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A Response to Vogel and Marcelin

In the foregoing Letter to the Editors (*1*), Vogel and Marcelin report that pure aluminum phosphate can be prepared from aqueous media only at a pH of less than 4.0. At a high pH, a mixed precipitate of alumina–aluminum phosphate forms.

The aluminum phosphate catalyst we used for cis-2-butene isomerization was prepared at a high pH; the pH increased during preparative procedure and finally became 7.0 (*2*). Following pretreatment at 750°C in a vacuum, the catalyst had a surface area of 163 m^2/g and showed an amorphous X-ray pattern. The possibility that our catalyst was alumina–aluminum phosphate cannot be excluded.

From the viewpoint of catalytic behavior, however, our catalyst exhibited the feature in *cis*-2-butene isomerization which is unique to aluminum phosphate catalyst; only geometrical isomerization to *trans*-2-butene occurs.

We prepared several aluminum phosphate catalysts by different preparative methods, and examined their catalytic properties for *cis*-2-butene isomerization. The results are summarized in Table 1. The catalysts I, II, and V were amorphous, while the catalysts III and IV were crystallized. Regardless of the preparative methods and the crystal structures, all aluminum phosphate catalysts exhibited extremely high *trans*/1 ratios in *cis*-2-butene isomerization as far as the catalysts were pretreated at about 750°C. The high *trans*/1 ratio can be regarded as the characteristic feature which distinguishes aluminum phosphate catalyst from other catalysts. Alumina catalyst shows a *trans*/1 ratio of about 1. If a small amount of alumina was included in an aluminum phosphate catalyst and acted in *cis*-2-butene isomerization, the *trans*/1 ratio would be lowered considerably. This was not observed for any aluminum phosphate catalysts. Therefore, it is considered that the observed reaction in our previous paper was totally contributed from aluminum phosphate and not from alumina even if the catalyst was alumina–aluminum phosphate.

TABLE 1

Physical and Catalytic Properties of Aluminum Phosphate Catalysts Prepared by Different Methods

$AlPO_4$ catalyst	Preparation method	Reference for preparation	Pretreatment temperature[a] (°C)	Crystal structure	Surface area	Ratio of *trans*/1[b]
I[c]	$Al(NO_3)_3 + H_3PO_4 + NH_4OH$	(*3*)	750	Amorphous	163	∞
II	$Al(NO_3)_3 + H_3PO_4$ + urea	(*3*)	700	Amorphous	95	∞
III	$\gamma\text{-}Al_2O_3 + H_3PO_4$	(*4*)	750	Cristobalite	22	∞
IV	$\gamma\text{-}Al_2O_3 + H_3PO_4$	(*3*)	750	Tridymite Cristobalite Berlinite	101	55
V	$AlCl_3 + H_3PO_4 + C_2H_4O$	(*5*)	750	Amorphous	193	∞

[a] All catalysts were calcined at 300°C in air before pretreatment in a vacuum.

[b] Ratio of *trans*-2-butene to 1-butene produced in *cis*-2-butene isomerization at 25°C.

[c] The catalyst was used in our previous paper (*2*).

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In general, use of well-defined or well-crystallized materials in determination of catalytic properties is of great advantage. In some cases, however, well-crystallized catalysts possess only a small number of catalytically active sites, or exhibit different catalytic properties from amorphous catalysts. It is also important to reveal the catalytic properties of highly active catalysts even though their crystal structures are amorphous. Ideally, use of both well-crystallized catalysts and highly active catalysts and comparison of their catalytic properties are desirable.

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