A preliminary investigation of microreactor designs for supercritical water oxidation using hydrothermal flames for space applications

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1. Introduction

The advancement in space technology has led to envisioning long-term human space missions thereby creating need to address diverse technological challenges to make human life sustainable aboard these missions. One such key area is the treatment of organic and human waste. Among several probable solutions, supercritical water oxidation (SCWO) has been sought as the leading candidate to address this problem owing to its several advantages [1]. The underlying principle of the process arises due to change in the behavior of water in its supercritical state (T > 373 C and P > 22.1 MPa). While water is polar at ambient conditions, it becomes non-polar in supercritical conditions (SCW). It thereby acts as a very good solvent for organic matter and several non-polar gases, such as oxygen, nitrogen etc. With both organic matter and oxygen dissolved in it, it acts as a homogeneous medium for the oxidation of the organic matter. The oxidation occurs completely in a single phase which circumvents the limitations posed by interfacial mass transfer and thus, very fast reactions are obtained with up to 99.9% degradation [2]. The organic waste is completely converted to gaseous products such as CO_2. Furthermore, no toxic products such as NOx, CO and dioxins are formed as temperatures are relatively low for their production rendering SCWO a clean disposal method.

Despite these advantages, SCWO process has been faced by two major challenges, generally termed as a dual problem in SCWO. Firstly, corrosion of the process equipment due to reactive ions such as Cl−, F−, H3O+, and secondly, plugging of the reactors/process equipment due to the precipitation of ionic/inorganic salts (their solubility decreases with decrease in dielectric strength of SCW). One remedy to tackle this problem is to avoid the temperature rise of the inlet streams beyond a certain limit to prevent the precipitation of the inorganic salts. This is made possible by injecting the feed (water + waste) at subcritical temperatures and attain supercritical conditions in the reactor by virtue of hydrothermal flames acting as an internal heat source [3]. Hydrothermal flames refer to the flames produced in the water due to combustion reaction between organic matter with oxidant in a SCW environment, primarily attributed to the reduction in the autoignition temperature of organic salts at high pressures, for example methanol. Hydrothermal flames can be obtained as low as 400 C [3] and are generally characterized by temperatures above 1000 C [4]. In addition to solving the plugging problem, this methodology also presents the following advantages, (a) up to 99.99% conversion of the reactants, (b) lower reaction times, ~10ms -100ms (c) formation of less harmful products, (d) smaller reactor designs due to low reaction times.

In order to use this methodology for space applications, it is inevitable to develop the technology at microscale owing to space and weight limitations in space missions. The scientific challenge lies to understand the behavior of hydrothermal flames confined within microreactor in zero-gravity environment and how it will govern the SCWO behavior when compared to ground conditions. The current study marks a first step towards achieving this goal wherein microreactors have been designed accounting for various factors to effectively carry out the process at microscale.

2. Results: Design considerations for microreactor

One of the key features in the current setup is the appropriate choice of material which can withstand high pressures (~250 bar) and extreme temperature conditions in addition to being chemically compatible with SCW. While sapphire is found to meet all the desired conditions and is planned for final application, the preliminary designs as presented in the current study are based on Si-Pyrex system.

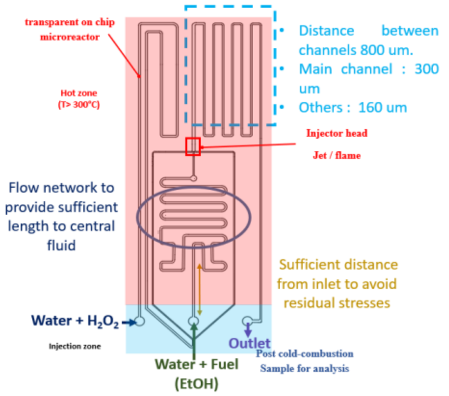

Figure 2: Schematic of the reactor

From design perspectives, various factors are addressed. One of the primary requirements is to have a sufficient gap between the channels of the microreactor to minimize the risk of the reactor being ruptured by mechanical stresses due to high pressure operating conditions (~250 bar). After several iterations in the design and testing, the dimensions of the channel were set to be 300 μm for main channel with gap of 800 μm between them (Fig. 1). The second design parameter is laying out the flow network and path which is motivated by two primary objectives: (a) the oxidizer in the reactor will be derived from the decomposition of H_2O_2 into H_2O and O_2. This requires that adequate flow path (and thus time) be available for decomposition of H_2O_2. (b) as the reactor is uniformly heated, it is desired that both the fuel and oxidizer stream enter the main channel at the same temperature. This necessitates providing sufficient residence time for the fuel stream (comparable to oxidizer stream) following which a serpentine like flow path has been designed as illustrated in Fig. 1. The last design aspect accounted for has been the injector head, $i.e.$, region of the main channel where the fuel and oxidizer are injected. In the current framework, a non-premixed flame configuration is expected where the combustion is primarily governed by mixing dynamics. However, laminar operating conditions can lead to inefficient mixing and thus higher ignition lag. Further, this can also lead to the formation of flame further downstream in the main channel, a phenomenon not desired owing to space constraint at microscale. The problem has been addressed by designing secondary flow channels as well as providing island like structure in the main channel for anchoring the flame. Experimental result on Si-Pyrex system supports the premixing behavior and that the injected H_2O_2 was decomposed into O_2 and H_2O (Fig. 2). More sophisticated injector heads based on tesla-valve configurations have also been designed and investigated.

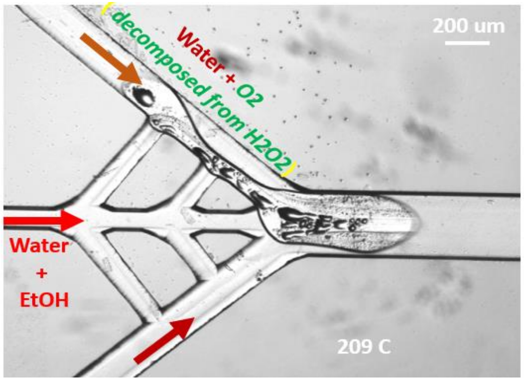

Figure 1: Observation of 2 phase flow due to decomposition of H_2O_2 to $H_2O + O_2$ (gas) at 110 bars.

3. Perspectives

The presented design considerations have been validated for their functionalities on model system setup. The next step is to carry out experiments on sapphire microreactor under real conditions. Further, the DNS studies of hydrothermal flames in the presented conditions using single step finite rate chemistry for oxidation of ethanol in supercritical water environment will be undertaken to optimize the experimental conditions in terms of flow rate and fuel/oxidizer concentrations.

References

[1] Zhang S., Zhang Z., Zhao R., Gu J., Liu J., Örmeci B., Zhang J., A Review of Challenges and Recent Progress in Supercritical Water Oxidation of Wastewater, Chemical Engineering Communications, 204 (2017) 265-282.
[2] Bermejo M.D., Cocero M.J., Supercritical water oxidation: A technical review, AIChE Journal, 52 (2006) 3933-3951.
[3] Reddy S.N., Nanda S., Hegde U.G., Hicks M.C., Kozinski J.A., Ignition of hydrothermal flames, RSC Advances, 5 (2015) 36404-36422.
[4] Augustine C., Tester J.W., Hydrothermal flames: From phenomenological experimental demonstrations to quantitative understanding, The Journal of Supercritical Fluids, 47 (2009) 415-430.