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## Mesoporous and Macroporous Mixed Mo-V-Te-Nb Oxides for Propane (Amm)oxidation

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### Introduction

Ordered porous mixed metal oxides are highly promising for applications in selective oxidation catalysis because of their tunable "bulk" and surface compositions, variable metal oxidation states, high surface areas, as well as large and uniform pore sizes. In particular, vanadium, molybdenum, tellurium and niobium-based mixed oxide phases are very attractive for selective oxidation and ammoxidation of lower alkanes, such as propane. Mesoporous single and mixed oxides of vanadium<sup>[1]</sup>, molybdenum<sup>[1]</sup>, and niobium<sup>[1-4]</sup>, and macroporous niobium oxide<sup>[5]</sup> have been reported. However, preparation of thermally stable ordered porous mixed multicomponent metal oxides with high surface areas still remains a challenging task.

In this work, we explore the synthesis of orthorhombic (M1) Mo-V-Te-Nb-O catalysts inside the pore channels of ordered mesoporous niobate host. The use of such mesoporous host allows confining the synthesis of the catalytic Mo-V-Te-Nb-O phases to the nanoscale region of their pore space. In the second approach, ordered hexagonal-structured macroporous Mo-V-Te-Nb-O was synthesized by sphere templating method.

### Materials and Methods

Ammonium paramolybdate, ammonium metavanadate, telluric acid, niobium oxalate, niobium chloride, Pluronic P123 were used as received. Polystyrene spheres were synthesized using emulsion polymerization process<sup>[6]</sup>.

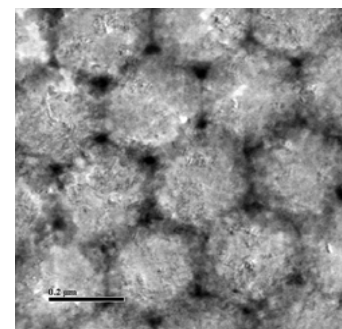
Mesoporous mixed Mo-V-Te-Nb-O catalysts were synthesized by incipient-wetness impregnation (IWI) technique and evaporation-induced self-assembly (EISA) method. Macroporous Mo-V-Te-Nb-O catalyst was synthesized using polystyrene spheres as template. The bulk structure of these mixed metal oxides obtained was characterized by XRD, SEM and high resolution TEM. The fixed-bed microreactor studies of propane oxidative dehydrogenation and propane ammoxidation were complemented by *in situ* Raman studies of these novel catalysts and reference bulk M1 catalyst.

### Results and Discussion

High surface area mesoporous mixed MoVTeNb oxides were synthesized, which exhibited higher activities than the binary system<sup>[4]</sup> in propane ODH reaction (Table 1). No characteristic peak of M1 phase was observed in Raman spectra of these oxides. This could explain their lower activities than that of pure M1 phase. Ordered hexagonal-structured macroporous MoVTeNb oxide, synthesized using polystyrene spheres as the template, possessed pore diameter of 250 nm and surface area of 70 m<sup>2</sup>/g. It exhibited rutile phase according to its XRD pattern. The selectivity to acrylonitrile in propane ammoxidation was low due to lack of M1 phase.

**Table 1. Propane ODH over mesoporous mixed MoVTeNb Oxides compared with reference catalysts**

Catalyst	Surface area (m <sup>2</sup> /g)	Rate at 420 °C (μmol/m <sup>2</sup> s)	Activation energy (kJ/mol)
V <sub>0.076</sub> Nb <sub>0.245</sub> O <sub>x</sub> <sup>[4]</sup>	159	0.355	80-102 <sup>[7]</sup>
Mo <sub>0.031</sub> Nb <sub>0.307</sub> O <sub>x</sub> <sup>[4]</sup>	159	0.266	99-117 <sup>[8]</sup>
EISA-1	151	0.523	69
IWI-1	84	0.891	51
IWI-3	48	1.646	56
Pure M1	8	5.75	138



**Figure 1.** SEM image of hexagonal-structured macroporous MoVTeNb oxide obtained by sphere templating method.

### Significance

The methods described here represent promising synthesis approaches for the design of novel mixed transition metal oxides for catalytic applications.

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*This research was supported by the National Science Foundation CAREER CTS-0238962 grant to Dr. Vadim V.Guliants.*