Direct photochemistry for C-C bonding up to C5 gas phase products and high molecular weight slurry

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Abstract

In this study, we propose an innovative C-C bonding strategy consisting of the use of reactive atomic oxygen from the direct photolysis of gaseous CO_2 for subsequent CH_4 activation. This is achieved in one step under mild conditions and without a dedicated catalyst.

As a photon source, we use a VUV lamp, which emits intense photons with sufficient energy to induce the photodecomposition of CO_2 into CO and O, with a unity primary quantum yield[1] (the threshold for CO_2 photolysis being $\lambda<210$ nm)[2].

The products obtained were analysed in the gas phase via FTIR, GC-MS, and RGA, evidencing the production of C2-C5 functionalized molecules. Surprisingly, a viscous slurry was collected on the reactor surface. This slurry was further analysed by ESI/ASAP-MS, ATR-FTIR, and NMR, revealing a high-molecular-weight complex of functionalised molecules. ^{13}C labelling experiments and simulations (0D kinetic and multiphysic modelling) were conducted to clarify the potential mechanism.

We believe that such an activation strategy is a starting point for new gas-phase chemistry and that a combination of experiments and simulations will allow further optimisation.

References

1. Okabe, H. *Photochemistry of Small Molecules*. (Wiley, 1978).

2. Schmidt, J. A., Johnson, M. S. & Schinke, R. Carbon dioxide photolysis from 150 to 210 nm:

   Singlet and triplet channel dynamics, UV-spectrum, and isotope effects.

Keywords

CO_2-to-fuels ; Direct photolysis ; CO_2/CH_4; High molecular weight slurry