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Catalytic performance of MoVTeNbO catalyst supported on SiC foam in oxidative dehydrogenation of ethane and ammoxidation of propane

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ABSTRACT

MoVTeNbO catalysts have been supported on pre-oxidized SiC foam by dip coating from slurry containing the precursors for all constituents and tested as catalysts for the oxidative dehydrogenation of ethane and the ammoxidation of propane. The characterization of the coating obtained by different techniques showed that a one layer coating contained almost no active M1 phase and mainly M2 phase whereas a two layers coating presented a significant amount of M1 phase. The structured catalyst thus obtained with two layers appeared highly efficient and stable in both reactions between 380 and 420 °C and the oxide coating exhibited fairly good mechanical properties. When compared to the most efficient powdered pure M1 phase in the case of ethane oxidative dehydrogenation and M1 + M2 phase mixture in the case of propane ammoxidation, the coated foam appeared less active; the reduced activity was attributed to the lower content in M1 phase of the supported catalyst. The selectivity to ethylene was lower, that to acrylonitrile was comparable; the decrease in ethylene selectivity was due to the presence of other active but not selective phases like $TeMo_5O_{15}$ and $Mo_{0.97}V_{0.95}O_5$. In the case of acrylonitrile the synergy effect that takes place between the M1 and M2 phases also took place between the $Mo_{0.97}V_{0.95}O_5$ phase and the M2 phase and maintained the selectivity to acrylonitrile. Improvements in preparation conditions are still possible to open the door to intensification and miniaturization on the process of propane ammoxidation on these efficient coated SiC foam structured reactors.

1. Introduction

Research efforts on light alkanes oxidation or ammoxidation have led to catalytic systems with yields approaching those that could compete with the current technologies indicating the probability of viable industrial processes. The best catalysts designed up to now correspond to complex multi-component MoVTeNbO oxide systems [1–3].

By now it is well established that the active and selective phase of the MoVTeNbO catalyst system corresponds to an orthorhombic phase denoted M1 with the composition $(AO)_{2-2x}(A_2O)_xM_{20}O_{56}$ ($0 \leq x \leq 1$, A = Te, Sb and M = Mo, V and Nb) [4,5]. The first patents published on the MoVTeNbO system in the ammoxidation of propane reported that the presence (or addition) of another phase called M2, also orthorhombic (pseudo hexagonal) with the composition $(TeO)_2M_6O_{18}$ (M = Mo, V and either with or without Nb), increased the yield in acrylonitrile [5–7]. The synergetic effect could be explained by intermediate propene, formed from propane on M1, being allowed to readsorb on M2, where it was selectively transformed to acrylonitrile [8–12]. Such effect that was evidently not observed for oxidative dehydrogenation of ethane to ethylene occurred only for Te containing catalysts since the Sb containing M2 phase was poorly active for propene ammoxidation or oxidation [12]. During the last decade several other ways to improve the catalytic properties of the MoVTeNbO catalytic system have been explored. Studies to enhance directly the catalytic performances of the M1 phase based on modifying its cationic composition appeared rather disappointing [13–15]. Limited but effective successes have been obtained with alkali metals and phosphorus added as surface doping [14,15]. Attempts to improve the catalysts by supporting them on various supports like alumina, silica, TiO_2 or Nb_2O_5, appeared unsuccessful since the supporting appeared to strongly inhibit the crystallization of the M1 phase, almost undetectable [16,17]. In the case of SiO_2 it was shown that addition of SiO_2 in small amount was without any effect on selectivity and beneficial to the conversion for ODH of ethane since it decreased of the agglomeration and sintering of the M1 phase crystallites [18]. At high content, this effect was still present but did not lead to an increase of activity as shown in the case of propane ammoxidation [19]. Marginally one may note that efficient doping of the M2 have been

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