Summary

In this thesis several new insights and possibilities concerning gold catalysis are presented. One of the goals was to investigate the role of the Au particle size. For the activity of gold based catalysts, it is very important that the gold particles have a size below 5 nm. The question arises is this effect unique for gold catalysis or can a similar effect be found with the related metals silver and copper? Therefore the catalytic behavior of comparable copper and silver based catalysts were investigated. This was mainly done by testing the activity of the catalyst from room temperature up to 400 °C in the following reactions: selective oxidation of CO in chapter 2, ammonia oxidation in chapter 3 and the oxidation of methanol, ethanol and propanol in chapters 4, 5, 6 and 7. Also the composition and particle size of the catalysts are determined. Copper and silver were chosen as they chemically resemble gold. The effect of particle size is described in chapter 2 and 3. The results show that nanoparticles of copper and silver show different behavior or enhanced activity compared to larger particles and hence the particle size effect is important in copper and silver based catalysts.

When the active metals are deposited as nanoparticles, the catalytic activity is greatly influenced by the interaction with support and additives. In this thesis the role and interaction of Li$_2$O and CeO$_x$ with the metal nanoparticles and support is investigated. Earlier, it was found that Li$_2$O acts as a structural promoter of gold by preventing sintering of the gold nanoparticles. In the studies described in this thesis this same effect was also found for the copper and silver based catalysts. However, it was found that the role of Li$_2$O is not solely the prevention of sintering but it also poisons the acidic sites, which are active in the catalytic reactions of alcohols, of the alumina support and so has a great influence on the activity and, especially, the selectivity of the catalysts. The addition of Li$_2$O results in a catalyst with is capable of converting ethanol with high selectivity to ethylene oxide, especially when low oxygen concentration of oxygen in the gas feed is used. This has not been reported before. The use of Li$_2$O, which poisons sites which are catalytic active, gave the
researchers more insight into the mechanism of the concerned reactions and several mechanisms could be proposed.

CeO$_x$ is a well-known co-catalyst, which can be used as a catalyst itself. One of the most active catalyst in oxidation reactions contains both Au and CeO$_x$. On this catalysts the reactant is activated on the gold and the cerium oxide is providing active oxygen. Here the structure and average size of the metal oxide is important. Indeed we found that for all investigated reactions and all used metals the addition of cerium oxide resulted in a catalyst with stronger oxidizing characteristics, and an increase in oxygen conversion. This resulted in higher selectivities towards CO and CO$_2$ despite the relatively low content of oxygen used.

The results concerning the formation of ethylene oxide from ethanol on gold based catalysts motivated us the investigate the possibility of converting 1-propanol and/or 2-propanol directly to propylene oxide. This would be a very interesting reaction as there is no suitable catalyst for this process. In this exploratory study in chapter 7 we found that gold based catalysts are capable of converting propanol to acetone and propylene. Minute traces of propylene oxide are found. It is suggested that the conversion of 1-propanol to acetone and 2-propanol is proceeding via a propylene oxide intermediate.