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Selective and efficient synthesis of benzonitriles by direct ammoxidation of alkylbenzenes in the sub–nano spaces

Abstract

A nanotech for selective and effective synthesis of benzonitriles by ammoxidation reaction of alkylbenzenes under the normal concentration of reactants is presented. The high selectivity and efficiency for the synthesis of benzonitriles is due to the ammoxidation reaction takes place in the sub-nano space.

Volume 7 Issue 3 - 2018

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Received: April 12, 2018 | **Published:** June 07, 2018

Introduction

Benzonitrile and its derivatives are used in manufacturing other organic compounds such as pharmaceuticals, dyes, brightening agents, rubber chemicals and agrochemicals, epoxy curing agents, solvents, etc. Benzonitrile is industrially produced by the ammoxidation process of toluene on mixed transition metal oxides under the reaction conditions of low toluene concentrations (0.5–2%) to avoid the combustion reactions. Any new catalysts with high performances have not been reported to date.

In the Wang Institute of Nanotech for Advanced Catalysts (WINAS), it has developed the methods for fabricating the transition metal oxide clusters in the pores of β-zeolites, which are used as the catalysts for the selective ammoxidation reactions of toluene, xylenes and chlorotoluenes to benzonitrile, methylbenzonitriles and chlorobenzonitriles, respectively. The catalysts were characterized by XAFS,XRD,TEM, etc to prove that the transition metal oxide clusters were successfully fabricated in the pores of the beta zeolites. The transition metal (Ni,Co,V,Mo,W, etc.) oxide clusters in the zeolite pores were active for catalyzing the gas-phase selective ammoxidation of alkylbenzenes in the sub-nano spaces as shown in Figure 1. The combustion reactions of aromatic reactants and ammonia to CO_2 and N_2 are effectively suppressed in the confined space of β-zeolite pores at the higher toluene concentration of 10-20%. The formation rates of benzonitrile, methylbenzonitriles and chlorobenzonitriles at 703K were 0.8g h-1 gcat-1, 0.6g h-1 gcat-1 and 1.1g h-1gcat-1, respectively with the nitrile selectivity of up to 99%. Figure 2 shows the performance of the novel catalysts developed in the WINAC and compared with the commercial catalysts for the synthesis of benzonitriles reported in references.[1,2]

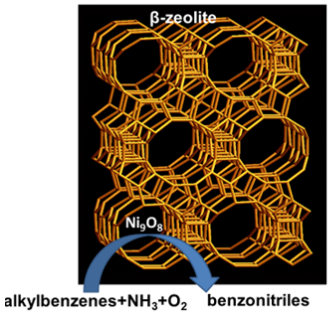
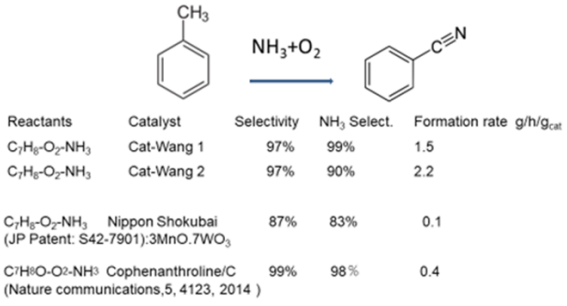


Figure 1 The structure of the transition metal oxide clusters fabricated in the β-zeolite pores as the catalyst for the synthesis of benzonitriles in the sub-nano spaces.



Reactants	Catalyst	Selectivity	NH_3 Select.	Formation rate $g/h/g_{cat}$
C_7H_8-O_2-NH_3	Cat-Wang 1	97%	99%	1.5
C_7H_8-O_2-NH_3	Cat-Wang 2	97%	90%	2.2
C_7H_8-O_2-NH_3	Nippon Shokubai (JP Patent: S42-7901):3MnO.7WO_3	87%	83%	0.1
C_7H_8O-O_2-NH_3	Cophenanthroline/C (Nature communications,5, 4123, 2014)	99%	98%	0.4

Figure 2 Comparison of the catalysts-wang (WINAC) with other catalysts used in industry catalysts for benzonitrile synthesis (Higher selectivity with munch higher activity).

Conclusion

The transition metal (Ni,Co,V,Mo,W, etc.) oxide clusters are successfully fabricated in the beta-type zeolite pores which are highly active and selective for synthesis of benzonitriles via ammoxidation of alkylbenzenes under the normal concentrations of the reactants.

Acknowledgments

None.

Conflicts of interest

The author declares there is no conflict of interest.R

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2. Jagadeesh RV, Junge H, Beller M. Green synthesis of nitriles using non-noble metal oxides-based nanocatalysts. *Nat Commun*. 2014;5:4123.

Citation: Wang L. Selective and efficient synthesis of benzonitriles by direct ammoxidation of alkylbenzenes in the sub–nano spaces . *J Nanomed Res*. 2018;7(3):200–201. DOI: 10.15406/jnmr.2018.07.00188