



DIRECT ULTRASOUND SYNTHESIS OF VANADYL  
PYROPHOSPHATE CATALYST FOR PARTIAL OXIDATION OF  
N-BUTANE TO MALEIC ANHYDRIDE

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*Peer-review under responsibility of 3<sup>rd</sup> Asia International Multidisciplinary Conference 2019 editorial board  
(<http://www.utm.my/asia/our-team/>)*

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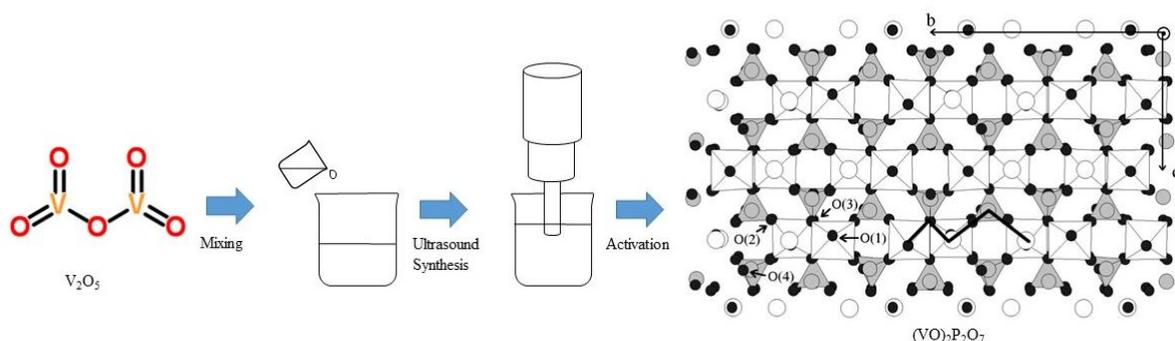
## RESEARCH HIGHLIGHTS

This research revealed that the vanadyl pyrophosphate catalysts can be produced using direct ultrasound irradiation technique throughout the whole synthesis process. The findings indicated that the ultrasound produced catalysts gave higher activity and selectivity to maleic anhydride by showing smaller particle sizes, polycrystalline phase, increased of  $V^{5+}$  contribution, higher removal of oxygen species during reactivity testing compared to bulk catalysts. The ultrasound technique further showed significant reduction in catalyst production time from 48 hours to only 2 hours.

**Keywords:** *Ultrasound Synthesis, Vanadyl Pyrophosphate Catalyst, Organic, Dihydrate, Sesquihydrate*

## GRAPHICAL ABSTRACT

Highly crystallised vanadyl pyrophosphates were synthesised from vanadium pentoxide by different routes via direct ultrasound irradiation synthesis process. The effect of direct ultrasound onto different preparation route towards the physico-chemical properties, reactivity and catalytic performances of the VPO catalysts is reported.



## RESEARCH OBJECTIVES

As the conventional catalyst production methods still show limitation such as high preparation time, limited catalytic performances and etc. Therefore this research aims to imply an alternative energy source in developing catalyst faster and cost-efficient. Up to few years ago, use of ultrasound technique in chemistry was mainly domain in physical and polymer chemistry. The situation has now changed as the ultrasound technique is discovered to be not only enhance reaction rates but also direct reactions onto different pathways due to its acoustic cavitation effect. Thus, this research further examines the effect of ultrasound on physico-chemical properties, reactivity and catalytic performances on the catalysts.

## MATERIALS AND METHODS

Three VPO catalysts were prepared via direct ultrasound technique using organic, sesquihydrate and dihydrate routes (1–5), denoted as VPO<sub>ou</sub>, VPO<sub>us</sub> and VPO<sub>ud</sub> respectively. These catalysts were synthesised solely on direct ultrasound technique and calcined in *n*-butane/air mixture. The physico-chemical, reactivity and catalytic properties of the vanadyl pyrophosphate catalysts synthesised via direct ultrasound synthesis were characterised by using X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), chemical analyses, field emission scanning electron microscope (FE-SEM), high resolution transmission electron microscope (HR-TEM), XANES spectra and temperature-programmed reduction ( $H_2$ -TPR).



The effect of direct ultrasound onto different preparation route towards the physico-chemical properties, reactivity and catalytic performances of the VPO catalysts were examined.

## RESULTS

All ultrasound produced catalysts exhibited well-crystallised  $(VO)_2P_2O_7$  phase (1,3,5,6). VPO<sub>us</sub> and VPO<sub>ud</sub> showed  $\alpha_{II}$ -VOPO<sub>4</sub>, which led to an increase in average oxidation state of vanadium (3,5). The ultrasound produced catalysts were showing O1s approx. 530 eV, similar P2p value and V2p<sub>3/2</sub> at approx. 517 eV, giving vanadium oxidation state of approx. 4.0 – 4.2. FE-SEM micrographs showed the secondary structure consisting of thin plate-like crystals in different sizes agglomerate to each other due to cavitation effect (4,6). HR-TEM demonstrated the existence of polycrystalline phase. The nature of the oxidants was investigated by TPR in H<sub>2</sub>. VPO<sub>us</sub> showed highest amount of total removal of oxygen species suggesting that it had highest activity compared to VPO<sub>uo</sub> and VPO<sub>ud</sub>. The XANES measurement of these catalysts showed the occurrence of vanadium oxide reductions in flowing hydrogen gas, which indicates the presence of V<sup>4+</sup> and V<sup>5+</sup> species. Catalytic tests demonstrated that the activity and selectivity to maleic anhydride increased with direct ultrasound technique.

## FINDINGS

The crystallite size obtained was much smaller compared to the conventional while comprising well crystallised  $(VO)_2P_2O_7$  phase. Catalyst produced via direct ultrasound method were quite abundance of V<sup>4+</sup> with slight V<sup>5+</sup> present, because the ultrasonication produces strong convective currents which may lead to reduce in thickness of platelets of the catalyst revealing a better catalytic activity and selectivity. Thus, the catalysts in regardless of route prepared by direct ultrasound synthesis, were more reduced (V<sup>5+</sup> to V<sup>4+</sup>) and more polycrystalline nanometre particles, and indeed a large improvement in synthesis time to only 2 hours required for catalyst production.

## ACKNOWLEDGEMENT

This research work is supported by Immortal Green Industrial Sdn. Bhd. was gratefully acknowledged. Resources support from JKS Engineering (M) Sdn. Bhd. was gratefully appreciated.

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