Journal of Catalysis **194,** 491 (2000)
doi:10.1006/jcat.2000.2960, available online at http://www.idealibrary.com on IDEAL®

LETTER TO THE EDITOR

In the recent article by Ueda and co-workers (Y. H. Han, W. Ueda, and Y. Moro-Oka, *J. Catal.* **186**, 75 (1999)), the authors report results in the selective oxidation of propylene using a mixed metal oxide catalyst described as $a(Bi_2Mo_3O_{12}/CeO_2)/Na_{0.5-3x}La_{0.5+x}MoO_4$, where a is up to 0.0025 and $x = 0$, 0.03 or 0.08. They attributed the observed enhancement in catalytic activity and selectivity for selective oxidation of propylene to acrolein with the inclusion of bismuth, molybdenum, and cerium to the existence of "supported" phases of $Bi_2Mo_3O_{12}$ and CeO_2 on the $Na_{0.5-3x}La_{0.5+x}MoO_4$ "support." The authors invoke their X-ray diffraction (XRD) and X-ray photoelectron spectroscopic (XPS) data to conclude that separate bismuth molybdate and cerium oxide phases exist. However, these data do not uniquely support this conclusion. Instead, the reported results are more justifiably explained by the likely formation of a surface solid solution phase of the type already described extensively in the literature (J. F. Brazdil L. C. Glaeser, and R. K. Grasselli, *J. Catal.* **81**, 142 (1983)); (J. F. Brazdil and R. K. Grasselli, *J. Catal.* **79**, 104 (1983)) as providing superior activity and selectivity for oxidation (ammoxidation) of propylene.

Specifically, the authors point to the XRD evidence that no change in unit cell parameters was observed with the inclusion of bismuth, molybdenum, and cerium in the sodium lanthanum molybdate scheelite phase. This evidence is, however, specious. At the levels used, it would not be possible to detect solid solution formation by the bulk analytical measurement of a standard XRD experiment. Using the authors' lattice parameter measurements one can estimate that at the 0.005 level of bismuth, molybdenum, and cerium, formation of a scheelite solid solution would result in a change in unit cell volume of only about 0.2 $Å^3$ out of 334.9 $Å^3$, well within the error of their measurement. Thus, failure to observe a change in lattice parameters does not indicate that a solid solution is not present ("Absence of evidence is not evidence of absence"). Likewise, the authors' finding by XPS that the surface of "supported" catalysts is enriched in bismuth and cerium is not evidence of the existence of separate bismuth molybdate or cerium oxide phases. It can equally indicate the existence of a scheelite solid solution enriched at the surface in the bismuth and cerium additives. This would not be surprising given that these cations were impregnated onto the sodium lanthanum molybdate from solution, then calcined 200°C below the calcination temperature of the sodium lanthanum molybdate. At 450°C, the likelihood of solid solution formation for scheelite molybdate compounds cannot be discounted, based on previously published work.

It would have been useful if the authors had used the same approach they employed in their earlier similar study to gain direct evidence of any purported surface bismuth molybdate and cerium oxide phases. In that work (*Catal. Lett.* **12**, 35 (1992)), the authors used transmission electron microscopy (TEM) to identify a $Bi_2Mo_3O_{12}$ phase on the surface of a $Co_{11/12}Fe_{1/12}MoO_x$ phase. Similarly applying TEM in the present work could identify any extant surface phases.

Thus, lacking any direct evidence of separate surface bismuth molybdate and cerium oxide phases, the catalytic results of Ueda and co-workers are equally likely to be due to the formation of a bismuth cerium molybdate containing sodium lanthanum molybdate scheelite solid solution, localized at the surface of the catalyst particle, and exhibiting the characteristic catalytic functions for selective olefin oxidation described previously (J. F. Brazdil and R. K. Grasselli, *J. Catal.* **79**, 104 (1983)).

James F. Brazdil
Raymond G. Teller

BP Amoco Chemicals
Naperville, Illinois 60563-8460

Received October 20, 1999; accepted February 11, 2000

0021-9517/00 $35.00