. Screenivasulu and J. J. Vittal, *Angew. Chem., Int. Ed.*, 2004, **43**, 5769; (*h*) M. A. Alam, M. Nethaji and M. Ray, *Angew. Chem., Int. Ed.*, 2003, **42**, 1940.

10 Z.-Q. Yan, Y.-Y. Zhou and L. Nie, *Chin. J. Appl. Chem.*, 2005, **22**, 1258.

11 T. Higashi, *ABSCOR*, Rigaku Corporation, Tokyo, Japan, 1995.

12 G. M. Sheldrick, *Program for Structure Refinement*, University of Göttingen, Germany, 1997.

13 (*a*) S. V. Ley and A. W. Thomas, *Angew. Chem., Int. Ed.*, 2003, **42**, 5400; (*b*) M. Negwer, *Organic-chemical drugs and their synonyms: (an international survey)*, Akademie Verlag GmbH, Berlin, 7th edn, 1994; (*c*) P. Y. S. Lam, G. Vincent, C. G. Clark, S. Deudon and P. K. Jadhav, *Tetrahedron Lett.*, 2001, **42**, 3415.

14 (*a*) P. Y. S. Lam, S. Deudon, K. M. Averill, R. Li, M. Y. He, P. Deshong and C. G. Clark, *J. Am. Chem. Soc.*, 2000, **122**, 7600; (*b*) G. I. Elliott and J. P. Konopelski, *Org. Lett.*, 2000, **2**, 3055; (*c*) P. Y. S. Lam, C. G. Clark, S. Saubern, J. Adams, K. M. Averill, D. M. T. Chan and A. Combs, *Synlett*, 2000, 674; (*d*) K. Mori, K. Yamaguchi, T. Mizugaki, K. Ebitani and K. Kaneda, *New J. Chem.*, 2002, **26**, 1536; (*e*) K. Mori, K. Yamaguchi, T. Hara, T. Mizugaki, K. Ebitani and K. Kaneda, *J. Am.*

Chem. Soc., 2002, **124**, 11572; (*f*) B. M. Choudary, C. Sridhar, M. L. Kantam, G. T. Venkanna and B. Sreedhar, *J. Am. Chem. Soc.*, 2005, **127**, 9948.

15 (*a*) J. P. Collman and M. Zhong, *Org. Lett.*, 2000, **2**, 1233; (*b*) J. P. Collman, M. Zhong, C. Zhang and S. Costanzo, *J. Org. Chem.*, 2001, **66**, 7892.

16 (*a*) J. P. Collman, M. Zhong, L. Zeng and S. Costanzo, *J. Org. Chem.*, 2001, **66**, 1528; (*b*) S. S. van Berkel, A. van den Hoogenband, J. W. Terpstra, M. Tromp, P. W. N. M. van Leeuwen and G. P. F. van Strijdonck, *Tetrahedron Lett.*, 2004, **45**, 7659; (*c*) J.-B. Lan, L. Chen, X.-Q. Yu, J.-S. You and R.-G. Xie, *Chem. Commun.*, 2004, 188; (*d*) M. L. Kantam, B. Neelima, Ch. V. Reddy and V. Neeraja, *J. Mol. Catal. A: Chem.*, 2006, **249**, 201.

17 (*a*) M. L. Kantam, G. T. Venkanna, C. Sridhar, B. Sreedhar and B. M. Choudary, *J. Org. Chem.*, 2006, **71**, 9522; (*b*) L.-Y. Zhang and L. Wang, *Chin. J. Chem.*, 2006, **24**, 1605.