



# Another Critical Look at Three-Phase Catalysis

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## Abstract

Three-phase catalysis, for example, hydrogenation, is a special branch of chemical reactions involving a hydrogen reactant (gas) and a solvent (liquid) in the presence of a metal porous catalyst (solid) to produce a liquid product. Currently, many reactors are being used for three-phase catalysis from packed bed to slurry vessel; the uniqueness for this type of reaction in countless processes is the requirement of transferring gas into liquid, as yet there is not a unified system of quantifying and comparing reactor performances. This article reviews current methodologies in carrying out such heterogeneous catalysis in different reactors and focuses on how to enhance reactor performance from gas transfer perspectives. This article also suggests that the mass transfer rate over energy dissipation may represent a fairer method for comparison of reactor performance accounting for different types/designs of reactors and catalyst structures as well as operating conditions.

## Keywords

- ▶ phase catalysis
- ▶ reactor performances
- ▶ mass transfer coefficient
- ▶ energy dissipation rate

## Introduction

Three-phase catalysis is one of the key synthesis steps in petrochemical, chemical, cosmetic, pharmaceutical, and food industries, for instance, converting alkenes and aromatics to saturated alkanes (paraffins) and cycloalkanes (naphthenes) in petrochemical industry to reduce toxicity and reactivity; converting unsaturated alkenes, alkynes, aldehydes, imines, and nitriles to alcohols and amines in chemical/pharmaceutical industries to produce purer products; processing vegetable oils into solid or semisolid fats, e.g., margarine, in food industry to add values, aid transportation, and lengthen products shelf times. To initiate any three-phase catalysis, e.g., hydrogenation, it requires both gas ( $H_2$ ) and liquid reactants diffusing into the surfaces of solid catalysts, as illustrated in ▶ **Fig. 1** focusing on one single spherical catalyst particle of a radius  $R$ . The reaction mechanisms of many commercial heterogeneous catalyses, although successfully operated, are still a matter of debate and controversy, and generally consist of seven well-known steps<sup>1–4</sup>: (1) mass transfer of gas, e.g.,  $H_2$ , from bulk into the liquid phase and then to the external surface of a catalyst particle; (2) diffusion of *both* the dissolved  $H_2$  (gas) and organic (liquid) compounds through the pores of the catalyst to the

internal catalytic surface; (3) adsorption of the gas and organic species onto the inner surfaces of the catalyst; (4) reaction on the inner surfaces of the catalyst; (5) desorption of the products from the surfaces; (6) diffusion of the products from the interior through the pores to the external surfaces; (7) mass transfer of the products from the external surfaces to the bulk fluid.

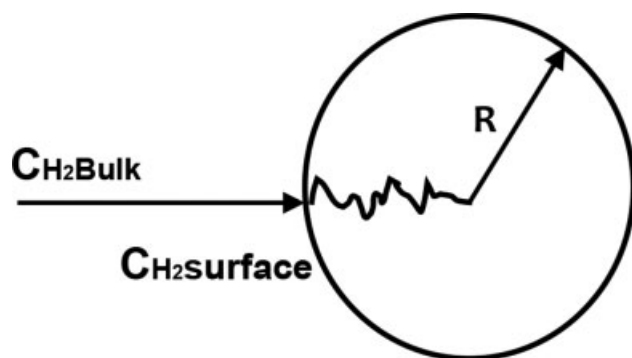
The steps (3) to (5) are regarded as the catalytic reaction, while the rest of the steps are associated with mass transfer. These reaction and transport processes occur concurrently in such catalyses.<sup>5,6</sup> The degree of resistance in mass transfer increases significantly from liquid–solid systems to gas–solid and further to gas–liquid–solid catalysis. For liquid–solid catalysis, solid catalyst particles are readily wetted by the surrounding liquid, the dispersion of liquid into the pores of the catalyst is a relatively straightforward process. For three-phase catalysis, on the other hand, the gas reactant must “travel” through the gas–gas interface, gas–liquid interface, and then the gas–solid interface before reaching the outer surfaces of the catalyst. In each of the boundary crossings, the concentration of gas is reduced, leading to the arriving concentration of  $H_2$  at the surfaces of solid catalysts ( $C_{H_2, \text{surface}}$ ) being significantly smaller than that of the input gas ( $C_{H_2, \text{Bulk}}$ ),

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**Fig. 1** Reaction mechanism of hydrogenation involving spherical solid catalyst particles.

as illustrated in ►Fig. 2. Since the reaction rate is proportional to  $C_{H_2\text{surface}}$ , three-phase catalysis is often severely restricted by the limitations in the aforementioned mass transfers, affecting the reaction rate, selectivity, productivity, and prolonging reaction times.<sup>7</sup> From chemical engineering perspectives, how to minimize the difference of  $(C_{H_2\text{Bulk}} - C_{H_2\text{surface}})$  is the key in ensuring effective heterogeneous catalysis.

In 1924, Murray Raney, an American engineer, discovered that by fusion of a 50:50 Ni/Al alloy and then leaching out the Al using aqueous NaOH, a nickel sponge was obtained, which was much more active than other commercial catalysts.<sup>8,9</sup> The Raney nickel catalysts are often large in diameter, e.g., 1 to 10 mm, it is easier to imagine mass transfer of gas species into these catalyst spheres. For modern metal catalysts, their sizes are usually range between micro and nanometers, thus, it becomes more difficult to envisage the aforementioned mass transfer processes taking place *physically* within these minute catalyst particles, but the truth is that all occur at the molecular level. Visible bubbles with a diameter from 100s to 1000s micrometers are at the macroscopic level, similar to catalyst particles, mixing, catalyst surface structure, and physical interactions that affect the outcome of catalytic

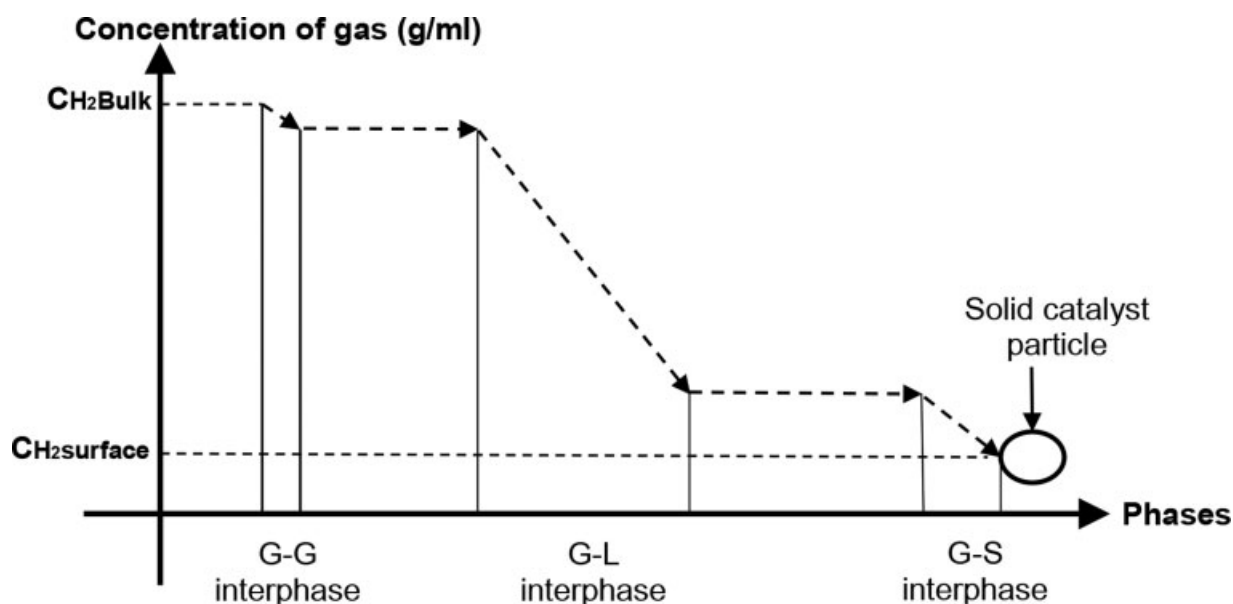
**Table 1** Dissolved hydrogen in water at 298 K<sup>14</sup>

$P$ (bar)	g $H_2$ dissolved/g $H_2O$	mol $H_2$ dissolved/mol $H_2O$
1	$1.54 \times 10^{-6}$	$1.39 \times 10^{-5}$
2	$3.09 \times 10^{-6}$	$2.78 \times 10^{-5}$
3	$4.62 \times 10^{-6}$	$4.16 \times 10^{-5}$
5	$7.72 \times 10^{-6}$	$6.94 \times 10^{-5}$

reactions. In order for any reactive gas to arrive at the surfaces of catalysts that are surrounded by liquid, macroscopic bubbles must be broken into smaller and minute ones, the latter would reach equilibrium with liquid and become dissolved at the microscopic/nanoscale level, and it is thus the dissolved gas in liquid, not visible bubbles, that holds the key for mass transfers in multiphase catalysis. From the chemical engineering viewpoint, how to increase the dissolved gas species in liquid is an effective measure of how efficient various reactors are for carrying out heterogeneous catalyses.

Note that improving the structure of catalysts, e.g., monoliths, would enhance the areas of catalyst surfaces, and the subsequent reaction efficiency.<sup>10-12</sup> Reactors with monolith catalyst packing are hydrodynamically superior to existing industrial reactors<sup>13</sup>; however, the physical transport process of gas dissolution into liquid must be driven by fluid mechanic forces, in combination with reactor designs/additions and catalyst structure.

Majority of three-phase catalysis is operated at elevated pressures, as pressure increases the solubility of gas into liquid. ►Table 1 displays such an effect where four times more hydrogen is dissolved in water at 5 bar compared with that at 1 bar.<sup>14</sup> However, it should be noted that the amount of hydrogen dissolved in water is measured in terms of micrograms per gram of water, i.e., parts per million. As a result, the effect of increasing pressure as a means of increasing



**Fig. 2** Mass transfer through phase boundaries in three-phase catalysis.

$C_{H_2}$  surface is rather small in three-phase catalysis; subsequently how to improve mass transfer of gas into liquid is THE fundamental chemical engineering parameter in most, if not all, of heterogeneous catalyses in industries. This is the focus of this review article.

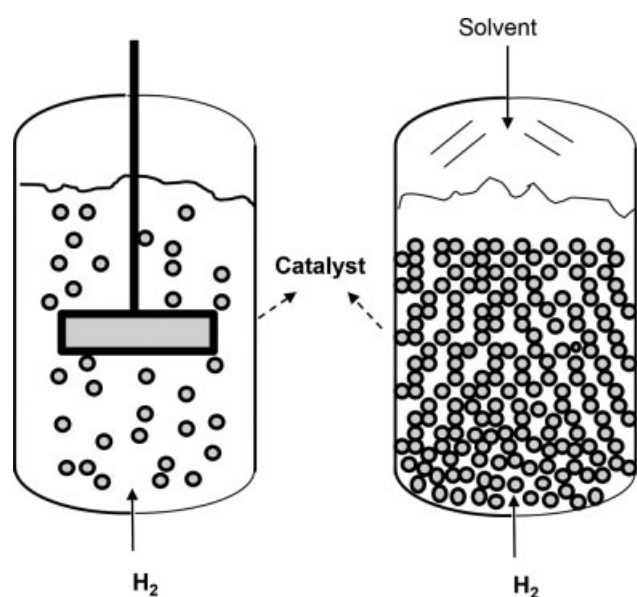
## Reactors for Three-Phase Catalysis

Mixing and reactor designs play a critical role in heterogeneous catalysis. The commonly used reactors in industrial multi-phase catalyses are generally divided into two categories: suspended or fixed bed reactors. The former is associated with mobile catalyst particles that are suspended in reactors, also referred as slurry reactors, including bubble columns (suitable only for very small catalyst particles), agitated tanks, and three-phase fluidized beds. The fixed bed reactors involve stationary catalysts, including packed bed, trickle bed, and pulsed bed reactors. ▶ Fig. 3 illustrates the basic principles of the two types of reactor set-ups.

In the following sections, each type of the reactors is assessed in terms of the common yardsticks for transferring gas into liquid, such as (1) mechanisms of breaking and maintaining gas bubble sizes; (2) mass transfer coefficient ( $k_L a$ ) per energy dissipation rate ( $W m^{-3}$ ), as mass transfer coefficient alone cannot provide any meaningful comparison of reactor performances; and (3) where the dissolved gas concentration is measured in each type of contactors is critical, as it differentiates local from overall mass transfer rate.

### Packed Bed Reactors

In a packed bed reactor (PBR), stationary solid catalyst particles are either packed or shelved in the reactor. The main choice for design and operation with this type of reactors is the direction of flow for both gas and liquid phases, e.g., co- or countercurrent.



**Fig. 3** Reactor type for three-phase catalysis: slurry bed (left) and packed bed (right).

### Trickle Bed Reactors

A trickle bed reactor is a variant of packed bed where the liquid solvent is showered down from the top, thus increasing the surface area of the liquid, and gas can go either co-currently or counter-currently with the liquid. Hydrodynamics of trickle bed reactors were studied<sup>15,16</sup> using transport modeling,<sup>17</sup> computational fluid dynamics (CFD) modeling,<sup>18</sup> electrical resistance tomography,<sup>19</sup> as well as by high pressures.<sup>20</sup> Due to the reliability of their operation, trickle bed reactors have won a great use in oil industry, and also found applications in  $SO_2$  oxidation,<sup>21</sup> glucose hydrogenation over ruthenium catalyst,<sup>22</sup> hydro-treating atmospheric residue,<sup>23</sup> hydro-purification,<sup>24</sup> catalytic hydro-treatment of vegetable oils,<sup>25</sup> fuel production via Fischer-Tropsch synthesis,<sup>26</sup> hydrogen production by aqueous-phase reforming of xylitol,<sup>27</sup> hydrogenolysis,<sup>28,29</sup> continuous thermal oxidation of alkenes with nitrous oxide,<sup>30</sup> liquid-phase selective hydrogenation of methylacetylene and propadiene,<sup>31</sup> hydrogen peroxide,<sup>32</sup> as well as continuous operation.<sup>33</sup>

There are two possible mechanisms in trickle bed reactors for breaking down bubble sizes and initiating dissolution of gas into liquid: (1) the interactions of liquid and gas flows, (2) through the tortoise routes that are formed by the packed catalysts, the denser the solid particles, the smaller the diameters of bubbles so formed. There are however no facilities of maintaining bubble sizes in such reactors, and once gas has passed the dissolution zones, bubble coalescence occurs readily. The determinations of gas-liquid or liquid-solid mass transfer coefficients in packed beds were performed in systems involving either nonreaction schemes, for example, absorption/desorption of  $O_2$  or  $CO_2$  in water,<sup>34-37</sup> or actual reactions.<sup>38,39</sup> Iliuta et al compiled mass transfer data from more than 3,200 experiments in 52 gas-liquid systems, with over 60 packing sizes/geometries and 17 column diameters<sup>40</sup>; however, there were neither information on where dissolved gas concentration was measured nor on energy dissipation rates in trickle bed reactors.

### Pulsed Trickle Bed Reactor

In pulsed trickle bed reactors (a variant of packed bed), either gas or liquid flow is subjected to a pulse motion<sup>41</sup> that can be generated using hydropneumatic, self-propelled, or elastic membranes.<sup>42</sup> The pulses cause the transition to bubble flow, while the parts of the bed in between pulses reside at the transition to trickle flow. Properties of pulsing flow were studied,<sup>43-45</sup> including hydrodynamics of trickling to pulsing flow transition<sup>46</sup> and bubbly to pulsing flow regimes,<sup>47-49</sup> since the majority of industrial processes operate at or near the transition from trickling to pulsing flow. The operation of trickle bed reactors at elevated temperatures was also reported.<sup>50</sup>

The same mechanisms for breaking bubbles and initiating dissolution of gas into liquid in trickle bed reactors apply to pulsed trickle beds, with an additional feature of pulsing. The purpose of pulsing liquid is effectively to “hold” bubbles for a fraction of time (e.g., 0.5 Hz), this enhances overall heat and mass transport while reducing axial dispersion. Keeping all other parameters constant, reactor operation in the presence of pulses resulted in up to 30% increase in reaction rate,<sup>44</sup> 15% increase in styrene concentration,<sup>51</sup> and 45% improvement in

styrene selectivity in hydrogenation of phenylacetylene over Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> catalyst compared with that without. Some mass transfer data in pulsed trickle bed reactors were reported,<sup>52–54</sup> once again, no information was given on both where dissolved gas concentration was measured and energy dissipation rate.

### Slurry Bed Reactors

In slurry bed reactors, solid catalyst particles are suspended in a liquid solution. There are many means of creating slurry suspension, e.g., mechanic, pneumatic, oscillatory, and combinations of flow directions of gas and liquid phases.

### Stirred Tank Reactors

Stirred tank reactors (STRs) are the widely used industrial workhorse for chemical reactions, including heterogeneous catalysis,<sup>55–57</sup> catalytic cracking,<sup>58</sup> catalytic foaming,<sup>59</sup> and Fischer–Tropsch synthesis,<sup>60</sup> just to name but a few. In terms of operation, solvent and catalyst particles are added to the tank, and impeller or impellers are used for mixing and suspending mobile catalyst solids. A sparger is implemented for introducing gas, either from the top or the bottom, with or without recycling of the product stream back to the reactor.

The mechanisms of breaking gas bubbles in STR usually include the rotating motions of impeller(s) and the interactions of fluid with the presence of wall baffles. In laboratory-scale STRs, mixing is regarded as uniform, thus, the dissolved gas concentration measured anywhere in the reactor can be used for the determination of the overall mass transfer coefficient. With the increase of reactor volume, however, a gradient in bubble sizes is often generated, in which the smaller bubbles appear near the tip of the impeller, and the larger bubbles appear elsewhere. The balance between bubble breaking and coalescence is determined by the hydrodynamic force of impeller rotation in the presence of mixing aids, e.g., wall baffles, thus the designs of impeller and turbulence promoter<sup>61</sup> are critical in providing more uniform bubble size, and in turn the higher mass transfer rate,<sup>62,63</sup> e.g., mass transfer using either a dual impeller<sup>64</sup> or radial–axial impeller combination<sup>65</sup> was 15 to 35% higher in comparison to a single impeller. Mass transfer of a gas into a liquid has intensively been studied in STR, such as air in water,<sup>66–68</sup> H<sub>2</sub> in water,<sup>69–72</sup> O<sub>2</sub> in liquid hydrocarbons,<sup>73</sup> and in *n*-octacosane processes,<sup>74</sup> in fermentation vessels,<sup>75</sup> in gas–liquid–solid systems,<sup>76–78</sup> and in scale-up STRs.<sup>79,80</sup> Modeling of gas–liquid mass transfer<sup>81</sup> was performed using an Euler–Lagrange approach<sup>82</sup> and CFD.<sup>83</sup> Additionally, liquid–solid mass transfer<sup>84,85</sup> and solid–liquid mass transfer<sup>86</sup> were also reported in STR.

Energy dissipation rates in STR takes the form of Equation 1:

$$\frac{P}{V} = \frac{P_o \rho N^3 D_s^5}{\pi D_v^2 L_h / 4} \quad (\text{Equation 1})$$

Where  $P/V$  is the energy dissipation rate per volume (W m<sup>-3</sup>),  $P_o$  the power number for the impeller depending on the impeller type and dimensions,  $N$  the rotational speed of

the stirrer (s<sup>-1</sup>),  $L_h$  and  $\rho$  are the height (m) and density (kg m<sup>-3</sup>) of the liquid in the reactor, and  $D_s$  and  $D_v$  the diameters of the agitator and the vessel (m), respectively. This is the energy that is received and utilized by the reaction media to achieve the measured mass transfer rate in the said reactor.<sup>87–94</sup> Available mass transfer and energy dissipation data are used for comparison in a later section.

### Bubble Column

A bubble column by definition consists of a column filled with liquid and solid catalyst particles with gas being sparged from the bottom. The hydrodynamics and heat/mass transfer of slurry bubble columns were reported.<sup>68,95–106</sup> In addition, slurry bubble columns were also used in Fischer–Tropsch synthesis,<sup>107,108</sup> CO methanation over La-promoted Ni/Al<sub>2</sub>O<sub>3</sub> catalyst<sup>109</sup> and green fuel production via hydrocracking of vegetable oil.<sup>110</sup> It should however be noted that the bubble column itself has no physical mechanisms of breaking bubbles as well as preventing bubbles from coalescing, and various restrictions, delays, and recycles are implemented to aid the physical process of bubble breaking, e.g., down pipes, loop, jet, and pulsing.

Jet flow/loop reactors are the variants of bubble columns; the implementation of jet is to break gas bubbles and the looping is to increase the time for dissolution of gas. In jet loop reactors, an external pump is used to circulate liquid (along with the catalyst and often some gas) through an ejector type nozzle, as such gas-inducing nozzle is a critical design component of jet loop reactors,<sup>111,112</sup> Fundamental scientific studies of flow in and out of a nozzle were reported in terms of mixing,<sup>113,114</sup> hydrodynamics,<sup>115–117</sup> bubble size distribution,<sup>118</sup> mass transfer,<sup>119,120</sup> as well as reaction kinetics.<sup>121</sup>

The jet in jet loop reactor sends liquid plume downwards, causing dispersion and entrainment of minute bubbles, this is where the maximum dissolution of gas into liquid and excellent gas–liquid mass transfer take place. When the jet plume reaches the bottom of the down-flow, the jet stream comes up, leading to bubble coalescence and visible bubbles rising. Effectively the dissolution zone is the length of the jet core. A study by Mandal<sup>114</sup> shows that the core length was 30 to 45 mm in a 1,500-mm tall column of a diameter of 52 mm, i.e., approximately 3% of the full length, although the core length can reach 50% of the full reactor length in industrial-scale operations. Dissolved gas concentrations were mainly measured at the tail end of the core, where bubble sizes were generally from 1 to 6 mm.<sup>122,123</sup> The energy requirement for forming such a jet is usually high, e.g., 1.2 to 1.5 m<sup>3</sup>/h for liquid flow and 0.25 to 1 m<sup>3</sup>/h for gas flow in a jet loop reactor of 200 mm diameter and 700 mm tall<sup>118</sup>; however, neither equations nor data were given on energy dissipation rates due to the combination of wide-ranging fluid mechanical zones with nonstandardized designs of jet promoters for this type of reactors.

Jet loop reactors have been used for model reactions such as hydrogenation, chlorination, phosgenation, hydroformylation,<sup>124,125</sup> as well as for processes of CO<sub>2</sub> absorption,<sup>126</sup> anaerobic codigestion of olive mill wastewater and liquid poultry manure,<sup>127</sup> treatment of slaughterhouse wastewater,<sup>128</sup> imidacloprid preparation,<sup>129</sup> and microbial fermentation.<sup>130</sup>

### Fluidized-Bed Catalytic Reactors

Heterogeneous catalysis has also been performed in fluidized-bed reactors where the gas reactant is bubbling through a liquid bed containing a solid catalyst.<sup>131</sup> The desirable features of a fluidized bed reactor can briefly be summarized as its the favorable heat transfer, temperature uniformity, high effectiveness factors, low pressure drop, and ability to add/remove catalysts, and some of the disadvantages are entrainment, attrition, wear, as well as nonuniform residence time distributions, and unpredictability.<sup>131</sup> In reality, the design and scale-up of fluidized-bed reactors rely heavily on experience, mechanistic understanding, and models.<sup>132</sup> There is a void in the literature for the evaluation of energy dissipation rate in fluidized bed reactors.

In summary, the advantages of PBRs are (1) there is lesser demand on particle size of catalyst; (2) it is relatively easy to design this type of reactors; and (3) there is no need for catalyst separation after reaction, reducing unit operation and associated requirement for energy and labor. The major drawback for stationary catalysts is that the required quantity of the catalyst is significantly higher in comparison to mobile catalyst arrangement for the same conversion due to reduced surface areas of the catalysts. In addition, bubble sizes are relatively large, as there is generally lack of means of holding small bubbles, leading to lower mass transfer capability and longer reaction times.

For slurry bed reactors, there are mechanic means of breaking bubbles and achieving gas dissolution, thus increasing mass transfer and reaction rates; mobile catalysts offer significantly higher surface areas, leading to much less catalyst and shorter reaction time to achieve the same conversion in comparison to stationary catalysts. The shortcoming for this type of operation is that a separation of solid catalyst particles is mandatory at the end of reaction, leading to potential loss of catalyst due to attrition. Filtered catalyst particles can be reused for a few times depending on processes.

### Micro Bubbles

Process intensification in multiphase reactors have been used<sup>133–135</sup> with various mechanisms, e.g., continuous flow mixing vessels,<sup>136</sup> vortical ciliary flows,<sup>137</sup> micro-mesh,<sup>138</sup> microbubble generators,<sup>139</sup> microreactors,<sup>140–143</sup> and in scale-up flow reactors.<sup>144</sup> The key feature of these intensified devices is the ability of producing fine and minute bubbles; consistently, some show the capability of maintaining fine bubble sizes, leading to enhanced mass transfer rates. However, currently, there are no publications on energy dissipation rates in these new/novel reactors.

### Oscillatory Baffled Reactors

Oscillatory baffled reactors (OBR) generally consist of a jacketed cylindrical column and a set of orifice baffles. The up and down movement of the baffle set within the column generates intensive eddy current that moves fluids from wall to center, creating equal radial and axial velocity components, which is the essential measure of uniform mixing. The

intensity of mixing in a batch OBR can be controlled by varying either oscillation amplitude or/and frequency when the orifice diameter and baffle spacing are fixed. Oscillatory amplitudes from half to one baffled cell length and oscillation frequency from 1 to 8 Hz can be employed, the latter is much higher than that in pulsed trickle bed reactors. The energy dissipation rate in OBR was developed by assessing inertial and frictional effects of the flow together with pressure drop due to a static head,<sup>145–148</sup> as in Equation 2:

$$\frac{P}{V} = \frac{2}{3} \frac{N_b \rho}{\pi C_D^2} \frac{1 - \alpha^2}{\alpha^2} x_o^3 \omega^3 \quad (\text{W m}^{-3}) \quad (\text{Equation 2})$$

Where  $N_b$  is the number of baffles per unit length in OBR ( $\text{m}^{-1}$ ),  $\alpha$  the ratio of the effective orifice area to the tube area,  $x_o$  the oscillation amplitude (m),  $\omega$  the angular oscillation frequency (radians  $\text{s}^{-1}$ ) and  $C_D$  the orifice discharge coefficient.

When gas is involved, gas bubbles are broken down by the formed vortices; the reciprocal movement of the baffles also holds and maintains bubble sizes. When compared with mass transfer of air in water,<sup>149–151</sup> air in cultures,<sup>152</sup>  $\text{O}_2$  in water,<sup>153,154</sup> ozone in water,<sup>155,156</sup> and  $\text{CO}_2$  in water<sup>157</sup> in STRs or bubble columns,  $k_L a$  values in OBR were much improved, which was attributed to three distinct features: (1) smaller and more uniform bubble sizes due to the mechanism of maintaining bubble sizes; (2) higher gas hold-up, as a foaming layer at the top of the column acts as a “blanket” to prevent minute bubbles from disengaging, and the oscillatory motion drags these bubbles back into the liquid; and (3) significantly prolonged residence times of bubbles due to the reciprocal motion of eddies.

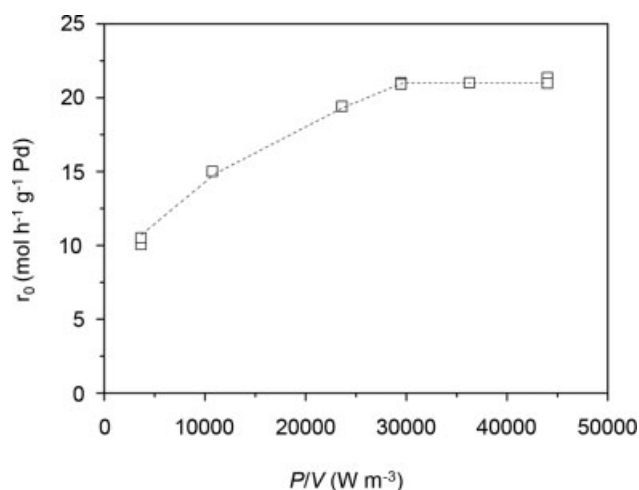
The above key features are manifested in a comparative study of catalytic hydrogenation of 3-butyn-2-ol over Pd/ $\text{Al}_2\text{O}_3$  catalyst to generate 3-buten-2-ol (an intermediate) in both a commercially available stirred tank PARR reactor (PARR in short) and an OBR,<sup>158</sup> where an increase in the initial reaction rate ( $r_0$ ) with the increase of energy dissipation rate (stirring speed) is seen up to  $29,500 \text{ W m}^{-3}$  in the PARR reactor (→Fig. 4), beyond which  $r_0$  is unaffected, indicating that the capacity of mixing in terms of stirring speed in the PARR has reached its ceiling and no longer affects the reaction rate.

Under the same reaction conditions, rising profiles of the initial reaction rate against energy dissipation are still seen for all pressures tested in the OBR (→Fig. 5), indicating that the capacity of mixing is not only significantly larger, but also more energy efficient than the PARR reactor, for example, approximately six times less energy dissipation in the OBR was required to achieve the same reaction rate obtained in the PARR working at the same pressure or approximately three times less energy dissipation if the operating pressure in the OBR was halved.

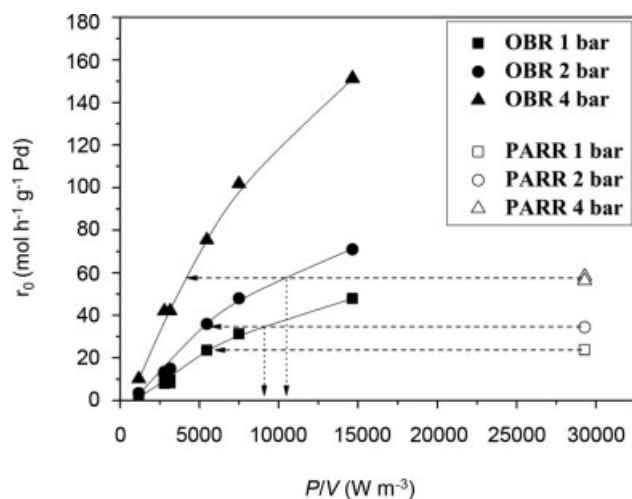
### Comparison

In this article, several reactors for heterogeneous catalysis have been introduced, each involves different physical





**Fig. 4** Effect of energy dissipation ( $P/V$ ) on initial reactor rate ( $r_0$ ) in the stirred tank PARR reactor at 1 bar.<sup>148</sup>



**Fig. 5** Effect of energy density ( $P/V$ ) on initial reaction rate ( $r_0$ ) at different pressures in PARR and OBR. Working conditions: initial molar ratio 3-butyn-2-ol/Pd = 1,360 and temperature = 323 K.<sup>148</sup> OBR, oscillatory baffled reactor.

designs and operating conditions. How could reactor performances be compared and what would be the common basis for such a comparison? Using the 100 m sprint as an example, if you race against Usain Bolt, Usain Bolt will win every time, because Usain Bolt is the fastest sprinter in the world, while you are an amateur runner. We accept the outcome without questioning. If you challenge Usain Bolt with you on a motor bike, you will win the race. This leads to the key question: what is the *common* basis for comparison? From the above example, power consumed by the motor bike is more than that by the human being, and the sprint time divided by the power consumption would provide a level-playing field for comparison of different racing modes; in this case Usain Bolt will win it again. The exact principles are applied here. The basis is the energy dissipation rate ( $W m^{-3}$ ) that is received by the reaction media in a given reactor, which is neither the electric power of the motor that is used for stirring in STRs nor the power of pump that is employed for generating jets in jet loop reactors. Using either mass

transfer coefficient or bubble mean size *alone* does not serve any meaningful purpose of comparison, since higher energy dissipation experienced by the reaction mixture generally leads to higher mass transfer rates. By dividing mass transfer coefficient over energy dissipation that is consumed by reaction media to generate the measured transfer rate would offer a fairer and better comparison of reactor performances, counting for different designs and operating conditions. This ratio is used in this article.

While there are plentiful research papers on the evaluation of energy dissipation in both STRs and OBRs, no research articles are found for other types of both slurry and packed bed contactors. In some cases, data of mass transfer coefficient or mean bubble sizes or gas hold-up were presented as a function of energy dissipation, but no information was given on how these dissipation rates were derived. Taking a leap of faith, ►Table 2 compiles the ratio of  $k_L a$  over their corresponding energy dissipations as the key indicator (last column). Note that the lowest energy dissipation data were taken for comparison in ►Table 2, and this is due to the unavoidable fact in gas-liquid systems where the percentage increase in mass transfer coefficient is much smaller than that in energy dissipation (mixing), e.g., often 100% increase in energy dissipation leads to 1 to 5% increase in mass transfer rates.

While the ratio of  $k_L a$  over ( $P/V$ ) in ►Table 2 gives the indicative comparison of the capability of delivering gas to liquid mass transfer for different types of gas-liquid contactors, there are three factors to note:

- Each reactor type has means of breaking bubbles; it is however the mechanism of maintaining achieved bubble sizes that are critical to the overall mass transfer rate, as gas bubbles coalesce naturally. Reactors with higher ratios of mass transfer over energy dissipation are generally equipped with better mechanisms of maintaining bubble sizes.
- Most gas-liquid contactors exhibit nonuniform mixing patterns, or have different fluid dynamic zones, where exactly the dissolved gas concentration was measured in a given reactor can have significant impact on the determination of the overall mass transfer rate; unfortunately very few details in this aspect were disclosed in published papers.
- The contribution of static pressure head, e.g.,  $\rho g u_g$ , or  $\rho_L g H_s Q_g$ , or  $\rho g v_g V_L$  in ►Table 2, to the overall energy dissipation is generally very small; the inclusion of such a term makes little difference in energy dissipation data compared with the exclusion of it.

In summary, heterogeneous catalysis covers countless processes involving three phases. Breaking and maintaining minute bubble sizes throughout each reaction are the unique chemical engineering challenge, yet at present, there is not a unified system of quantifying and comparing reactor performances. Profiles of mean bubble size, gas hold-up,  $k_L a$  as function of aeration rates, jet velocity, stirring rate, nozzle diameter, etc. are rather bespoke, and have little value for any meaningful comparison. In this article, the mass transfer rate over energy dissipation is proposed as a fairer method for comparison of

**Table 2** Comparison of mass transfer rates based on energy dissipation

Reference	Reactor type	Energy dissipation ( $P/V$ ) equation	$P/V$ (kW m <sup>-3</sup> )	$k_L a$ (s <sup>-1</sup> )	Ratio $k_L a/(P/V)$
159	Sparged stirred tank	$P/V = (P_0 f^3 D^5 \rho N + \rho g v_g V_L)/V_L$ $P_0$ – power number $f$ – agitation speed $N$ – number of impellor $D$ – diameter of impellor $v_g$ – gas superficial velocity $V_L$ – volume of liquid	0.1	0.02	0.2
160	Sparged stirred tanks	$P/V = \alpha(P_0^2 ND^3/Q)^{0.56} \beta/V_L$ $N$ – agitation speed $D$ – stirrer diameter $Q$ – gas flow rate $P_0$ – energy input in un aerated system $\alpha$ and $\beta$ depend on type and numbers of stirrer	0.15	0.002	0.133
89	Sparged stirred tanks	$P/V = (2\pi NM + \rho_l g H_s Q_g)/V_L$ $N$ – agitation speed $M$ – torque $H_s$ – liquid height $Q_g$ – gas flow rate $V_L$ – liquid volume	1.3	0.04	0.03
161	Sparged stirred tank	$P/V = 2\pi NM/V_L$ $N$ – agitation speed $M$ – torque $V_L$ – liquid volume	0.7	0.019	0.027
162	Jet flow loop		0.8	0.014	0.018
120	Jet loop		2	0.6	0.3
163	Plunging jet bubble column		8	0.2	0.025
111	Gas–liquid ejector		20	1.1	0.055
164	Microbubble nozzle		0.05	0.002	0.04
	Perforated plate		0.005	0.0002	0.04
	Spiral liquid flow		1.3	0.001	0.008
	Venturi		1	0.0003	0.003
	Ejector		7	0.0004	0.00006
	Pressurized dissolution		10	0.001	0.0001
138	Jet array downflow bubble column		1.254	0.139	0.111
149	Oscillatory baffled column	$(P/V) = 2\rho N_b(1 - \alpha^2)x_0^3\omega^3/(3\pi C_D^2\alpha^2)$ $N_b$ – number of baffles per unit length $C_D$ – orifice discharge coefficient $\alpha$ – baffle-free cross-sectional area $x_0$ – oscillation amplitude $\omega$ – angular frequency of oscillation	0.05	0.02	0.4
153	Oscillatory baffled column	$(P/V)_0 = 2\rho N_b(1 - \alpha^2)x_0^3\omega^3/(3\pi C_D^2\alpha^2)$ $N_b$ – number of baffles per unit length $C_D$ – orifice discharge coefficient $\alpha$ – baffle-free cross-sectional area $x_0$ – oscillation amplitude $\omega$ – angular frequency of oscillation $(P/V)_B = \rho g u_g$ $P/V = (P/V)_0 + (P/V)_B$	0.02	0.005	0.25

reactor performance accounting for different types/designs of reactors and catalyst structures as well as operating conditions. Hope more papers on energy dissipation rates for both existing and new-type reactors are emerging to fill the gap.

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#### Conflict of Interest

The authors declare no conflict of interest.

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