

A NEW REACTION PAIR FOR MICROMIXING MEASUREMENTS IN GAS-LIQUID SYSTEMS WITH LARGE VAPOUR GENERATION

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The relevance of micromixing in fast reactions can be characterised by measuring the product distribution of a known test reaction. Although several test reaction systems have been published, none are suitable for boiling or nearly boiling conditions.

This paper presents details of a new test reaction system suitable for sparged aqueous systems at temperatures up to 100°C. This system, which is easy to use, provides the basis for experiments carried out in a standard semi-batch stirred tank reactor agitated with a Chemineer CD-6 hollow-blade impeller. The quantitative characterisation provided by liquid micromixing sheds light on the effect of temperature, and hence vapour pressure, in the operation of gas-liquid reactors.

Key words: micromixing, fast reactions, boiling, hot sparged systems, and vapour generation

INTRODUCTION.

Mixing is a multistage process involving large scale blending (macromixing) and fine scale homogenisation (micromixing) reflecting the scale of scrutiny applied to the operation. Chemical reactions depend on reagents interacting at a molecular level and are directly affected by the rate of micromixing. In many processes the reaction is fast and the time required to bring homogeneity to the molecular level is longer than the time needed to complete the reaction. When more than one possible reaction route is involved, the yields of desired and undesired products will depend on the rate of micromixing. Through optimising the design and operation of reactors to achieve more efficient micromixing, processes can be engineered to improve efficiency, increasing yields and reducing or eliminating subsequent product purification.

The effect of micromixing on fast reactions can be quantified by measuring the product distribution of test reactions. A new reaction system suitable for hot sparged or boiling aqueous systems is proposed in this paper. Experiments have been carried out in a standard semi-batch stirred tank reactor agitated with a Chemineer CD-6 hollow blade impeller. The quantitative characterisation of micromixing in the liquid phase sheds light on the relevance of temperature and vapour pressure in the operation of gas-liquid reactors.

TEST REACTION SYSTEMS IN THE LITERATURE

Many test reaction systems have been developed. These are of three main stoichiometric types, which have been summarised by Fournier et al. (1996):

- (1) single reaction $A + B \rightarrow R$
- (2) consecutive competing reactions $A + B \rightarrow R$ and $R + B \rightarrow S$
- (3) parallel competing reactions $A + B \rightarrow P$ and $A + C \rightarrow Q$

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Single test reactions are suitable for turbulent tubular reactors where reagent conversions as functions of distance and hence also of time can be followed. However, in stirred tank reactors (STRs) the local conversions in the moving fluid cannot usually be tracked as a function of time so the other schemes are more applicable.

CONSECUTIVE COMPETING REACTIONS

The most frequently used consecutive competing reaction system is the diazo coupling between 1-naphthol and diazotized sulphanilic acid. (Bourne et al., 1977).

This system is usually described by two second-order reaction schemes (Bourne et al., 1981),



At 298K and pH=10, $k_1 = 7.3 \times 10^3 \text{ m}^3 \text{ mol}^{-1} \text{ s}^{-1}$ and $k_2 = 3.5 \text{ m}^3 \text{ mol}^{-1} \text{ s}^{-1}$. Therefore, the first reaction is essentially instantaneous, so that if B is limited and the reactants are well-mixed, product R will predominate. Any deviation from perfect mixing allows newly formed R molecules to remain in contact with a high concentration of B, so that the product S is formed before more A can mix with the B molecules. The effect of incomplete mixing on reactions can, therefore, be detected by the distribution of the undesired secondary product S. The selectivity of the reaction is defined as

$$X_S = \frac{2c_S}{c_R + 2c_S} \quad [3]$$

X_S depends on the degree of homogeneity of the mixture of reagents. If completely blended to a molecular level X_S is determined by the reaction kinetics. On the other hand, in a segregated systems, X_S is controlled by molecular diffusion and in the limit a final value of unity is to be expected. For partial segregation, X_S will fall between these two limits.

Although the value of this test system for the investigation of the effect of mixing on reaction has been confirmed in many publications (Middleton et al., 1986; Baldyga and Bourne, 1988, 1989 and 1992, etc.) it has some disadvantages. There is evidence that other side reactions may occur leading to unidentified by-products (Bourne et al., 1990). Moreover the reagents are temperature sensitive.

PARALLEL COMPETING REACTIONS

Parallel competing test reactions usually consist of two irreversible, second-order reactions of the following type:



The first is usually a virtually instantaneous reaction, for example an acid-base neutralization or precipitation ($k_1 \approx 10^7 \text{ m}^3 \text{ mol}^{-1} \text{ s}^{-1}$) with the second a slower but still fast reaction ($k_1 \gg k_2$), e.g. the hydrolysis of an ester.

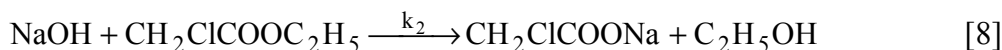
The reaction is carried out by adding a small quantity of A to a pre-mixture of known composition of B and C. The two reactions compete with limited A. The selectivity of product Q is defined by

$$X = \frac{Q}{P + Q} \quad [6]$$

In a perfectly mixed system, X would be determined by the chemical kinetics with the production of very little Q. On the other hand, if the reagents are completely segregated, X is controlled by molecular diffusion. When there is partial segregation on the molecular scale X

depends on chemical factors (reagent concentrations, catalysis, etc.) and the physical conditions (viscosity of solution, feed location, mixing intensity, etc.).

A typical parallel competing test system is the competition between neutralization and alkaline hydrolysis of ester, which was proposed by Bourne and Yu (1991, 1994).



Although this test system has been extensively validated, (e.g. Baldyga et al., 1997; Barresi, 1997, 2000), the system has some limitations. The low solubility of ethyl chloroacetate limits its initial reaction concentration and the consequent limited production of alcohol makes it impossible to use the system for characterising rapid mixing rates such as those in intensive mixers.

The neutralization reactions of Eq.7 can be replaced by precipitation Eq.9 (Baldyga and Bourne, 1990) and Eq.10 (Brucato et al., 2000)

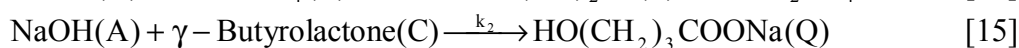
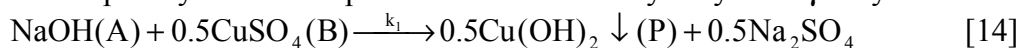


The precipitation of cupric hydroxide is preferred since it is believed to be less complex. Furthermore, the strong coloration of cupric ions in water makes simple colorimetric analysis possible.

Although these and several other test reaction systems have been published, none is suitable for boiling or nearly boiling conditions, either because the reactants or the products are volatile, with low boiling points, or temperature sensitive. It is self-evident that the reagents used for a reaction used to characterise a hot system need to have a much lower vapour pressure than that of the bulk liquid.

NEW PARALLEL COMPETING REACTION PAIR

The test system consists of two parallel reactions competing for limited hydroxyl ions: the precipitation of cupric hydroxide coupled with the alkaline hydrolysis of γ -butyrolactone.



The distribution of undesired product (Q) is defined by Eq. [16],

$$X = \frac{Q}{P + Q} \quad [16]$$

In this system, as in all the micromixing studies referred to here, a small value of the selectivity X corresponds to the most rapid mixing at fine scale, and the highest yield of the usually desired product.

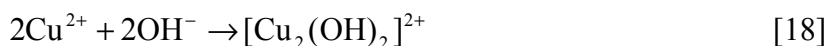
PRECIPITATION OF CUPRIC HYDROXIDE

Reaction mechanism

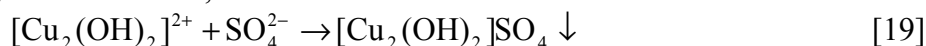
This reaction is a second-order, near instantaneous reaction ($k_1 \approx 10^7 \text{ m}^3 \text{ mol}^{-1} \text{ s}^{-1}$). When hydroxyl ions are in excess compared with copper (II) ions, or the reaction is carried out at high temperature, the following reaction takes place.



On the other hand, when the hydroxyl ion concentration is insufficient, Cu^{2+} complexes with OH^- .



and the following reaction occurs,



The amount of copper (II) ions left in the solution depends on the balance between these reactions. It will be necessary to quantify the possible influence of the formation of this complex on the determination of the residual copper in solution before the micromixing experiments are interpreted..

Measurements of copper ion concentration.

The strong coloration of cupric ions in water makes it possible to measure their concentration by a simple spectrophotometric method. In this work, the concentration of cupric ions in solution was determined from absorption at 800 nm as measured in 4.5 ml cuvettes in a Perkin Elmer Lambda2 UV/VIS spectrometer. At this wavelength the absorbance of cupric ions has a maximum, while the absorbance of all the other species present is zero.

ALKALINE HYDROLYSIS OF γ -BUTYROLACTONE

Physical properties of γ -butyrolactone

γ -butyrolactone ($\text{C}_4\text{H}_6\text{O}_2$) is an unstrained five-membered ring, shown in Fig. 1. At room temperature, it is a colourless liquid and miscible with water. It has a high boiling point (204-205 °C at 760 mmHg).

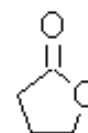


Fig. 1 Molecular structure

The alkaline hydrolysis mechanism.

The hydrolysis of the γ -lactone can be catalyzed by acidic or basic solutions but there is no detectable reaction in water. The acyl oxygen scission (Eq.15) occurs in both the acid- and base-catalyzed reactions. The base-catalyzed hydrolysis of the lactone is a second-order, irreversible fast reaction. The rate constant of the reaction is given by equation [20].

$$k_2 = 2.2 \times 10^8 \exp\left(-\frac{4.728 \times 10^4}{RT}\right) \quad (\text{L} \cdot \text{mol}^{-1} \cdot \text{s}^{-1}) \quad [20]$$

According to Eq. [20], the corresponding rate constants at 25 °C, 50°C, 100°C can be calculated to be 1.15, 5.0, 53.0 $\text{L} \cdot \text{mol}^{-1} \cdot \text{s}^{-1}$, respectively. This reaction rate is comparable with that for the alkaline hydrolysis of ethyl-chloroacetate (for which at 25°C, $k_2 = 30 [\text{L} \cdot \text{mol}^{-1} \cdot \text{s}^{-1}]$) so it could be suitable for micromixing measurements.

PRELIMINARY EXPERIMENTS

CALIBRATION EXPERIMENTS

To measure copper ion concentration, a calibration curve is needed. The linear dependence of absorbance on cupric ion concentration in CuSO_4 aqueous solution is shown in Fig. 2. The curve was prepared by carrying out aliquots of a standard solution. It can be seen that in the range of 0.001-0.04 M, copper ion concentration can be accurately measured.

The validity of the linear relation in the presence of the lactone and/or product (Q) in copper sulphate solution has also been checked. Experimental results (not shown) have confirmed that the linear dependence of solution absorbance on cupric ion concentration is still followed and no

significant variation is detected. Therefore, the interference from the reactant and product of the hydrolysis reaction on the copper ion measurements is excluded.

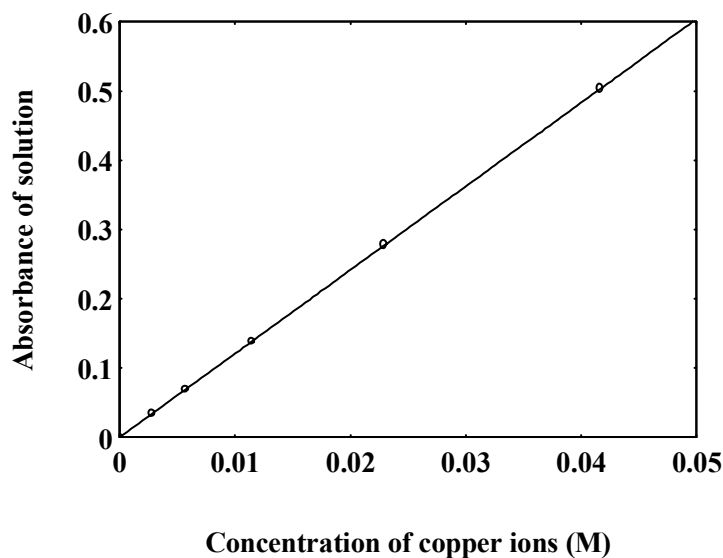


Fig. 2 Calibration curve (wavelength 800nm)

COPPER ION LOSS DUE TO COMPLEX SIDE REACTIONS

Many preliminary experiments according to the following procedure were done in a baffled beaker to quantify the influence of reaction [18] on the copper ion concentration in solution. The limited concentrated sodium hydroxide solution (the solution volume ratio of sodium hydroxide to copper sulphate is 1:50) was slowly fed into turbulent copper sulphate solution agitated by a magnetic or a mechanical stirrer. The concentrations of reagents are shown in Table 1. After the reaction was completed, a sample of the reacted mixture was first centrifuged at 5000 rpm for 20 minutes in order to remove the precipitate. The residual copper concentration in the solution was then determined spectrophotometrically.

The amount of copper precipitated as $[\text{Cu}_2(\text{OH})_2]\text{SO}_4$ can be calculated by the following equation:

$$B_2 = B_0 - B_1 - B_s \quad [21]$$

where B_2 is the amount of copper precipitated as $[\text{Cu}_2(\text{OH})_2]\text{SO}_4$, (mole); B_0 is the total copper in the feed, (mole); B_1 the copper precipitated as $\text{Cu}(\text{OH})_2$, (mole); and B_s are the copper left in the final solution, (mole). B_1 and B_s can be obtained from Eqs.22 and 23, respectively.

$$B_1 = \frac{A_0}{2} \quad [22]$$

$$B_s = C_s \cdot V_T \quad [23]$$

where C_s is the copper ion concentration in the final solution, (M); V_T is the volume of the final solution, (L).

A large number of experiments (some experimental results are listed in Table 1) have confirmed that when limited concentrated sodium hydroxide solution is **slowly** added to copper sulphate solution, the influence of reaction [18] on the copper ion concentration in the final solution cannot be neglected..

Table 1 The effect of side reaction on the final concentration of copper ions

$B_0 * 10^{-2}$ (mol)	$A_0 * 10^{-2}$ (mol)	V_T (L)	$B_1 * 10^{-2}$ (mol)	$(B_0 - B_1) / B_1$ %	$c_s * 10^3$ (M)	$B_2 * 10^{-2}$ (mol)	f %
3.1910	4.4288	1.590	2.2144	44.10	1.7355	0.7007	21.96
3.1929	4.4444	1.590	2.2222	43.70	1.5289	0.7273	22.78
1.4700	1.7988	0.800	0.8994	63.45	3.4050	0.2983	20.29
1.3321	1.7420	0.800	0.8710	52.94	2.5950	0.2535	19.03
1.0554	1.6505	0.780	0.8253	27.89	0.1900	0.2153	20.40
1.7575	2.7988	0.785	1.3994	25.59	0.2810	0.3360	19.12
4.1255	5.0583	1.490	2.5292	63.12	4.9752	0.8556	20.74
4.2039	4.5585	1.495	2.2793	84.44	7.7850	0.7617	18.12
0.6866	0.7088	0.195	0.3544	93.75	9.8430	0.1403	20.43

In Table 1, a term f defined by Eq.[24] is employed to describe the influence of the side reactions on the copper ion concentration in solution.

$$f = \frac{B_2}{B_0} \quad [24]$$

In the experimental range of interest (Cu^{2+} in excess by between 25 and 94%), about 20% of the fed copper ions are precipitated as product of the side reaction [18]. This proportion is sufficiently constant to allow a consistent correction to be applied..

Brucato et al. (2000), however, did not consider this side reaction when they chose the precipitation reaction as one of the two parallel competing reactions used to investigate the effect of mixing on fast reactions. This is probably because in their case, the limited concentrated sodium hydroxide was fed **rapidly** into the vessel. The fast injection may reduce the chance of the formation of $[\text{Cu}_2(\text{OH})_2]\text{SO}_4$. It should be pointed out that in the rapid feed case, it is macro- and meso-mixing rather than the micromixing that determines the distribution of product of complex reactions.

BLANK TEST

In the new test reaction system, the copper ions are always in excess; thus a certain amount of copper will be left in solution after the reaction is complete. This amount can be determined by means of 'blank reaction' runs, which are performed exactly as the usual runs, except that only the precipitation reaction is involved.

If the absorbance of the solution at the end of the blank experiment is Abs_b , and the absorbance before and after the introduction of NaOH in a normal experiment are Abs_1 , and Abs_2 , respectively, the selectivity X in Eq.[16] can be simply expressed as

$$X = \frac{Q}{P + Q} = \frac{\text{Abs}_2 - \text{Abs}_b}{\text{Abs}_1(1 - f) - \text{Abs}_b} \quad [25]$$

where f is the factor of copper ion loss due to side reactions. In this case, $f = 0.2$.

MICROMIXING MEASUREMENTS.

EXPERIMENTAL PROCEDURE

Micromixing experiments have been carried out in an insulated and fully baffled 0.45m diameter tank with four baffles which was described in Zhao et al. (2001). The height of the filled liquid is 0.50m. The reactor was agitated by a Chemineer CD-6 hollow blade impeller with a diameter of 0.176m. The water in the tank could be heated by two immersion heaters located in the tank bottom. A weighed amount of $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ was pre-dissolved and transferred into the stirred tank. The dilute aqueous lactone solution was added and the vessel mixed before being brought to the desired final volume by adding water and again mixing. The resulting concentrations of cupric ions and lactone in solution were 0.015M and 0.02M respectively. Once the contents were completely mixed, a sample (20ml) was withdrawn and the absorbance of the solution, Abs_1 , determined. 1.5L 1M NaOH solution was then added slowly, either into the impeller discharge flow or just below the liquid free surface. The feed time was carefully chosen in order to preclude the effects of macro- and meso-mixing on the product distribution as is discussed in the next section. Then 70ml water was used to flush the funnel. After the reaction was complete, a sample of the solution was centrifuged at 5000 rpm for 20 minutes. Then the absorbance of the solution, Abs_2 , was measured. Finally, enough NaOH solution was added to the tank to convert the lactone into the less physiologically harmful sodium salt.

DETERMINATION OF INJECTION TIMES

In order to determine the effect of micromixing on the selectivity of a complex mixing-sensitive reaction system, the experiments must be operated in only the micromixing-controlled regime and the effects of macro- and mesomixing have to be excluded. If the injection rate of the concentrated reagent solution is high, the plume of feed is not well dispersed so that the macro- and meso-scale concentration gradients can be formed. This increased segregation leads to the production of more by-products. On the other hand, when the injection rate is infinitely slow, macro- and meso-mixing effects are eliminated and the selectivity of the by-product of fast reactions is only micromixing relevant. When the injection time is longer than a critical injection time t_c , the selectivity X reaches a constant value. This means that macro- and mesomixing do not play a role during injection. The critical injection time depends on local power dissipation, reagent concentrations and other operating conditions, thus each experimental run may have a different t_c . We can find the longest t_c under the condition where the weakest micromixing is performed, for example, when the concentrated reagent is injected at the free surface of the liquid and the concentrations of the reagents in the fast reaction are high. Micromixing experiments should be carried out with an injection time longer than the longest t_c .

In this work, when the impeller was operated at the lowest speed and the NaOH solution was injected just below the liquid free surface, the selectivities obtained with the injection time of 30 and 45 minutes are within 3% of each other. We chose an injection time of 45 minutes for safety.

EXPERIMENTAL RESULTS AND DISCUSSION

HOT UNGASSED SYSTEMS

Because the rate constant of the alkaline hydrolysis reaction is relatively small at 25°C, the reaction is not fast enough at ambient temperature to quantify the micromixing. The test reaction pair can thus only be used in hot systems. The measured by-product selectivity with the change of the average shaft power dissipation in hot ungasged systems (89°C) is shown in Fig. 3. The solid line is the simulated data using the E-model (Baladyga et al., 1997). Very good agreement is obtained between the experimental and modelling data. This evidence validates this reaction pair as a test probe for micromixing.

BOILING SYSTEMS

At a given mechanical energy input the by-product selectivity in a boiling system (Fig. 4) is much lower than in hot ungasged conditions. With the increase of temperature, liquid viscosity decreases. As a result, the engulfment rate E increases and this can increase the intensity of mass exchange between the reaction zone and its surroundings. The segregation between reactants is reduced and fewer by-products are produced. The reduction of liquid viscosity between 89°C and 100 °C is 11.4%; As predicted by the E-model, the increase can lead to a 4% decrease of the selectivity of by-product at the average energy dissipation rate 0.07W/kg. Therefore, the change in liquid viscosity is not sufficient to fully account for the considerable improvement of micromixing in boiling conditions.

In seeking the reason for the improvement of micromixing in a boiling liquid, the dynamics of vapour bubbles should be considered. It has been shown in previous chapters that the rapid growth of vapour bubbles can input considerable kinetic energy to the liquid. The violent expansion and collapse of these vapour bubbles can generate local turbulence which may help in decreasing the segregation of reactants in solution and lead to the reduction of the formation of undesired by-products. The mechanism involved in this phenomenon is very complicated and much more work needs to be done before a general conclusion can be drawn.

HOT SPARGED SYSTEMS

Quite scattered data shown in Fig. 5 were obtained when air was sparged into the reactive system, especially when the average power dissipation is low. The suspicion that the small amount of CO₂ present in the sparged gas might have affected the by-product selectivity can be excluded since there was little improvement in the data scatter when N₂ was used instead of air. A similar situation was found by Lin and Lee (1997) with the scatter of data for the by-product selectivity in gas-liquid systems greater than that in single phase systems. In their work, the micromixing index was determined by the measurement of the iodine by-product of their chosen reaction. Some loss of I₂ due to gas stripping could be one of the key factors leading to in the scatter of their data. In the present work, however, this problem does not arise since the reactants and products of the fast reaction have high boiling points and are not easily evaporated.

Generally speaking, the effect of aeration on micromixing is not significant, especially in conditions of high power dissipation (Fig. 5). Although the bulk motion induced by gas bubbles can increase the rate of macromixing, it has little effect on liquid micromixing. This is probably due to the turbulent length scales generated by the motion of gas bubbles being larger than the micromixing length scale in the reactor. The turbulent length scale caused by bubbles depends on bubble size. In general the bubble size is usually too big to affect micromixing. Lin and Lee (1997) found that aeration could improve or retard liquid micromixing depending on impeller speed, gas flow rate and feed location. They assumed the tiny bubbles generated in the region near the impeller tip could influence micromixing.

Here it is worthwhile again stating that the situation in a hot sparged system is different from that in a boiling system. In boiling systems, the rapid expansion from nuclei to small vapour bubbles may well contribute to liquid micromixing. Furthermore vapour bubbles are easily collapsed by local temperature inhomogeneity and this collapse of bubbles may generate local turbulence and also increase micromixing. In a hot sparged system, the bubble sizes do not fluctuate so violently and in any case do not collapse completely. The contribution of these gas bubbles to micromixing is very limited.

FEED LOCATION.

It is well known that the feed location of the limiting reactant in a complex competing reaction system has a large effect on selectivity. The results shown in Fig. 6 confirm this. The solid symbols represent the data obtained when the concentrated NaOH solution was added just below the free surface of the liquid while the open symbols are the results with the feed located in the impeller discharge flow. Compared with feed into the impeller discharge, surface feed apparently gives much higher selectivity, i.e. poorer primary product yield, whether in hot ungasged systems or in boiling conditions. This can be explained by the widely varying energy dissipation rates throughout a stirred tank reactor with the local energy dissipation rates near the liquid surface much smaller than those in the impeller discharge. This leads to the segregation between reactants being greater offering a larger chance for the side reaction to occur. The difference in selectivity between hot ungasged systems and boiling conditions is reduced when fluid is fed just below the liquid surface. Compared with the much improved macromixing in boiling systems when a tracer was injected at the liquid free surface, (Zhao et al., 2001) the improvement in liquid micromixing is less significant. This result implies that, as usual, the optimal operating strategy will depend on the process objective. With a truly boiling liquid, addition at the free surface provides faster bulk blending but slower micromixing. The nature and complexity of the reacting system will be decisive.

CONCLUSIONS

This paper primarily focuses on looking at the effect of boiling and the presence of inert gas bubbles on liquid micromixing in a stirred tank vessel. A new test reaction system, suitable for micromixing investigation in boiling and hot sparged systems, has been developed. The system consists of two parallel competing reactions - the precipitation of cupric hydroxide and the alkaline hydrolysis of γ -butyrolactone. Experimental results have demonstrated that this reaction pair is suitable for the determination of liquid phase micromixing in hot sparged and boiling aqueous systems while being straightforward in use.

Micromixing is apparently improved in boiling reactors. This may be attributed to the decrease of liquid viscosity and the contribution of rapid expansion and violent collapse of vapour bubbles, but further work is needed before general conclusions can be drawn. In contrast gas sparging into a hot liquid, when significant vapour generation is also occurring, has little effect on the rate of micromixing. This is probably because of the much less violent bubble growth or collapse in hot sparged, as opposed to boiling liquids. Feed location has a significant effect on the product distribution in both hot ungasged and boiling systems which is consistent with other published results made in ambient temperature systems.

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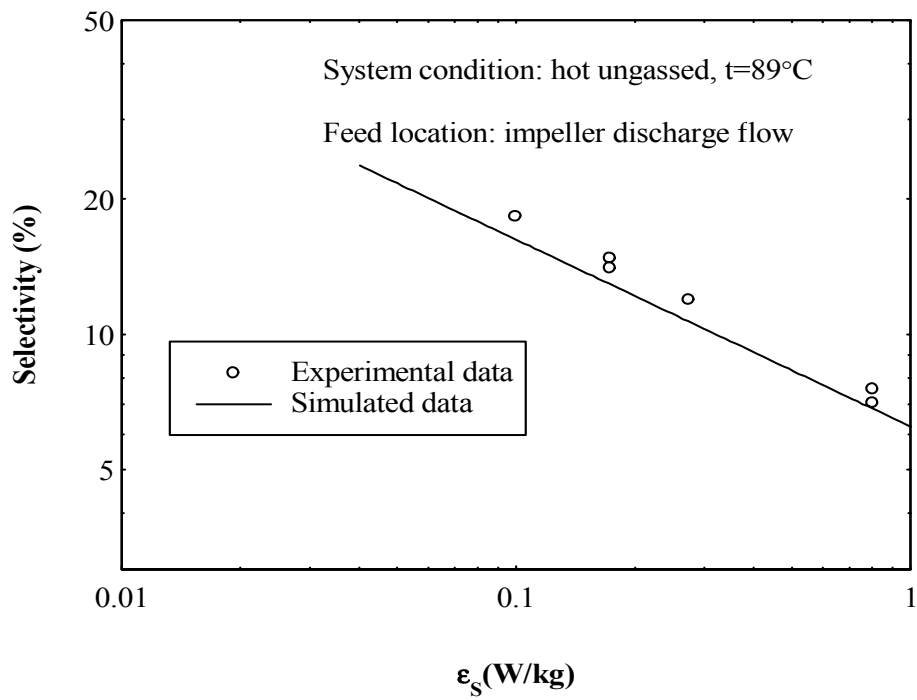


Fig. 3 Comparison of experimental and engulfment modelled data in hot ungasged systems

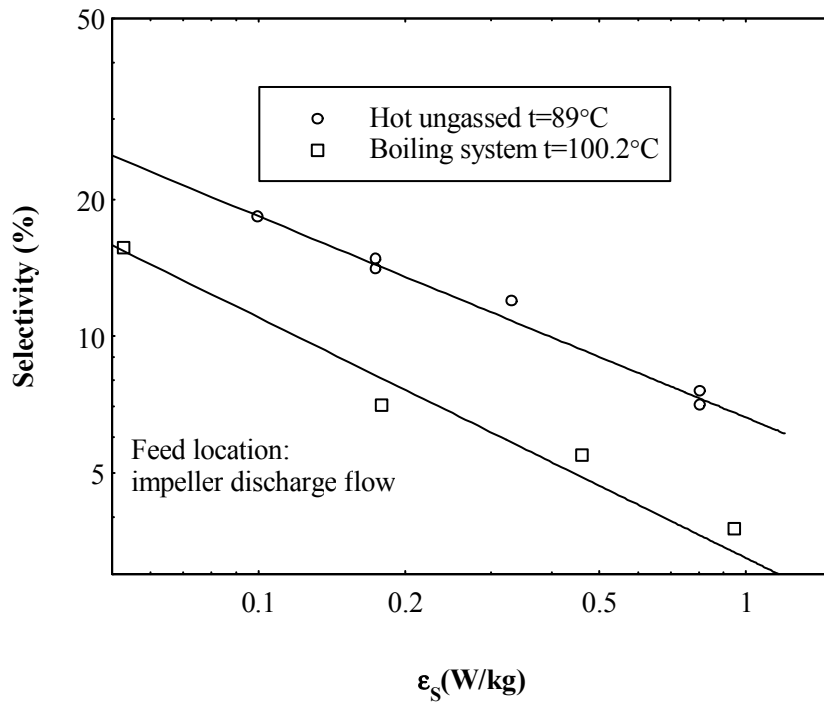


Fig. 4 Micromixing in hot and boiling liquid

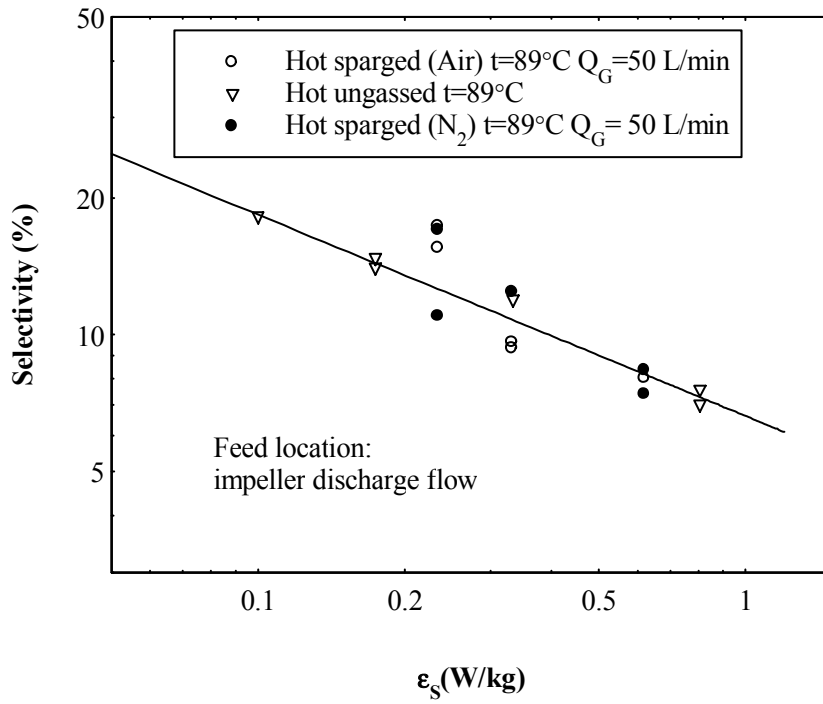


Fig. 5 Micromixing in hot ungasged and sparged systems

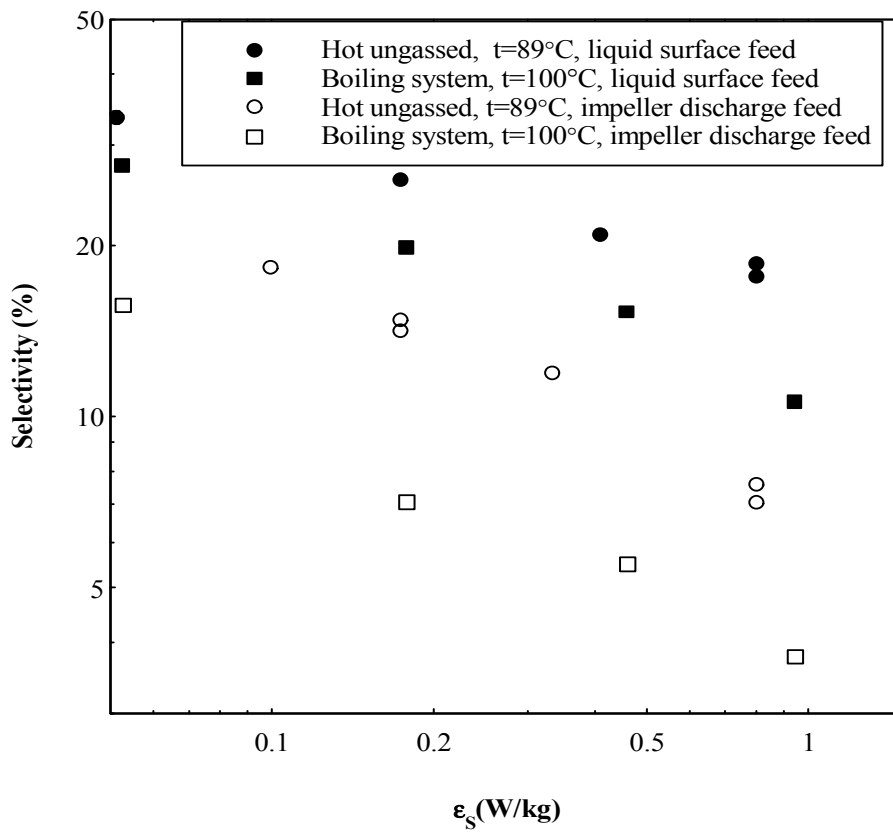


Fig. 6 The effect of feed location on micromixing