



Synthesis and Properties of Oxygen Complex Compounds of 3d-Transition Metals

Djumaeva Maxfuza Kayumovna ¹

¹ Degree-seeking student, assistant of Medical Chemistry Chair, Bukhara State Medical Institute, Uzbekistan, Bukhara

Abstract: The fact of significant activation of molecular oxygen and butene as a result of their coordination by transition metal compounds in the process of complex formation has been established. A combined catalyst is proposed that makes it possible to carry out the reaction of butene oxidation to methyl ethyl ketone under mild conditions (at low temperature, atmospheric pressure) with a high yield and selectivity of the target product.

Keywords: complex compound, molecular oxygen, methyl ethyl ketone, oxidation.

In modern applied chemistry, the specific activation of simple molecules is often of great interest, many of which can be used as ligands. When such molecules form complexes, they become so active that they can actually carry out important processes. Complex compounds of transition metals with organic ligands that bind molecular oxygen have very important properties. Complex compounds of this type are promising from the point of view of the possibility of their use as oxidizing agents in the oxidation of organic substances.

In living nature, the role of such oxygen complexes with transition metal ions in their composition is played by enzymes. From this point of view, it is important to search for new catalytic systems that are models of enzymes capable of carrying out substrate oxidation reactions and forming stable oxygen complexes with transition metals under mild conditions. There is little information on the reactivity of oxygen-containing transition metal complexes. At the same time, the available data indicate that the coordinating molecular oxygen in many compounds is in an activated state [1, 2].

Interest in the activation of coordinating molecular oxygen is also due to the potential formation of short-lived intermediates in homogeneously catalyzed autoxidation reactions. Of particular importance for elucidating the mechanism of the reaction is the study of the reactivity of O₂ in oxygenated complexes. In the well-studied Wacker process using an aqueous Pd-Cu redox system, the oxidation of propylene to acetone is relatively easy, but the reaction is slow for butenes and higher olefins with 5 or more carbon atoms. Olefins are slightly soluble in water, so the production of methyl ethyl ketone using this catalyst is impractical. To search for effective catalysts for the oxidation of butene-1 to methyl ethyl ketone, the authors carried out extensive studies of the catalytic properties of 3d-transition metal chlorides in the oxidation of butene-1 with molecular oxygen [3, 4].

Complexes of monovalent copper with molecular oxygen were synthesized by dissolving anhydrous chloride in hexamethylphosphoramide. Benzonitrile was used as a modifying ligand. Thus, it was found that the addition of sulfolane, which is the main solvent, increases the yield of methyl ethyl ketone. This method was used to synthesize 3d-transition metal complexes. [6,7,9]

It has been established that the synthesized copper complex is an effective catalyst for this reaction, forming a stable complex with molecular oxygen at an intermediate stage. Typically, when a solution of Cu(I) compounds absorbs oxygen, copper is oxidized by oxygen to copper. A similar oxidation

reaction was also expected in this case. However, the solution of the divalent copper compound complex has a reddish-brown color.

If the solution absorbs oxygen, the complex turns dark green. The UV absorption spectra of solutions of these substances were taken. The spectra of Cu(1) and O₂ scavenging complex solutions are completely different from the Cu(2) complex solution spectra, with the green oxygen scavenging complex having an absorption maximum at 265 nm. the formation of oxygen complexes with coordinated oxygen molecules was shown [5].

A feature of this oxygen complex is that the coordinating oxygen is not desorbed even when heated, i.e., the absorption is irreversible. Apparently, O₂ molecules are coordinated by metal ions, polarized, and activated as a result of electron transfer. As mentioned above, methyl ethyl ketone is produced by the oxidation of butene-119 with bound oxygen activated by a transition metal. Assuming that butene-1 can also form complexes and be activated as a result of complex formation, then it will be possible to carry out oxidation reactions at low temperatures and pressures. Therefore, various complexes of transition metals of the palladium group were investigated. It was found that Pd(2)Cl₂ chloride forms a complex with GMPA.

Various studies have been carried out to identify the conditions for the formation of more stable butene complexes. Nitriles such as C₆H₅CN were added as modifying ligands (additional complexing agents). In this case, a new complex is formed.

Absorptive properties of the complex When comparing a single solvent consisting of HMPA and benzonitrile (C₆H₅CN) with the Pd(2) complex, the pure solvent absorbs about 1.6 times more butene than the Pd(2) complex, although the absorption of butene-1 is large in case. Because this reaction is driven by the formation of new butene complexes, the resulting butene-1 complexes contain significantly activated butene-1. The coordinated butene-1 is then oxidized with complexing oxygen to give methyl ethyl ketone. The implementation of the method for producing methyl ethyl ketone according to the developed method by coordinating with a transition metal and thereby activated butene and oxygen has many differences from existing methods for producing methyl ethyl ketone and some advantages. The process is carried out under mild conditions at atmospheric pressure and a temperature of 80°C, which ensures high yields and selectivity in obtaining target products. In this case, the valencies of 20 transition metal ions remain unchanged and water does not participate in the formation of methyl ethyl ketone. The yield of methyl ethyl ketone is 92% in 1 hour and 98% in 2 hours [5].

The proposed binary system is capable of coordinating molecular oxygen with butene-1, so the reaction between butene-1 and O₂ requires a specific composite catalytic system, rather than a direct oxidation reaction between them.

With its help, it becomes possible to carry out the oxidation reaction in activated coordination. Mild reaction conditions significantly reduce the amount of by-products and simplify the stages of obtaining and isolating the main product - methyl ethyl ketone.

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