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Preface

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22-23 AUGUST 2019

***Enhancing Engineering
Innovation Towards
A Greener Future***

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IOP Conference Series: Materials Science and Engineering

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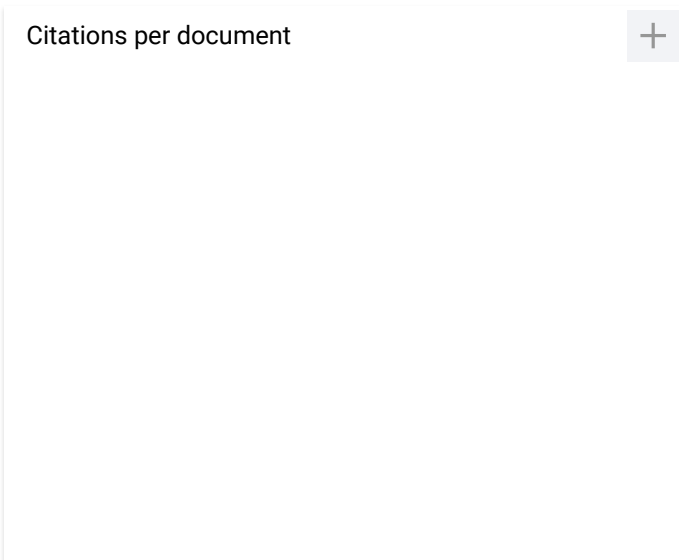
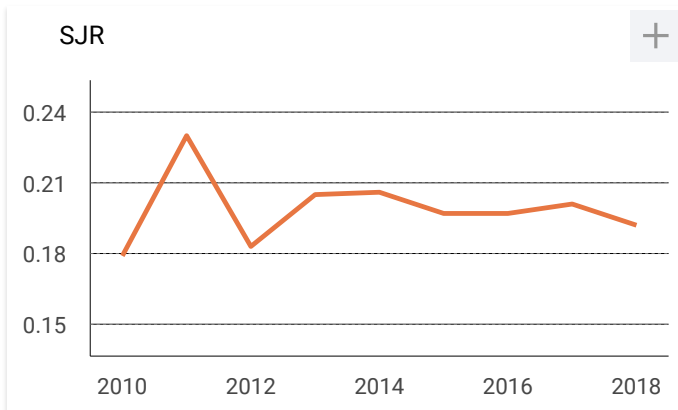
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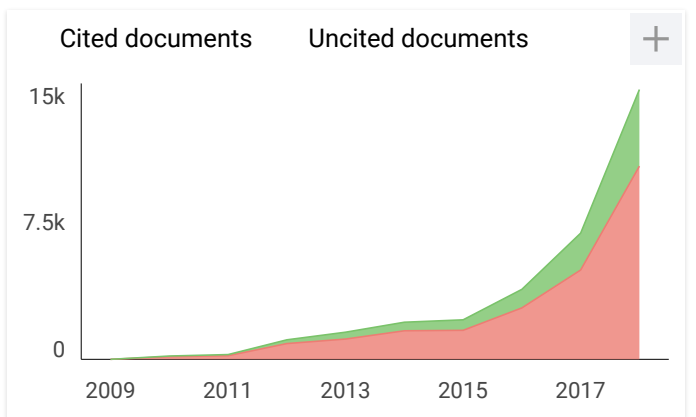
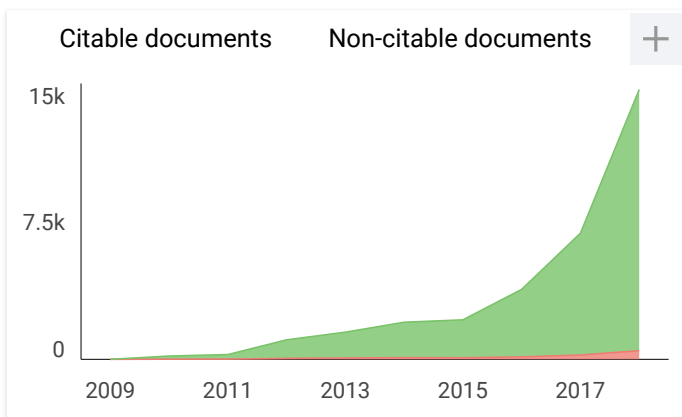
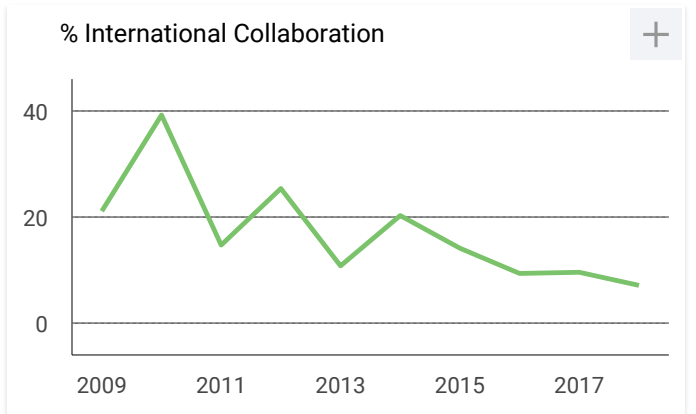
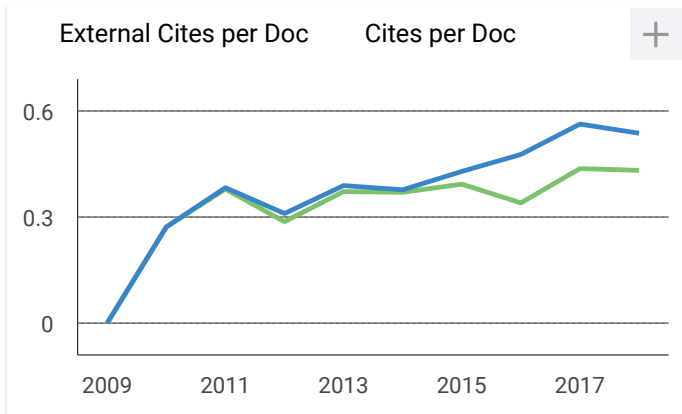
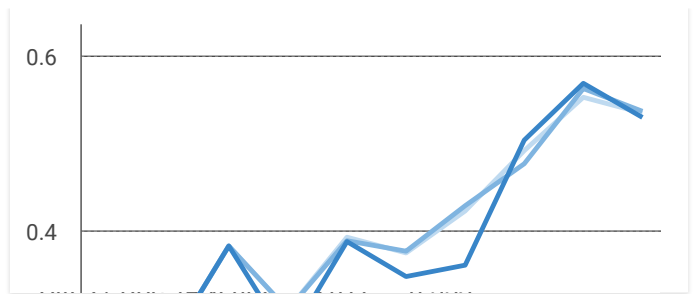
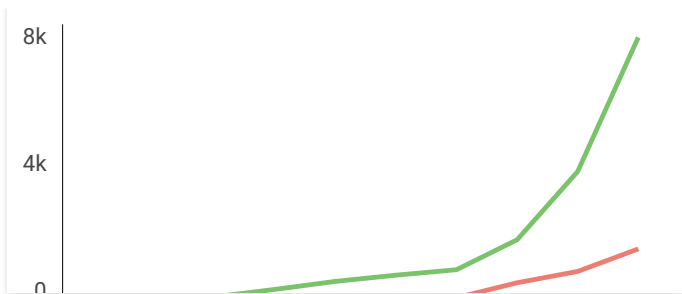
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Welcome Remarks,
Chair of the Steering Committee

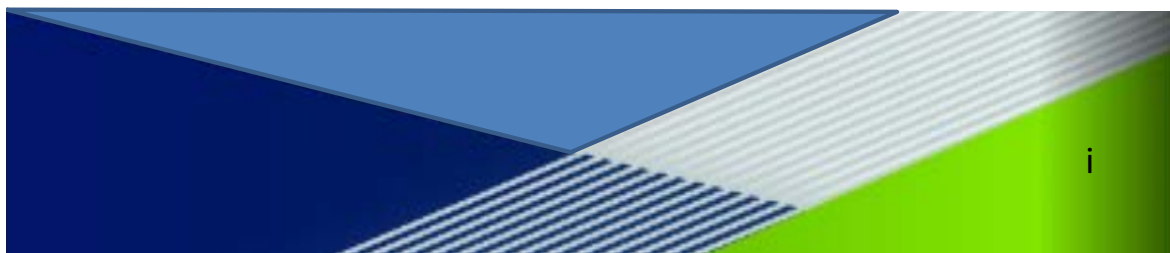
It is a great pleasure to welcome all of you to Bali and to the International Conference on Informatics, Technology, and Engineering 2019 (InCITE 2019) held by the Faculty of Engineering, University of Surabaya (UBAYA) in collaboration with The University of Adelaide, Australia and Sirindhorn International Institute of Technology (Thammasat University), Thailand. The first InCITE has been successfully held in Bali, Indonesia in 2017. We are very delighted to host the second InCITE here in Bali, Indonesia again.

There are more than 75 presentations in this conference. We welcome leading experts not only from Indonesia, but also from different parts of the world. The experts will share the knowledge and experiences in the fields of informatics, technology, science, and engineering. The main theme of this conference is **Enhancing Engineering Innovation Towards A Greener Future** in response to several world challenges including sustainable development, global convergence of information and communications technologies, climate change and global warming as well as the depletion of unrenewable natural resources. We hope this conference will provide you a good opportunity to get to know each other better and consolidate bonds of friendship and mutual trust.

We would like to express our sincere gratitude to the Keynote and Plenary speakers, International Scientific Committee, Steering Committee, and Organising Committee for their huge efforts to make this conference successful.

Thank you all for your support and attendance at InCITE 2019. Please enjoy the conference and Bali !

Asst. Prof. Djuwari, Ph.D.





Preface

Welcome Remarks,
Chair of The Organizing Committee

Welcome to Bali, Indonesia to all delegates and presenters. It is my pleasure and privilege to welcome all of you to the 2nd (second) International Conference on Informatics, Technology, and Engineering 2019 (InCITE 2019) held by the Faculty of Engineering, University of Surabaya (UBAYA) in collaboration with The University of Adelaide, Australia and Sirindhorn International Institute of Technology (Thammasat University), Thailand.

InCITE 2019 has received more than 75 papers to be presented in this conference. All papers represent four following parallel clusters: Green Design and Innovation, Green Manufacturing and Green Processes, Power System and Green Energy Management, and The Role of IT in Innovation Enhancement. Each cluster supports the main theme of the conference, which is **Enhancing Engineering Innovation Towards A Greener Future**. The engineering innovation is the key to increase our awareness in maintaining the sustainable growth and development in the world.

The Organising Committee of InCITE 2019 would like to express our sincere gratitude for the tremendous supports and contributions from many parties. The supports from The Faculty of Engineering of UBAYA, keynote and plenary speakers, our International Scientific Committee, the Steering and Organising Committees are really acknowledged.

The last but not the least, thank you for your supports, enjoy the conference and we hope through this meeting all of you can extend your networks and collaborations.

Asst. Prof. Putu Doddy Sutrisna, Ph.D.



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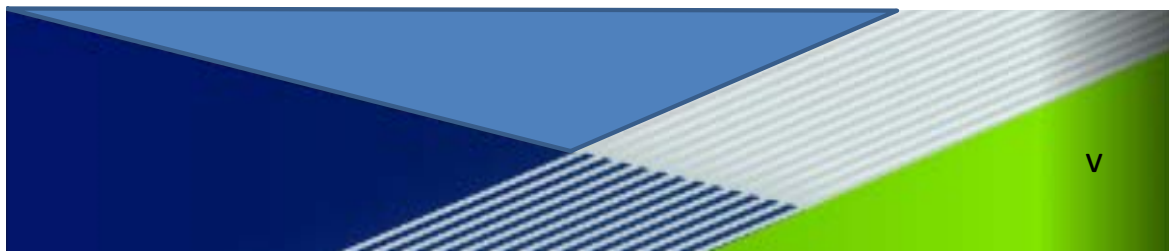


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The Influence of water and catalyst leach process toward propane oxidation on MoVTeNb catalyst

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The Influence of water and catalyst leach process toward propane oxidation on MoVTenb catalyst

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Abstract. The effect of the water stream to the propane oxidation on diluted MoVTenb catalyst has been investigated. The present work has elucidated that careful operation of high throughput instrumentation can be used in various beneficial ways to speed up the discovery process of improved catalysts in other forms than enabling efficient trial-and-error testing of compositional variations of a given catalyst system. The result shows that the addition of massive amounts of water to the feed should have a negative influence on the kinetics, as water will compete with all other polar molecules in the system for adsorption sites. This work also investigated the effect of catalyst leach process toward propane oxidation. From the result, it can be described that catalyst leach process tends to reduce the phase of the catalyst that responds to the total oxidation of propane. This work also proposed the reaction network and gave the comparison between the propane oxidation reaction kinetic using leached and un-leached catalyst. The result showed that the activation energy of the acrylic acid formation on the leached catalyst was slightly higher than that of on un-leached catalyst. On the other hand, the activation energy of the carbon dioxide formation on the leached catalyst was much higher than that of on un-leached catalyst. It can be described that the leaching process to the catalyst can reduce the phase of the catalyst responsible for the total oxidation of propane.

Keywords: *MoVTenb catalyst, propane, oxidation, water, leach process*

1. Introduction

The oxidation process using a catalyst has developed into a very important technology in the modern chemical industry. One of the important processes that utilize this technology is the functionalization of aromatics and alkanes, due to the advantageous economy and the easy processing. Acrylic acid is one of the most important catalytic oxidation products, in which its consumption in 2020 is expected to reach 8,000-kilotons. Acrylic acid is widely used in the industry as intermediate for polyacrylates and methacrylate esters production. These products can be used for various purposes, such as super adsorbents. Acrylic acid is presently factory-made from propylene in two steps via acrolein in a gas phase using special catalysts which the yield up to 80% [1]. Despite the promising yield of the product, the price of propylene has seen attention being drawn to develop a method for producing acrylic acid alternatively by direct oxidation of propane in the presence of a catalyst. One of the promising catalysts that are widely studied is a mixed metal oxide, such as MoVTenb catalyst [2-7].



The problem that is still faced in the utilization of this catalyst is the control of the complexity of the reaction network consisting of consecutive reactions and parallel reactions [3, 8-10]. The phase-in MoVTeNb which is responsible for the production of acrylic acid from direct oxidation of propane is referred to as "M1" phase [2, 4, 11-13]. Mo and V are very suitable for the oxidation of alkanes via alkenes to carboxylic acids [14, 15]. Some results of previous studies have reported the proposed reaction mechanism [3, 9, 16], as well as the structure of the catalyst during the reaction [4, 17-21]. The presence of water in this oxidation reaction provides a reaction advantage as has been reported by previous researchers [3, 22-25].

In this paper, we will discuss the effect of the presence of water on catalytic reactions and the effect of the leaching process on diluted MoVTeNb oxide catalyst. Water is believed to have an indirect role in the process of propane oxidation reactions to acrylic acid. The leaching process is believed to effect on the phase composition of M1 and M2 in the catalyst which has a very important role in the oxidation reaction mainly related to the reaction network of propane to acrylic acid.

2. Experimental

2.1. Materials

The materials used for catalyst synthesis were Telluric acid (Aldrich), Ammonium metavanadate (Riedel-de Haën), Niobium ammonium oxalate (Aldrich), Ammonium heptamolybdatetetrahydrate (Merck), Aerosil300 (Degussa). The gases used for reactants were oxygen and propane (MOX).

2.2. Instrumentation

Nanoflow catalytic reactor is one of the equipment that is used for this high throughput experimentation study.

2.3. Procedure

Preparation of MoVTeNb oxide catalysts and kinetic studies was carried out according to the procedure described previously, and some kinetics data were also reported previously [4, 5, 7]. The catalytic reaction was done by presenting a small quantity of catalyst (0.1 g) mixed in SiO₂ to a total volume of 0.5 ml. The high total flow rates (15 mlN/min, GHSV = 1800 h⁻¹). The reaction rate is calculated by Equations presented below:

$$\text{Reaction rate (mol/h. gcat)} = \frac{\%vol \times total \text{ flow} \times d \times 60}{Mr \times catalyst \text{ weight}} \quad (1)$$

Where Mr is molecular weight and d is density.

To study the effects of water dependence, the concentration of water varies and other concentrations are fixed. To determine activation energy, the oxidation reaction was studied at four different temperatures, namely, 653, 663, 673 and 683 K. After taking the natural logarithm of the equation, it can be seen that a plot of ln k versus 1/T should be a straight line whose slope is proportional to the activation energy.

$$k = Ae^{-\frac{E_A}{RT}} \quad (2)$$

Where k is the rate constant, T is the absolute temperature (in Kelvin), A is the pre-exponential factor, E_a is the activation energy for the reaction, and R is the universal gas constant.

3. Result and Discussion

The characteristic of the MoVTeNb oxide catalyst has been reported in our previous work [4]. The catalyst consists of two main phases namely M1 and M2 phases. M1 phase is more related to the structure of Mo₅O₁₄ and is responsible for the oxidation process of propane to acrylic acid, while the M2 phase is

more related to the orthorhombic variant of HTB structure which is responsible for the total oxidation process of propane to CO_x. Based on catalyst characterization, it shows that the M1 phase is more dominant than the M2 phase.

3.1. Effect of water

There are several opinions about how water affects the performance of the catalyst and the distribution of products, such as by facilitating desorption of oxygenating from the catalyst or modifying the catalyst structure. The present study has substantiated that the modification of the catalyst surface required for partial oxidation by water concentration in the feed is a fact for the systems studied here. The beneficial effect of water in helping to wash off the partial oxidation products and to protect them from over-oxidation by blocking sites next to the adsorption site of the freshly formed products and so inhibiting the access of activated oxygen from neighboring sites have also been identified in the present study.

To interpret this point, kinetic investigations were lead with water as the varied feed component. The rate of propane disappearance and acrylic acid formation were calculated and plotted against the water concentration (see Figure 1 and 2).

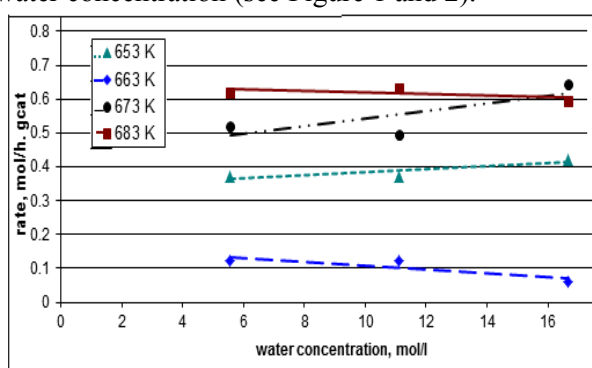


Figure 1. Rate of propane disappearance with respect to water concentration at 653, 663, 673, and 683K

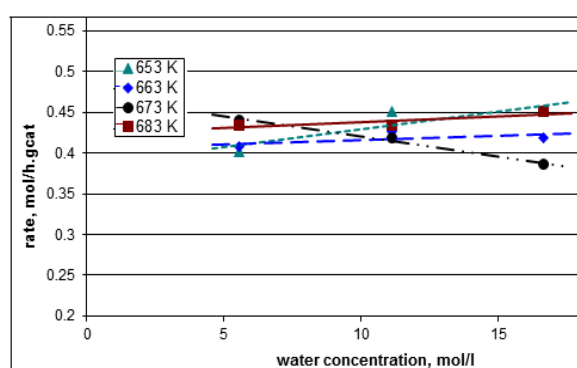


Figure 2. Rate of acrylic acid formation with respect to water concentration at 653, 663, 673, and 683 K

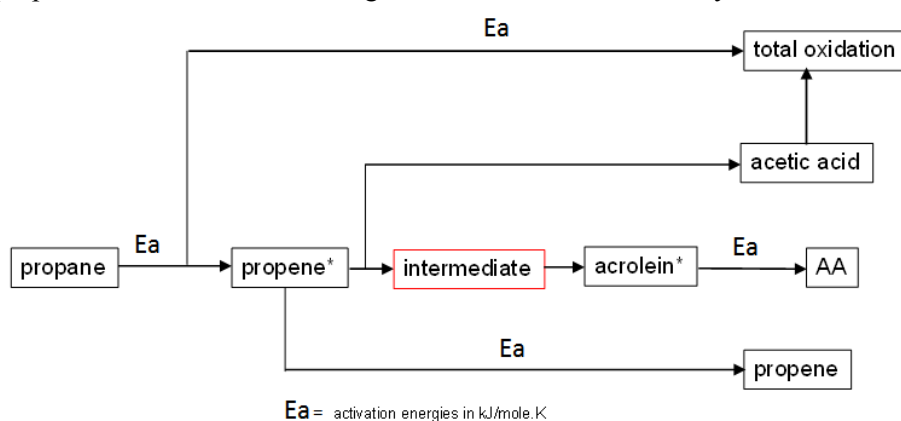
The reaction stoichiometry for propane to acrylic acid is $C_3H_8 + 2 O_2 \rightarrow C_3H_2COOH + 2 H_2O$. From the result above, the rate of propane disappearance (r_{prop_dis}) and acrylic acid formation (r_{AA_form}) from the selective oxidation of propane over this catalyst with respect to water concentration can be determined by the equation (see table 1).

In all cases, a small value of the reaction order with respect to water was obtained. This is unexpectedly taking into account that water is a reaction product in all processes considered here and that it will control the processes of C-H activation leading to (intermediate) OH groups. The addition of massive amounts of water to the feed should have a negative influence on the kinetics, as water will compete with all other polar molecules in the system for adsorption sites. It has to be assumed that the integrity of the catalysts requires its surface to be covered to a substantial extent with water. On such a surface the additional water created during the reaction of propane will not affect greatly the coverage and hence will not lead to a significant reaction order detectable by formal kinetics. In contrast to current opinion, the low values of reaction order do not imply that water cannot interfere with the reaction kinetics.

For acrylic acid a strong dependence of the reaction order on temperature is observed, showing at low temperatures a slightly beneficial and for high temperatures a slightly inhibiting effect of water. As the C-H chemistry should not be relevant, it may be concluded that at low temperatures the beneficial washing off effect is at work whereas at high temperatures the site competition effect prevents the optimization of the surface coverage with other species than water indicating that water desorption must be a strongly activated process.

3.2. Effect of leach process

For further development of the catalyst system, two conclusions can be drawn. It is very important to design the dilution of the active phase in such a way as to maximize the hydrothermal stability and to optimize the heat management of the entire catalyst. Concerning the active phase, it became evident that a single phase is essential as then the chances to bring about a single type of active sites, capable to perform minimum consecutive total oxidation the selective oxidation of propane. To this end, leaching and nanostructuring of the surface are suitable concepts. To clarify this point, a reaction mechanism has been proposed (Scheme 1, [7]) and then it will be compared the amount of energy activation of the propane oxidation reaction using unleached and leached catalysts.



Scheme 1: Schematic representation of the reaction network

Overall, this propane oxidation reaction consists of three parallel processes paths. These processes are one main path that produces the desired product, acrylic acid. The other pathway is the total oxidation of propane to carbon dioxide, and another pathway is the process of desorption of propene. One thing that is quite good in this reaction process is that the total oxidation pathway of propane requires a fairly large activation process, so it is technically quite difficult to occur. The proposed reaction mechanism is consistent with the presence of 2 types of active catalyst sites referred to as M1 for the partial oxidation pathway to produce acrylic acid and M2 for the total oxidation pathway.

To further understand the effect of washing or leaching the catalyst on the presence of phases responsible for the formation of acrylic acid, activation energy calculations for each process are presented in table 1. This washing is intended to reduce the M2 phase which leads to a total oxidation reaction. It is expected that the reduction of the M2 phase in total in the catalyst material will be dominated by the M1 phase which pushes towards selective oxidation so that the desired product will be produced more.

Table 1. Comparison of activation energy in each process of selective propane oxidation using leached and unleached MoVTeNb [5,7]

Processes	Rate of reaction (r)		Activation Energy (kJ mol ⁻¹)	
	Unleached catalyst	Leached catalyst	Unleached catalyst	Leached catalyst
Propane disappearance	$k[\text{C}_3\text{H}_8]^{1.00} [\text{O}_2]^{0.24}$	$k[\text{C}_3\text{H}_8]^{1.00} [\text{O}_2]^{0.21}$	21,8	27,3
Propene formation	$k[\text{C}_3\text{H}_8]^{1.00} [\text{O}_2]^{0.00}$	$k[\text{C}_3\text{H}_8]^{1.00} [\text{O}_2]^{0.00}$	62,7	40,9
CO _x formation	$k[\text{C}_3\text{H}_8]^{1.00} [\text{O}_2]^{0.21}$	$k[\text{C}_3\text{H}_8]^{1.00} [\text{O}_2]^{0.31}$	123,2	177,3
Acetic acid formation	$k[\text{C}_3\text{H}_8]^{0.22} [\text{O}_2]^{0.24}$	$k[\text{C}_3\text{H}_8]^{0.83} [\text{O}_2]^{0.42}$	46,9	91,6
Acrylic acid formation	$k[\text{C}_3\text{H}_8]^{0.50} [\text{O}_2]^{0.11}$	$k[\text{C}_3\text{H}_8]^{0.49} [\text{O}_2]^{0.09}$	32,9	30,3

From table 1, it can be described that the process of removal (conversion) of propane requires slightly higher energy in the reaction using leached catalysts. It can be logically explained that the washing process causes several active catalytic sites both M1 and M2 phases to be washed away. The reduced active site will complicate the process of propane removal/conversion. However, in the process of using

leached catalysts, it appears that the formation of propene is easier. Propene is an intermediate product which will immediately turn into the next oxidation product, acetic acid, and acrylic acid. The formation of acetic acid which is a by-product requires higher energy, while the formation of acrylic acid as a product of the main reaction pathway requires lower energy. This proves that the washing process is quite successful in eliminating the M2 phase which is responsible for the total combustion of propane. This is supported by the calculation of the activation energy of CO_x formation which shows a very high increase. This suggests that CO_x formation is difficult. On the other hand, converting propane towards propene and continuing towards acrylic acid is even easier. This confirms that the washing process succeeded in reducing the M2 phase which is responsible for the total oxidation reaction of propane to CO_x. In addition the washing process successfully maintains the M1 phase which is responsible for the selective oxidation reaction of propane to acrylic acid.

4. Conclusion

In general, this study has provided a phenomenological framework for understanding the action of MoVTeNb in propane oxidation catalysis. It was found that there are two types of reaction sites present, one of which requires the presence of excess water to function. The extent to which these sites will be present depends on the details of the bulk material nanostructuring and can be modified by synthetic and post-synthetic steps. One of these steps is the washing process. Parallel reaction pathways for partial oxidation need to be treated more carefully so that acrylic acid can be produced optimally.

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