JOC Note

Transesterification of Various Methyl Esters Under Mild Conditions Catalyzed by Tetranuclear Zinc Cluster

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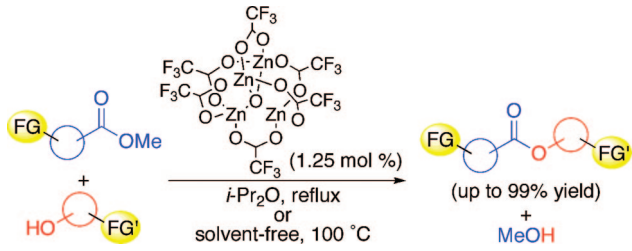

A new catalytic transesterification promoted by a tetranuclear zinc cluster was developed. The mild reaction conditions enabled the reactions of various functionalized substrates to proceed in good to high yield. A large-scale reaction under solvent-free conditions proceeded with a low *E*-factor value (0.66), indicating the high environmental and economical advantage of the present catalysis.

Esterification is one of the most general and important reactions in organic synthesis because of the ubiquity of esters in various biologically active natural products and drugs.[1] Common synthetic routes to esters include condensation reactions of carboxylic acids with alcohols and reactions with highly reactive acylating reagents such as acyl halides and acid anhydrides. These methods require stoichiometric amounts of the condensation reagent or base, resulting in the formation of greater than stoichiometric amounts of unwanted chemical waste.[2–4] In terms of atom-economy and environmental concerns, the catalytic transesterification of esters, especially methyl and ethyl esters, with higher and/or functionalized alcohols is desirable for the synthesis of diverse esters. Moreover, the transesterification of esters is more advantageous than the condensation reaction of carboxylic acids with alcohols due to handling ease and high stability of esters as well as their high solubility in most organic solvents.[5] Because the transesterification is an equilibrium reaction, it is difficult to attain high conversions. The following methods have been used to force the reaction toward the product side: (i) use of excess amounts of either of the reactants,[6] (ii) use of an enol ester as a reactant, accompanied by the formation of the corresponding aldehyde or ketone,[7] and (iii) removal of the resulting lower alcohol by molecular sieves[8] or continuous distillation. The last approach is the most ideal method, and several catalytic transesterifications at high temperature using esters of lower alcohols were developed using this approach.[3,9] Moreover, Otera et al. recently reported transesterification at room temperature promoted by a fluorous distannoxane catalyst in a fluorous biphase system.[9m] There is great demand, however, for the development of a versatile transesterification under mild and harmless conditions to produce highly functionalized compounds such as pharmaceutical agents and so on.

Recently, we developed a direct conversion of carboxylic acids, esters, and lactones with β-amino alcohols to oxazolines catalyzed by a μ-oxo-tetranuclear zinc cluster $Zn_4(OCOCF_3)_6O$ (**1**) (Figure 1),[10a] in which zinc ions act cooperatively, similar to aminopeptidase[11] and efficient multimetallic catalysts.[12]

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(9) (a) Seebach, D.; Hungerbuehler, E.; Naef, R.; Schnurrenberger, P.; Weidmann, B.; Zueger, M. *Synthesis* **1982**, 138. (b) Schultheiss-Reimann, P.; Kuntz, H. *Angew. Chem., Int. Ed. Engl.* **1983**, *22*, 63. (c) Otera, J.; Yano, T.; Kawabata, A.; Nozaki, H. *Tetrahedron Lett.* **1986**, *27*, 2383. (d) Waldmann, H.; Kunz, H. *J. Org. Chem.* **1988**, *53*, 4172. (e) Klibanov, A. M. *Acc. Chem. Res.* **1990**, *23*, 114. (f) Gutman, A. L.; Shkolnik, E.; Shapira, M. *Tetrahedron* **1992**, *48*, 8775. (g) Blandy, C.; Pellegatta, J.-L.; Cassoux, P. *Catal. Lett.* **1997**, *43*, 139. (h) Ponde, D. E.; Deshpande, V. H.; Bulbule, V. J.; Sudalai, A.; Gajare, A. S. *J. Org. Chem.* **1998**, *63*, 1058. (i) Krasik, P. *Tetrahedron Lett.* **1998**, *39*, 4223. (j) Xiang, J.; Toyoshima, S.; Orita, A.; Otera, J. *Angew. Chem., Int. Ed.* **2001**, *40*, 3670. (k) Xiang, J.; Orita, A.; Otera, J. *Adv. Synth. Catal.* **2002**, *344*, 84. (l) Chavan, S. P.; Shivasankar, K.; Sivappa, R.; Kale, R. *Tetrahedron Lett.* **2002**, *43*, 8583. (m) Otera, J. *Acc. Chem. Res.* **2004**, *37*, 288. (n) Yoo, D.-W.; Han, J.-H.; Nam, S. H.; Kim, H. J.; Kim, C.; Lee, J.-K. *Inorg. Chem. Commun.* **2006**, *9*, 654.

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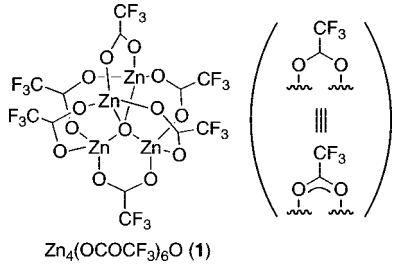
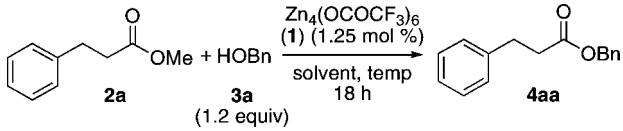


$Zn_4(OCOCF_3)_6O$ (**1**)

FIGURE 1. Structure of the μ-oxo-tetranuclear zinc cluster $Zn_4(OCOCF_3)_6O$ (**1**).

TABLE 1. Solvent Effects in Transesterifications Catalyzed by 1[a]



2a **3a** (1.2 equiv) **4aa**

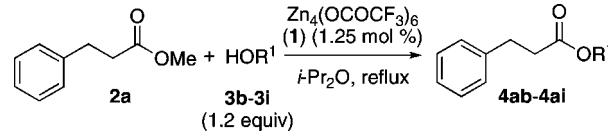
entry	solvent	temperature	yield (%)[b]
1	toluene	reflux (111 °C)[c]	95
2	toluene	80 °C	48
3	PhCl	80 °C	41
4	octane	80 °C	61
5	$ClCH_2CH_2Cl$	80 °C	52
6	$CHCl_3$	reflux (61 °C)[c]	14
7	CPME[d]	80 °C	38
8	*i*-Pr_2O	reflux (68 °C)[c]	79
9	THF	reflux (66 °C)[c]	20
10	CH_3CN	80 °C	16
11	DMF	80 °C	22
12	DMSO	80 °C	34

[a] Reaction conditions: A mixture of **2a** (1.0 mmol), **3a** (1.2 mmol), **1** (1.25 mol % = 5 mol % Zn), and solvent (1.7 mL) were heated for 18 h under an argon atmosphere (flowing Ar gas, 10 mL/min). [b] GC yield. [c] Boiling point of pure solvent. [d] Cyclopentyl methyl ether.

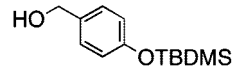
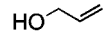
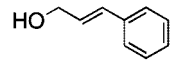
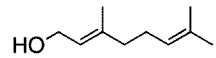
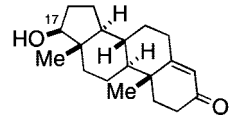
Furthermore, this catalysis was successfully applied to the *O*-selective acylation of amino alcohols.[10b] Herein, we report that the tetranuclear zinc cluster **1** catalyzes the transesterification of various methyl esters under mild conditions (almost neutral conditions, reaction temperature: 58−68 °C). The applicability of this catalysis was successfully demonstrated by reactions of various esters **2** and alcohols **3** containing functional groups and protecting groups sensitive to acid.

We initiated our studies of a transesterification using methyl 3-phenylpropionate (**2a**) and 1.2 equiv of benzyl alcohol (**3a**) as representative substrates. Under toluene reflux conditions, 1.25 mol % of the Zn cluster **1** efficiently promoted the reaction to afford the product **4aa** in 95% yield (Table 1, entry 1). When the reaction was performed at 80 °C in toluene, however, the yield decreased to 48% (entry 2). We thus examined solvent effects at 80 °C or lower. Chlorobenzene, octane, and halogenated solvents gave low to moderate yields (entries 3−6).

TABLE 2. $Zn_4(OCOCF_3)_6O$ (1) Catalyzed Transesterification of Methyl 3-Phenylpropionate (2a) with Various Alcohols 3[a]



2a **3b-3i** (1.2 equiv) **4ab-4ai**

entry	alcohol (HOR^1)		time (h)	yield (%)[b]
1	$HO(CH_2)_3CH_3$	**3b**	18	92
2	$HO(CH_2)_5CH_3$	**3c**	24	94
3	$HO(CH_2)_{17}CH_3$	**3d**	18	99
4		**3e**	24	92
5		**3f**	40	76
6		**3g**	40	91
7		**3h**	40	91
8		**3i**	40	96

[a] 3.0 mmol scale (flowing Ar gas, ~2.0 mL/min). [b] Isolated yield.

Diisopropyl ether was the best solvent (entry 8, 76% yield). Diisopropyl ether forms a roughly 1:1 azeotrope with methanol, which boils at ca. 10 °C lower than pure diisopropyl ether (bp = 68 °C) at atmospheric pressure,[13] making it easy to remove MeOH from the reaction mixture. Coordinating solvents, such as THF, acetonitrile, DMSO, and DMF were not effective for this reaction, probably because the zinc active site is occupied by solvent molecules (entries 9−12). Because this reaction is an equilibrium process, the conversion of the reaction is dependent on catalyst activity as well as the amount of methanol that is expelled from the system. We achieved high reproducibility by controlling the flow rate (10 mL/min) of Ar gas (flowing on top of the condenser).[14] Although the flow rate of the Ar gas affected the reaction rate, it had only a small effect on the final yield of the product **4**. Thus, the following experiments were performed at the Ar flow rate of ~2.0 mL/min.

Under the optimized conditions, we first investigated the scope and limitations of alcohols **3** (Table 2). The transesterification of **2a** with a variety of primary aliphatic alcohols **3b**−**3h**, including benzylic alcohols and allylic alcohols, was efficiently catalyzed by the Zn cluster **1** to afford the corresponding esters **4** in high yield (entries 1−7, up to 99% yield). Since the reaction conditions are almost neutral, the acid-sensitive TBDMS ether of phenol persisted (entry 4), and neither the isomerization nor cyclization of geraniol (**3h**) occurred (entry 7).[9m] Secondary alcohol **3i** was also applicable to the present system, in which there was no epimerization of the C17 stereocenter and the enone moiety survived to give the desired ester **4ai** in 96% yield (entry 8; see also entries 4 and 5 in Table 4). In contrast, transesterification with tertiary alcohols did not proceed due to steric hindrance. Acidic alcohols such as phenol

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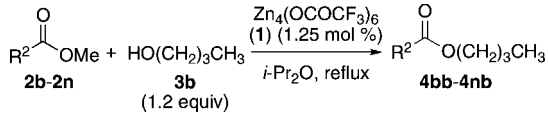
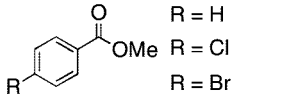
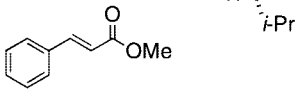
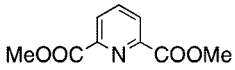
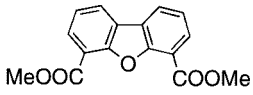
(13) Gmehling, J.; Menke, J.; Krafczyk, J.; Fischer, K. *Azeotropic Data*; Wiley-VCH: Weinheim, 2004.

(14) See Supporting Information for details.

TABLE 3. $Zn_4(OCOCF_3)_6O$ (1)-Catalyzed Transesterification of Various Methyl Esters 2 with *n*-Butanol (3b)[a]



entry	ester (R^2CO_2Me)		time (h)	yield (%)[b]
1	 R = H	**2b**	40	96
2	R = Cl	**2c**	24	91
3	R = Br	**2d**	40	90
4	R = CN	**2e**	24	77
5	$R = NO_2$	**2f**	40	>99
6	R = OH	**2g**	40	76
7	R = 	**2h**	44	93
8		**2i**	24	86
9	$CH_3(CH_2)_{16}COOMe$	**2j**	24	92
10	$THPO(CH_2)_9COOMe$	**2k**	44	87
11	$C_6H_{11}COOMe$	**2l**	40	97
12[c]		**2m**	40	>99
13[c]		**2n**	40	97

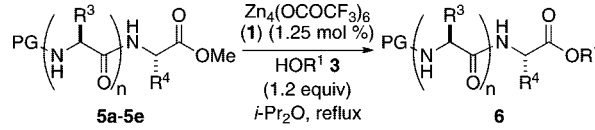
[a] 3.0 mmol scale (flowing Ar gas, ~2.0 mL/min). [b] Isolated yield. [c] 7.2 mmol (2.4 mol equiv) of *n*-butanol was used.

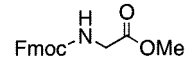
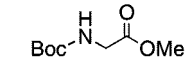
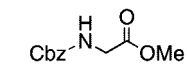
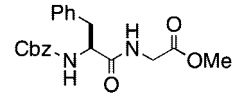
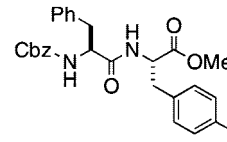
(pK_a 9.95) and 1,1,1,3,3,3-hexafluoro-2-propanol (pK_a 9.3) did not participate in the reaction. Taking advantage of this chemoselectivity, aliphatic alcohols can be selectively acylated in the presence of acidic alcohols (*vide infra*).

Next, we estimated the substrate generality of the ester component (Table 3). Aromatic esters with various substituents at the para position were converted to the corresponding butyl esters **4** in good to excellent yield (entries 1−7). Previously, we reported that nitrile is a good substrate for the Zn cluster-catalyzed oxazoline formation. Moreover, oxazoline reacts with alcohol in the presence of Lewis acid. Under the current reaction conditions, however, neither functional group reacted with the alcohol (entries 4 and 7). Due to the large reactivity difference between aliphatic and aromatic alcohols, the reaction of 4-hydroxybenzoate **2g** provided the corresponding product **4gb** in good yield without forming a phenol ester (entry 6). When methyl cinnamate (**2i**) was used as a substrate, 1,4-addition did not proceed and the desired product **4ib** was obtained in 86% yield (entry 8). Various aliphatic esters with higher reactivities can also be used (entries 9−11) without the loss of the highly acid-sensitive tetrahydropyranyl ether functional group (entry 10). Dimethyl esters were also converted to the corresponding dibutyl esters in excellent yield, accompanied by only trace amounts of monobutyl esters (entries 12 and 13).

We then applied this catalysis to the reaction of amino esters (Table 4). Amino esters are widely present in natural and unnatural bioactive compounds and play an important role in the development of pharmaceuticals.[15] The scope and limitation of the protecting groups, which are frequently used for amino group protection, were investigated. The reaction of *N*-Fmoc, *N*-Boc, and *N*-Cbz glycine methyl esters **5** with benzyl alcohol (**3a**) afforded the corresponding benzyl esters **6** exclusively without the loss of any protecting groups (entries 1−3). Transesterification with secondary alcohols including (+)-menthol **3k**gave the corresponding product in excellent yield (entries 4 and 5). Reactions of dipeptides also proceeded in good yield, and there was no epimerization of the stereocenters (entries 6 and 7).

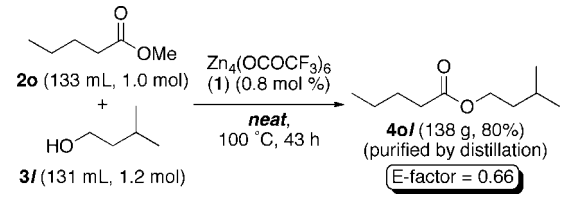
TABLE 4. $Zn_4(OCOCF_3)_6O$ (1) Catalyzed Transesterification of Various *N*-Protected Amino Esters 5 with Benzyl Alcohol (3a)[a]



entry	amino ester		alcohol	time (h)	yield (%)[b]
1		**5a**	**3a**	41	94
2		**5b**	**3a**	42	87
3		**5c**	**3a**	18	83
4		**5c**	**3j**	18	99
5		**5c**	**3k**	48	92
6		**5d**	**3a**	18	92
7		**5e**	**3a**	20	82

[a] 3.0 mmol scale (flowing Ar gas, ~2.0 mL/min). [b] Isolated yield.

SCHEME 1. Transesterification under Solvent-Free Conditions



The applicability of **1** to a large-scale synthesis was demonstrated by synthesizing isopentyl pentanoate (**4o*l***), which is well-known to have an apple flavor, under solvent-free conditions (Scheme 1). In the presence of 0.8 mol % of the Zn cluster **1**, the reaction of 133 mL (1.0 mol) of methyl pentanoate (**2o**) and 131 mL (1.2 mol) of isopentanol (**3*l***) at 100 °C under solvent-free conditions was completed within 43 h, and the resulting mixture was directly purified by distillation to give the pure product **4o*l*** in 80% yield. The *E*-factor value[16] (an

(15) (a) Han, H.-K.; Oh, D.-M.; Amidon, G. L. *Pharm. Res.* **1998**, *15*, 1382. (b) Yang, C.; Gao, H.; Mitra, A. K. *J. Pharm. Sci.* **2001**, *90*, 617. (c) Vig, B. S.; Lorenzi, P. J.; Mittal, S.; Landowski, C. P.; Shin, H.-C.; Mosberg, H. I.; Hilfinger, J. M.; Amidon, G. L. *Pharm. Res.* **2003**, *20*, 1381.

(16) (a) Sheldon, R. A. *Chem. Ind. (London)* **1992**, 903. (b) Sheldon, R. A. *Chem. Ind. (London)* **1997**, 12. (c) Sheldon, R. A. *Pure Appl. Chem.* **2000**, *72*, 1233. (d) Poliakoff, M.; Fitzpatrick, J. M.; Farren, T. R.; Anastas, P. T. *Science* **2002**, *297*, 807.

assessment of the waste generation and environmental impact of chemical manufacturing processes) of this reaction was only 0.66, indicating the high environmental and economical advantage of the present catalytic transesterification.

In summary, a general and highly efficient transesterificaton of various methyl esters was developed using the tetranuclear zinc cluster **1**. The current Zn catalysis has several appealing features, including high tolerance of functional groups, low catalyst toxicity, and ease of operation. Moreover, conducting the reactions under solvent-free conditions results in high volumetric productivity and low waste generation. Further studies on chemoselective acylation of highly functionalized natural products are ongoing.

Experimental Section

General Procedure for Transesterification of Methyl Esters with Alcohols as Illustrated by the Synthesis of 4-*tert*-Butyldimethylsiloxybenzyl 3-Phenylpropionate (4ae). A mixture of $Zn_4(OCOCF_3)_6O$ (**1**) (36 mg, 0.038 mmol), methyl 3-phenylpropionate (**2a**) (470 μL, 3.0 mmol), 4-*tert*-butyldimethylsiloxybenzyl alcohol (**3e**) (858 mg, 3.6 mmol), and diisopropyl ether (5.0 mL) was refluxed for periodic time under an argon atmosphere. The resulting mixture was concentrated and purified by silica gel column chromatography (silica gel, hexane/EtOAc = 40/1) to provide the title compound **4ae** (1017 mg, 92%) as a colorless oil; IR (neat NaCl, ν/cm^{-1}) 2956, 2930, 2859, 1736, 1609, 1513, 1257, 1159, 913, 839, 782, 734, 699; ^{1}H NMR (300 MHz, $CDCl_3$, 35 °C) δ 0.19 (s, 6H, Si*Me*$_2$), 0.98 (s, 9H, Si*t**Bu*), 2.65 (t, J = 7.8 Hz, 2H, PhC*H*$_2$CH$_2$), 2.95 (t, J = 7.8 Hz, 2H, PhCH$_2$C*H*$_2$), 5.02 (s, 2H, OC*H*$_2$Ar), 6.79 (d, J = 8.7 Hz, 2H, *Ar*), 7.1−7.3 (m, 7H, *Ph, Ar*); ^{13}C NMR (75 MHz, $CDCl_3$, 35 °C) δ −4.3, 18.3, 25.8, 31.1, 36.0, 66.1, 120.0, 126.1, 128.2, 128.4, 128.6, 129.8, 140.4, 155.6, 172.5; MS (EI) *m*/*z* (relative intensity) 370 ([M$^+$], 38), 313 (40), 280 (18), 221 (34), 207 (100); HRMS (EI) *m*/*z* calcd for $C_{22}H_{30}O_3Si$ 370.1964, found 370.1953.

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Supporting Information Available: Experimental procedures and characterization of the products. This material is available free of charge via the Internet at http://pubs.acs.org.

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