The Crystal and Molecular Structure of the Catalytically Active Organoaluminium Compound [$Me_2AlOCPhNPh,MeCHO,AlMe_3$]

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Summary The molecular structure of [$Me_2AlOCPhNPh,MeCHO,AlMe_3$] has been determined by *X*-ray structure analysis.

RECENTLY we reported the molecular structure of a stereospecific polymerization catalyst for acetaldehyde [$(Me_2AlOCPhNPh)_2$] (I)[1] and its monomer complex [$(Me_2AlOCPhNPh,MeCHO)_2$] (II).[2] We now report the molecular structure of [$Me_2AlOCPhNPh,MeCHO,AlMe_3$] which is obtained by mixing equimolar amounts of trimethylaluminium and (II) in n-hexane at 0°. Polymerization of acetaldehyde using this complex as a catalyst gives quite different results from those using (I) or (II); *atactic* polymer is obtained.

Crystal data: [$Me_2AlOCPhNPh,MeCHO,AlMe_3$], $a = 13{\cdot}60$, $b = 13{\cdot}74$, $c = 12{\cdot}40$ Å, $\beta = 95{\cdot}87°$; space group $P2_1/n$, $Z = 4$, D_c 1·06, D_m 1·07 g cm^{-3}, $\mu = 13{\cdot}0$ cm^{-1} (for Cu-K_α).

As the complex decomposes immediately in air, the crystal used was sealed in a thin-walled glass capillary tube under argon.

The intensity data were collected by the multi-film equi-inclination Weissenberg method, and measured visually. 2891 independent reflexions were obtained from the *0kl* to *9kl* layers. The structure was established by the heavy-atom method, and was refined by the block-diagonal least-squares procedure (isotropic temperature factors for hydrogen atoms and anisotropic temperature factors for non-hydrogen atoms) to an *R* index of 0·13 for non-zero reflexions.

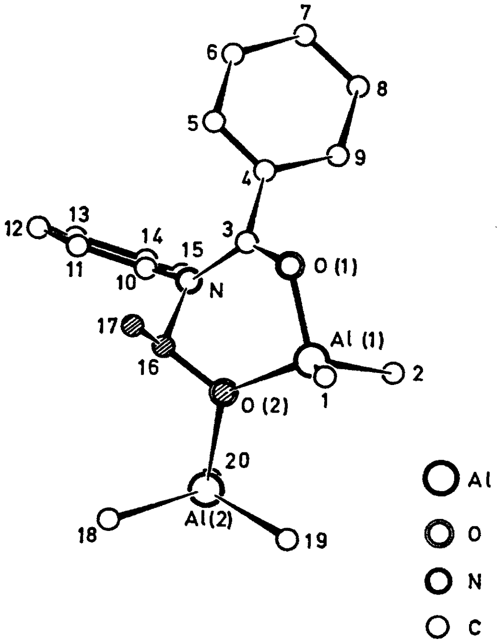

FIGURE. *Molecular structure of* [Me$_2$AlOCPhNPh,MeCHO,AlMe$_3$]. *Atoms belonging to the aldehyde moiety are hatched.*

The molecular structure is shown in the Figure. The molecule is *not* dimeric as were (I) and (II). No bridged-methyl group is found in the molecule. The shortest contact between methyl groups and aluminium atoms in the molecule is Al(1) ······ C(19) [3·397(14) Å]; the rest are all greater than 4·0 Å.

The AlMe$_3$ group is connected to one half of the molecule (II) by co-ordination between Al(2) and the aldehyde oxygen O(2). A similar type of co-ordination was reported in the *X*-ray analysis of [dioxan,(AlMe$_3$)$_2$].[3]

The geometry around Al(2) shows slight distortion from the tetrahedral conformation; the average value of the O(2)– Al(2)–C angles 104·0(0·4)° is smaller, while that of the C–Al(2)–C angles [114·3 (0·5)°] is larger, than the normal tetrahedral angle. The Al(1) atom, however, shows considerable distortion from tetrahedral co-ordination; the angle C(1)–Al(1)–C(2) is 122·2(5)° and O(1)–Al(1)–O(2) is 94·0(3)°. The Al(1)–O(1) distance [1·837(6) Å] is much shortened as compared with that [2·030(7) Å] in (II), while Al(1)–O(2) [1·869(6) Å] is equal to the corresponding distance [1·856(7) Å] in (II). The Al(2)–O(2) distance [1·928(6) Å] is shorter than the corresponding distance [2·02(2) Å] in [dioxan(AlMe$_3$)$_2$] but a little longer than those of Al(1)–O(1) and Al(1)–O(2) in the latter molecule.

The oxygen atom in the acetaldehyde moiety, O(2), co-ordinates to both aluminium atoms and shows the typical sp^2-type geometry, which was also found in (II). The four atoms Al(1), Al(2), O(2), and C(16) are coplanar within 0·002 Å.

The most noteworthy feature in the acetaldehyde moiety [O(2), C(16), and C(17)] is that the O(2)–C(16) distance [1·352(10) Å] is an intermediate value between single- and double-bond distances, while in (II) this distance [1·432(12) Å] is a normal single-bond distance.

In the amide moiety, interatomic distances and angles and the conformation of the two benzene rings correspond closely to those in (I) and (II).

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