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Are micro reactors inherently safe? An investigation of gas phase explosion propagation limits on ethene mixtures

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ABSTRACT

A method for the determination of safety properties for micro reactors and micro structured components is presented. Micro structured reactors are not inherently safe but the range of safe operating conditions of micro reactors are extended since the explosion region is reduced. The λ/3 rule was demonstrated to be applicable to micro scale tubes for stoichiometric mixtures of ethane–oxygen and ethane–nitrous oxide. Furthermore first results from an investigation concerning detonation propagation through a micro reactor of non-ideal geometry are shown. Initial pressure investigated is ranging from low pressure up to 100 kPa.

Keywords: Micro reactor; Explosion propagation limits; Chemical safety; Investigation method

1. Introduction

Micro reaction technology has become increasingly interesting for industrial applications since it provides several advantages over conventional reaction technology such as enhanced heat and mass transfer as well as large specific phase areas (Jähnisch et al., 2004). Often, it is assumed that micro structured devices are inherently safe since explosion propagation will be suppressed due to the small dimensions. This enables gas mixtures to process even in an extended range of reaction conditions that are within the explosion range as specified by the European standard investigation method for explosion limits (DIN EN 1839, 2003). Nevertheless, it is not possible to safely operate micro reactors at any condition. If an explosion can propagate through the micro device, it may cause ignition in a subsequent volume containing hazardous material. If it is intended to make use of the enhanced safety of micro devices, then for safety assessment of a plant containing micro structured components it will be essential to know the limits of explosion propagation. A detailed review of various safety aspects in micro structured reactors is given elsewhere (Gödde et al., 2009).

This paper presents a method for the determination of maximum safe capillary diameters and its results as well as an attempt to transfer this approach to a real micro scale reactor. This will enable to adequately describe the extended safety range of operating conditions for gas phase partial oxidation reactions in micro structures. In view of the ethene oxide process, the ethene–oxygen and ethene–nitrous oxide systems are investigated. Since the conservative consideration requires safety to be sustained even in the case of an erroneous volumetric educt dosing, the most hazardous mixture is to be investigated for the purpose of safety consideration. Furthermore, a new micro reactor with a channel shape and a catalyst area, based on the design of the DEMIS-reactor (Markowz et al., 2004; Klemm et al., 2008) is considered in the present study. First results and the work still to be done will be discussed.

2. Methods and materials

Two versions of experimental setup were used. In both of them a detonation was ignited in a macroscopic primary chamber and the propagation of the detonation through either stainless steel capillaries of different inner diameter or a micro reactor was traced by pressure transducers. The inner diameter of the macro tube was 40 mm in the first version and 18 mm in the second set-up version. The steel capillaries had inner diameters of $d1 = 1.00$ mm, $d2 = 0.50$ mm, $d3 = 0.25$ mm and

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Nomenclature

λ	detonation cell width
d_s	safe capillary diameter
P_0	initial pressure before ignition
T	initial temperature
v	propagation speed
y	molar fraction

d4 = 0.13 mm. The total length of the capillary was 1 m. It was intercepted by two pressure measurement tips to determine explosion propagation velocities inside the capillaries, maximum safe initial pressures and the corresponding maximum safe capillary diameter. Additional pressure sensors were mounted at the secondary macro tube to monitor whether explosion will be triggered in the subsequent macroscopic volume. Dynamic pressure measurement was performed using piezoelectric pressure transducers of Kistler type 601H providing a measurement range of P = 0–100 MPa with a resonant frequency of 150 kHz. Signal amplification was performed using a Kistler amplifier type 5001. The signals were sent to an A/D converter and recorded at a sampling rate of 1 MHz using a ScopeCorder from Yokogawa, Type DL 750. Investigations were performed on stoichiometric (with respect to total oxidation reaction) mixtures of ethene–oxygen and ethene–nitrous oxide at room temperature. Prior to explosion experiments homogeneous mixtures were prepared using the method of partial pressures in a separated mixing vessel. In the primary macro volume an (exploding wire type) igniter was initiated by a capacitive ignition energy unit providing an electric energy of 10 J to initiate a detonation in the primary chamber. Pressure–time histories of the pressure sensors were recorded. Fig. 1 depicts the simplified flow sheet diagram of the experimental setup used. An experiment in a similar setup using hydrogen–air-mixtures was reported in the literature (Pfeifer et al., 2004).

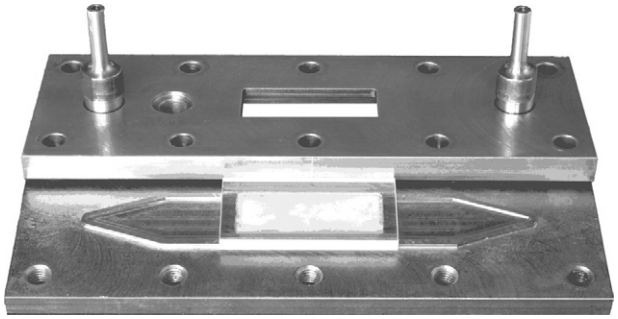

Fig. 2 – Photo of the channel-shaped micro reactor consisting of the top plate with two connections for in- and outlet, a window and the bottom plate containing the micro channel and the catalyst bed.

As mentioned above, instead of the small capillary, a micro reactor of non-ideal geometry was mounted as well. The micro reactor is shown in Fig. 2. It provides a channel height of 0.25 mm and channel width of 20 mm. Outside dimensions of the reactor top and bottom plates are 200 mm × 70 mm, the area of the visible range through the

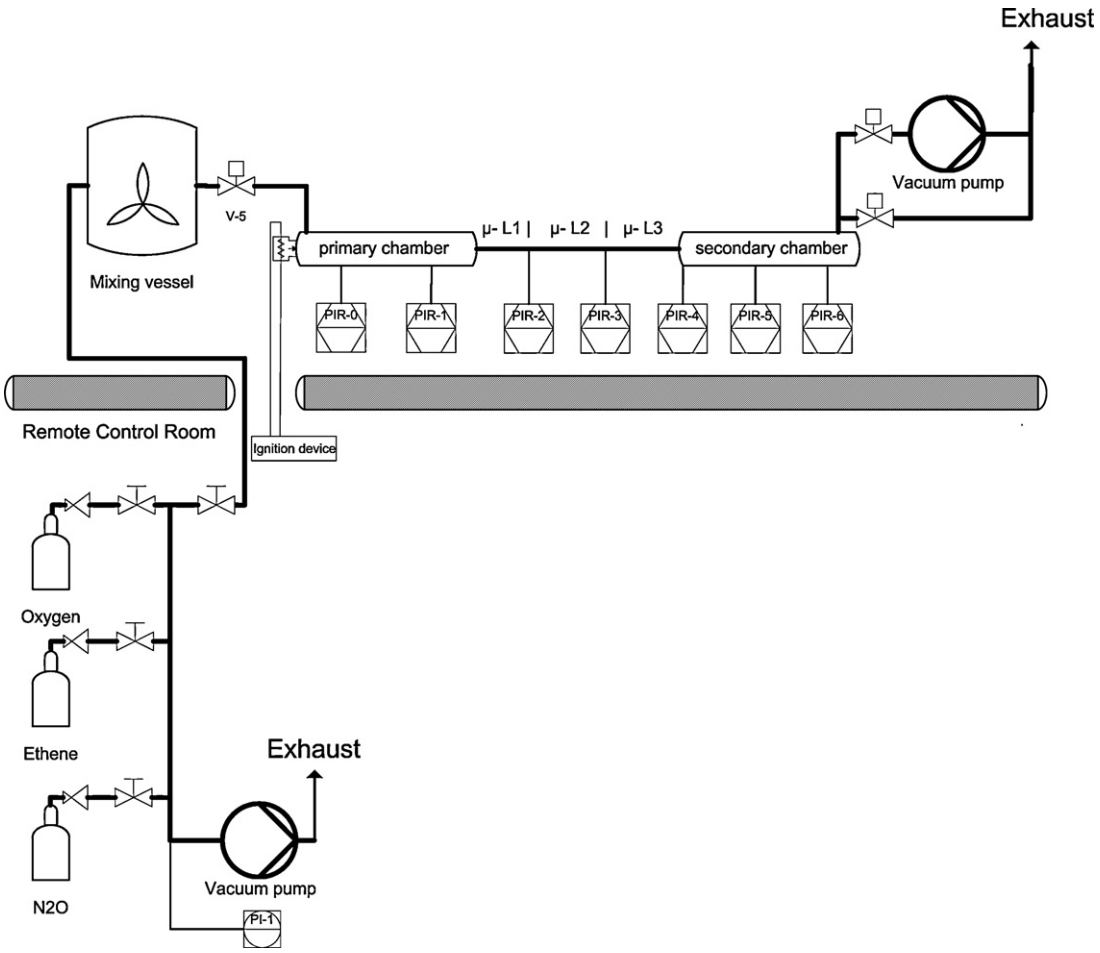

Fig. 1 – Simplified flow sheet diagram of the explosion propagation apparatus for micro structured devices.