

# COMPARISON OF MONOLITH AND CONFINED TAYLOR FLOW (CTF) REACTOR FOR PROPYLENE EPOXIDATION

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## Summary

Heterogeneous catalytic epoxidation of propylene to propylene oxide was investigated at various operating conditions in a monolith and confined Taylor flow (CTF) reactor. Titanium silicalite (TS-1) catalyst was immobilized on the outer surface of a rod in the CTF reactor and on the inner wall of a monolith channel, respectively. The effects of gas and liquid superficial velocity on the hydrodynamics were also investigated under Taylor flow regime. Both reactors showed high, stable productivity and selectivity under mild conditions but it was easier to control the catalyst coating in the CTF reactor.

## Keywords

Epoxidation; propylene; confined Taylor flow (CTF) reactor; monolith reactor

## Introduction

Propylene oxide is one of the most important chemical intermediates in the chemical industry. The propylene oxide industry has employed different types of commercial processes for several decades but interest in heterogeneously catalyzed oxidation of propylene with hydrogen peroxide as oxidant has recently emerged. Clerici and co-workers demonstrated that titanium catalyst (TS-1) is a promising catalyst for this route under mild conditions<sup>1</sup>.

Catalytic gas-liquid-solid reaction has played a vital role in a variety of chemical processes. Conventional reactors, such as stirred tank slurry, slurry bubble column and trickle-bed reactor, have been employed to cope with the catalytic multiphase reactions. However, due to the small particle size of TS-1 catalyst, there has been a demand for the application of a different type of reactor to the hydrogen peroxide-to-propylene oxide (HPPO) process. Monolith reactor can be considered as one of the alternatives. However, it has been known that it is not easy to make a suitable coating of catalyst materials on monolithic support. Therefore, a novel design of monolith reactor, confined Taylor flow reactor, was presented in this study. The performance of propylene epoxidation using hydrogen peroxide oxidant was investigated in a monolith reactor and compared with that of a confined Taylor flow (CTF) reactor.

## Experimental

TS-1 catalysts were synthesized following the procedure described in the literature<sup>1, 2</sup>. The TS-1 particles were

coated on the outer surface of a rod in the CTF reactor and on the inner wall of a channel in the monolith reactor, respectively. Silica nanoparticles were used to make a more stable coating of catalyst.

The monolith and CTF reactor consisted of a 500mm vertical column with temperature control. Gas and liquid flow rates were controlled by mass flow controllers and a peristaltic pump. The rod coated with TS-1 catalyst was positioned axis-symmetrically in a narrow channel of the CTF reactor. The reactors were operated in a Taylor flow regime at atmospheric pressure. An infra-red detection unit connected with a data logger and computer system was attached on the reactor column to collect and analyze the hydrodynamics data. The reaction products were sampled at regular time intervals. The concentrations of hydrogen peroxide were determined by iodometric titration and those of other components were analyzed on a gas chromatograph (Shimadzu GC-2014). Photographs of Taylor gas bubbles were taken using an Olympus high speed digital camera (i-SPEED 3).

TS-1 catalysts were characterized by XRD (PANalytical X'Pert PRO), FT-IR (Perkin-Elmer Spectrum 1), SEM (Leo Gemini 1525), TEM (JEOL JEM 2010), TGA (TA Q5000) and BET (Micromeritics TriStar 3000).

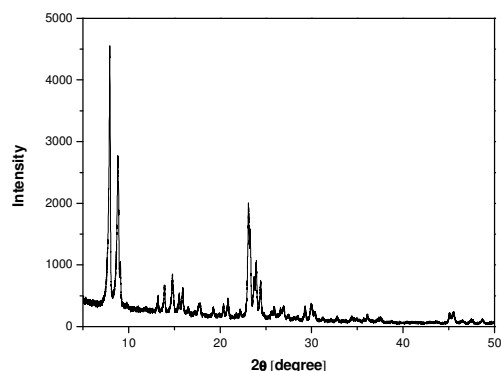
## Results

The characterization of TS-1 catalysts showed a good agreement with the results indicated in the original patent<sup>2</sup>. Figure 1 shows the XRD pattern of TS-1 catalyst. The single peaks at about 24.4 and 29.3 demonstrate a

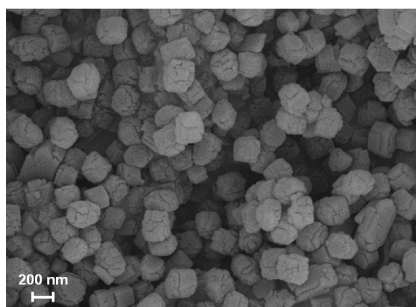
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conversion from a monoclinic symmetry (silicalite) to an orthorhombic symmetry (titanium silicalite). In the SEM image shown in Figure 2, well-defined, uniform cubic crystallites with rounding edges are observed. The size of particles is in the range of 200 – 300 nm.

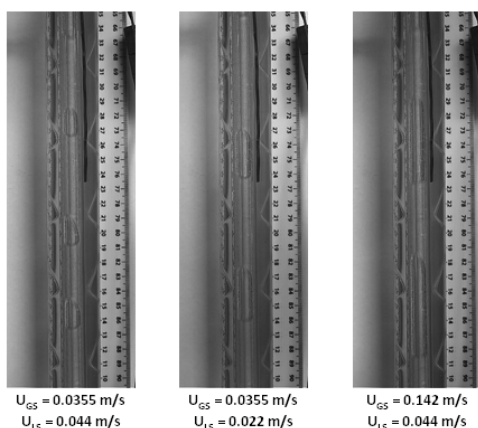


**Figure 1.** X-ray diffraction pattern of TS-1 catalyst.



**Figure 2.** Scanning electron micrograph of TS-1 catalyst.

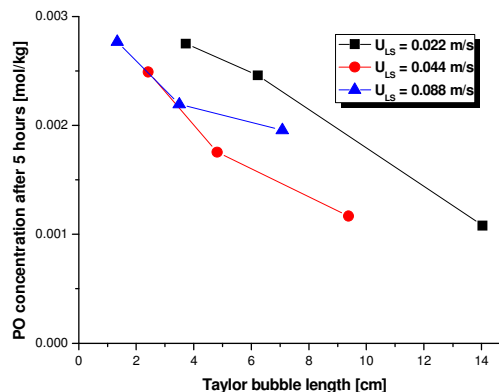
Figure 3 shows the photographs of different length Taylor bubbles for co-current upward flow in the CTF reactor. All the Taylor gas bubbles had spherical/elliptical nose and similar overall shape but their length was changed with gas and liquid flow rates.



**Figure 3.** Photographs of Taylor gas bubbles at various gas superficial velocities ( $U_{GS}$ ) and liquid superficial velocities ( $U_{LS}$ ) in the CTF reactor.

The performance of TS-1 catalyst for the epoxidation of propylene was tested in the CTF reactor (Figure 4). In

principle, propylene oxide production rate increased with decreasing Taylor gas bubble length. However, it was also affected by Taylor bubble rise velocity, liquid slug length and slug rise velocity at the same time.



**Figure 4.** Effect of Taylor bubble length on the propylene oxide concentration after 5-hour operation in the CTF reactor.

In addition, the effects of hydrodynamics with variations of liquid and gas flow rates on the productivity of propylene oxide in the monolith reactor were investigated. The monolith reactor also exhibited comparable performances but it was somewhat difficult to deal with catalyst coating in the monolith reactor.

## Conclusions

The epoxidation of propylene to propylene oxide with hydrogen peroxide was carried out over immobilized TS-1 catalyst in the monolith and CTF reactor. Various analytical methods confirmed that the TS-1 catalyst was prepared with a comparable activity reported in the literature<sup>1</sup>. The effect of hydrodynamics on the propylene epoxidation in the monolith and CTF reactor was investigated in detail. The two types of reactor showed somewhat different characteristics of hydrodynamics but comparable productivity and selectivity to propylene oxide over 95% under mild conditions (40 °C, 1 atm). However, the CTF reactor has the advantage of easy and cost-effective control of catalyst.

## Acknowledgement

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## References

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