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Gas-liquid(-liquid) slug flow in capillary reactors with intermediate gas feed via electrolysis

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1. Introduction

Multiphase contactors in the chemical industry often suffer from inadequate and poorly defined interfacial areas to the detriment of mass transfer. The use of slug flows in capillary reactors offers a solution to such deficiencies. Capillary reactors have already been shown to exhibit considerable benefits in terms of mass and heat transfer, as a consequence of their high surface to volume ratios, together with the internal Taylor circulation flows and regular slug structures. Over the last few decades, the hydrodynamics of two-phase liquid-liquid and gas-liquid slug flows have been thoroughly investigated, while applications, such as extraction and multiphase reaction, have received less attention. Several challenges remain in the application of such technologies. For instance, in the gas consuming reaction $A_{liq}+B_{gas}\rightarrow C_{liq}$ the gas slugs shrink and may even disappear, limiting the conversions and throughputs of capillary reactor systems. [1-5]

To overcome such shortcomings, an intermediate gas feed could be employed, which must nevertheless be implemented so as not to perturb the underlying slug flow. In order to maintain the well-defined slug flow characteristics, it is necessary to introduce the gas rapidly and precisely in small aliquots of < 10 µL. A miniaturised electrolysis cell was thus devised and appropriate operating conditions for generating and sustaining regular slug flow established, followed by the implementation of an intermediate gas feed via the electrolysis cell.

2. Methods

The electrolysis cell is fabricated from PMMA with nickel electrodes. The inner cell capacity is only around 1 mL, so as to eliminate unwanted gas compression phenomena, which might otherwise lead to irregular gas feed rates. For the initial hydrodynamic studies, an aqueous sodium hydroxide electrolysis to a mixture of hydrogen and oxygen gas was used, which was then introduced into the capillary slug flow via a T-junction. The power supply permits one to manipulate both the amperage and the duration of the current pulses.

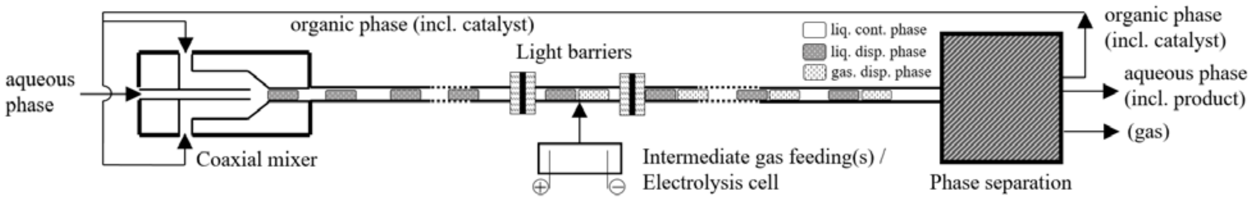

Figure 1: Experimental set-up for an intermediate gas feed for gas consuming reaction.

For the intermediate gas supply, 1-hexanol (VWR 99%) as the continuous phase and ultrapure water as the dispersed phase were first mixed in a proprietary coaxial slug generator. The intermediate gas feed comprised the miniaturised electrolysis cell and two photoelectric sensors, which monitor the upstream and downstream slug flow and synchronise the gas feed by regulating the electrolysis parameters (Figure 1).

3. Results and discussion

To start with, the slug generation by the electrolysis cell was assessed. The slug lengths formed were measured for different amperages and pulsation times and compared to the theoretical values from Faraday's law.

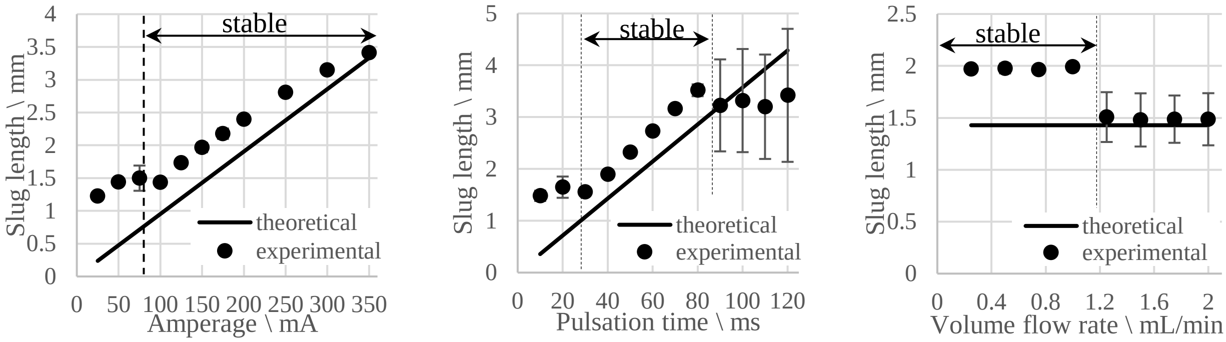

Figure 2: Experimental slug length determination. Left: Variation of the amperage of the electrolysis cell. Middle: Variation of the pulsation time of the current. Right: Variation of volume flow rate in the capillary.

It can be observed that the slug length can be easily adjusted with reasonable accuracy by both the amperage and the pulsation period. A minimum slug length was found to be around 1.5 mm, which is entirely adequate for the desired slug flow regime. The stability limit depicted in the right part of the diagrams is due to the break-up of the gas slugs in the capillary, arising from interaction of various forces in the capillary (e.g. the volumetric flow rate) from excessively long gas release phases (i.e. pulsation time). Overall, good agreement was found between the experimental data and the theoretical values. The slight deviations visible may well result from higher than expected pulsation times due to control equipment defects.

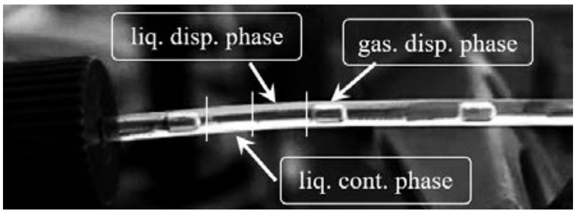

Figure 3: Successfully implemented intermediate gas feeding and resulting three-phase slug flow.

4. Conclusions

For gaseous slug generation, a miniaturised electrolysis cell represents a novel alternative, which is characterised by a rapid response and well-adjustable behaviour. The slug length can be easily be manipulated either by the duration of the current pulse or by the current itself. The volume flow rate inside the capillary and the geometry was found to influence gas slug break-up. By monitoring the upstream structure of the biphasic slug flow using photoelectric sensors, an stable intermediate gas feed without disrupting the existing slugs was successfully demonstrated, exhibiting gas slug lengths similar to those of the two liquids (Figure 3).

Controlled electrolytic gas introduction can be adapted for a whole range of gases, and thus reactions such as hydrogenation or hydroformylation, for example by exploiting an additional membrane to separate the gases generated at the anode and the cathode. It is important to avoid coalescence due to the gas slugs contracting below a certain minimum size. For a liquid reaction medium and an immiscible homogeneous or heterogeneous catalyst carrier phase uniform gas-liquid-liquid slug flow provides a powerful new high-performance tool for conducting multiphase reactions.

References

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