The structure of □-Al2O3, a crucial catalyst support, remains debated, particularly the Al³□ cation and vacancy locations. While models propose spinel and non-spinel structures, NMR and DFT data support a predominantly spinel structure (62.5-65% octahedral Al³□), contradicting non-spinel models. TEM reveals significant □-Al2O3 surface reconstruction, with (111) facets favored over atomically flat (110) surfaces. The non-spinel model requires reevaluation.