Process Intensification of Photocatalytic CO_2 Reduction Using 3D Printed Microreactors

Abdullahi Adamu, Fernando Russo Abegão, Kamelia Boodhoo

School of Engineering, Newcastle University, Newcastle upon Tyne, NE1 7RU, UK

It is well known that atmospheric CO_2 concentration is increasing at an alarming rate, with CO_2 being considered one of the greenhouse gases responsible for climate change. The effort to curtail carbon emissions requires the contribution of policymakers and scientific community through a series of approaches, including switching to renewable energy sources, carbon capture, storage and utilisation. CO_2 utilisation processes are limited by unfavourable thermodynamic equilibrium, making these energy intensive. A promising pathway around this, is the use of photocatalytic CO_2 reduction, which is a sustainable and energy efficient process for CO_2 conversion to solar fuels and chemicals.

Photocatalytic CO_2 reduction, usually referred to as 'artificial photosynthesis' utilises solar energy to reduce CO_2 in the presence of photocatalyst as shown in Figure 1. While many studies in this area focus on photocatalyst development, little work has been reported on development and design of efficient photocatalytic reactors. This is largely due to the complexity of the reactor design requirements to ensure homogeneity of light irradiation, or unclear understanding of the kinetics of the process. Applying the concept of Process Intensification to this process can address these reactor engineering challenges. Process intensification (PI) is aimed at developing smaller, safer and environmentally benign processes, utilising energy efficient resources and technologies. This work is aimed at utilising PI technology to holistically tailor photocatalysts with appropriate properties in conjunction with the design of novel catalytic intensified reactors for efficient light irradiation.

Conventional photocatalytic reactors suffer from low mass and photon transfer rates, slow reaction kinetics, as well as small area to volume ratios which leads to poor irradiation of the entire reactor volume. Microreactors, with their characteristic large surface area to volume, can be used to achieve high mass, heat and photon transfer rates to the reaction medium, thus offering the potential to improve productivity of photocatalytic reactions.

In this work, a photocatalytic microreactor system constituting of a microchannel chip was developed using high precision 3D printing technology, an innovative automated process that is capable of streamlining the traditional prototype manufacturing steps, allowing fast screening of reactor geometries at a reduced cost.

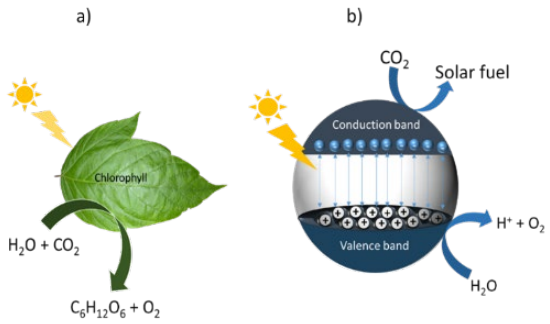

Figure 1. Illustration of (a) natural and (b) artificial photosynthesis.

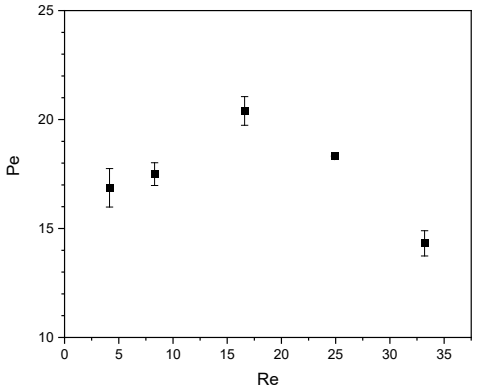

Figure 2. Peclet number (Pe) at different Re in a serpentine microchannel with 2 mm hydraulic diameter.

The computational fluid dynamics studies of the flow in the microchannel have shown a characteristic laminar profile within the range of Reynolds number (Re) of 8 - 141. Residence time distribution experiments were conducted to estimate the mixing behaviour within the microchannel. Peclet number (Pe) was estimated from the model of an open-open vessel at different Re to estimate axial dispersion and identify optimal plug-flow conditions. As shown in Figure 2, the Pe reached its maximum at Re=16.6, with the decrease being attributed to potential bypass of channel geometries for higher flow rates.

The microreactor was used to reduce CO_2 (in the form of bicarbonate) on Cu_xO/TiO_2 photocatalysts, prepared in-house via sol-gel method using Titanium (IV) n-butoxide as precursor, or using commercial P25. The average space time yield on $Cu_xO/P25$ is 0.176 g L^{-1} day^{-1} in the microreactor and 0.059 g L^{-1} day^{-1} in the batch reactor. This result shows nearly 200% productivity increase in the microreactor compared to batch reactor experiments using the same photocatalysts.

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