Investigation of droplet nucleation in CCS relevant systems – Design and testing of a CO_2 branch of the mixture preparation device

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Abstract. A unique in-house designed experimental apparatus for investigation of nucleation of droplets in CCS relevant systems is being developed by our team. The nucleation is measured with the help of a rapid pressure drop within an expansion chamber. In this study, a CO_2 branch representing an important part of the mixture preparation device (MPD) was designed, assembled and tested. MPD is intended for preparation of CO_2-rich mixtures in a sense of accurate setting of flow rates of individual gaseous components through the experimental section with the expansion chamber. In the CO_2 branch, the saturated liquid CO_2 coming from a cylinder with a dip tube is pressurized by a supercritical pump with a constant flow rate to the pressures of up to 340 bar. The required mass flow of the supercritical CO_2 of up to 180 g/h at pressures between 74 bar and 150 bar is accurately tuned with a help of a set of stainless steel capillary tubes with inner diameter of 0.1 mm. Some of the tubes are submerged in a thermostatic bath set to a constant temperature of 70 °C in order to assure the supercritical flow conditions of CO_2. Needed lengths of the capillary tubes were determined with a help of a one-dimensional numerical model allowing prediction of both isothermal and adiabatic flow conditions.

1 Introduction

The team of the Laboratory of Phase Transition Kinetics at the Institute of Thermomechanics of the CAS is developing a unique experimental apparatus for investing the formation of droplets in CO_2-rich systems. Motivation for this research is to obtain new valuable data for nucleation of droplets (the initial stage of droplet formation) in binary and multicomponent mixtures important in the design of new carbon capture and storage (CCS) technologies [1]. At the current state, the research of our team is focused mainly on binary systems of carbon dioxide with nitrogen and argon. Later, binary systems with water and ternary systems will be studied.

A unique mixture preparation device (MPD) for preparation of CO_2-rich mixtures with up to three components was designed by our team. Its function is based on precise setting of flow rates of individual components within the measuring system. In this study, the CO_2-branch representing the most important part of MPD was designed, assembled and successfully tested.

2 New apparatus for investigation of nucleation in CO_2-rich systems

In the new experimental apparatus, the nucleation of droplets in CO_2-rich systems is measured with the help of a rapid pressure drop inside a stainless steel expansion chamber [2-4]. The apparatus allows measurement under wide operating conditions at temperatures from –20 °C to +150 °C and pressures up to 130 bar. Consequently, the nucleation of droplets can be investigated both in a low pressure vapor and in a high pressure supercritical fluid. We note that the critical point temperature and pressure of CO_2 are 30.98 °C and 73.8 bar, respectively. The apparatus allows simulating various scenarios relevant in CCS technologies including sudden rupture of the pressurized pipeline with CO_2-rich fluid.

The expansion within the chamber, filled with a CO_2-rich mixture in the form of the superheated vapor or supercritical fluid, can be considered as isentropic due to the fast pressure drop. The heat transfer from the walls of chamber and the ambient into the fluid can be neglected in this case. As the decreasing pressure crosses the equilibrium two-phase envelope, the fluid becomes thermodynamically metastable, i.e. at supersaturated state. At certain conditions given by the initial state and the kinetics of the pressure drop, the supersaturation of the original phase is large enough such that the phase transition is initiated. Liquid droplets start to nucleate and subsequently grow.

Conditions inside the expansion chamber are measured with a fast-response pressure transducer and accurate temperature probes. The nucleation of droplets and their subsequent growth is detected optically by using an in-house designed interferometer.

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3 Mixture preparation device

Gaseous mixtures with CO_2 are prepared within MPD using the principle of an accurate adjustment of flow rates of individual components. The flow rates are tuned with a supercritical fluid pump (SCP) for CO_2, a set of flow controllers and stainless steel capillary tubes used as elements with defined pressure drop at a given flow rate. The temperatures and pressures of flowing gases are varied and measured with the help of a set of thermostatic baths, backpressure regulators, pressure transducers and precise temperature probes.

Preparation of mixtures by fine tuning of flow rates has an important advantage for the investigation of nucleation, which is highly sensitive to a presence of impurities. The gaseous mixtures can flow through MPD and the expansion chamber for longer time (even several hours if needed). This helps to equilibrate the adsorption of gas molecules on the walls of the MPD system and the expansion chamber and to flush away possible impurities. We note that the system is carefully evacuated to a low vacuum before each test. Most of the MPD tubbing is made from stainless steel tubes with outer diameter of ¼ inch and wall thickness of 0.036 inch and 0.049 inch for maximum pressures of 350 and 515 bar, respectively.

At the current state, the MPD system consists of three main parts: the CO_2 branch, the gaseous branch with carrier gas (nitrogen or argon), and the final section with mixed gases containing the expansion chamber. In this study, the design and testing measurements of the CO_2 branch are described in detail.

3.1 Gaseous branch with nitrogen or argon

The carrier gas, either nitrogen or argon, is delivered from the 50 liter gas cylinder with a maximum load pressure of 200 bar. The output pressure is varied with the pressure regulating valve and measured with 300 bar pressure transducer of 33 series from the Keller[a] manufacturer. The flow rates are set by two thermal mass flow meters of F-23xM series designed for high pressure operation up to 400 bar from the Bronkhorst[b] manufacturer. The two flow meters have different ranges from 0.01 to 0.50 l_n/min and from 0.04 to 2.0 l_n/min. Unit l_n/min denotes flow rate of gas calculated at the normal conditions, i.e. at the density of given gas at temperature of 0 °C and atmospheric pressure of 101 325 Pa. Nominal flow of 1.0 l_n/min corresponds to mass flow of 75 g/h and 107 g/h in case of nitrogen and argon, respectively. The gaseous branch is designed to operate with outlet pressures from 38 up to 160 bar.

The carrier gas is mixed with carbon dioxide coming from the CO_2 branch in ¼ inch tubes placed inside a thermostatic bath set to temperatures between 5 and 150 °C depending on the conditions for nucleation of droplets investigated within the expansion chamber.

3.2 CO_2 branch

The CO_2 branch allows fine adjustment of the CO_2 mass flow up to 180 g/h at the outlet pressures of the supercritical fluid from 74 to 150 bar. A simplified scheme of the setup is provided in Fig. 1. The saturated liquid CO_2 coming from cylinder (1) equipped with a dip tube enters the system at an approximate pressure of 63 bar corresponding to the vapor pressure of CO_2 at room temperature 24 °C. The inlet pressure is detected with pressure gauge (2). The saturated liquid CO_2 is compressed by supercritical fluid pump (SCP)[c] (3) up to pressure of 340 bar representing the allowed maximum set on the safety valve after the pump. Typical operating pressure of SCP is around 270 bar. The two-piston supercritical pump operates at a constant flow rate of up to 24.0 ml/min of the saturated liquid CO_2. In order to eliminate the pressure fluctuations caused by the operation of the supercritical pump, the system is equipped with a high-pressure buffer tank (accumulator – AC) with inner volume of 150 ml (item 4 in Fig. 1). Part of the supercritical CO_2 from the buffer tank returns back to the supercritical pump inlet through a backflow capillary tube (CAP 2) with inner diameter of 0.10 mm and length of 0.85 m. Within the backflow capillary tube, the supercritical CO_2 is adiabatically throttled to the saturated liquid state. At the SCP operating pressure of 270 bar, the mass flow through the capillary tube is around 340 g/h. The backflow capillary tube allows setting SCP approximately to the middle of its range, i.e. to the flow rate around 12.0 ml/min of the saturated liquid CO_2 corresponding to the mass flow of 511 g/h. The mass flow going to the rest of MPD and the expansion chamber varies typically between 50 g/h and 180 g/h. This part of the CO_2 flow is expanded to the required outlet pressure in two stainless steel capillary tubes (CAP 3 and CAP 4) placed in a thermostatic bath (5). The thermostatic bath with water is set to the temperature around 70 °C, i.e. far above the critical temperature of CO_2. The capillary tubes CAP 3 and CAP 4 have equal inner diameters of 0.1 mm and lengths of 3.5 m and 7.5 m, respectively. The capillary tubes CAP 3 and CAP 4 deliver mass flow of 112 g/h and 73 g/h at the SCP operating pressure of 270 bar and the CO_2 branch outlet pressure of 100 bar. Each of the two capillary tubes can be open / closed by a needle valve. Temperature along the system is measured with three small Pt100 resistance thermometers (15–17) attached to the outer surface of the ¼ inch tubing.

In the final design of MPD, the CO_2 branch will end at the location of the temperature sensor No. 17 where it will be connected to the final section with mixed gases. For the individual testing of the CO_2 branch, the setup was additionally equipped with two pressure transducers Keller[a] of the 33 type with the range from 1 to 300 bar and accuracy better than 0.45 bar (items 7 and 8 in Fig. 1). The transducers measured pressures after the buffer tank AC (7) and after the isothermal capillary tubes CAP 3 and CAP 4 (8). The mass flow through the system was measured with the Coriolis mass flow meter

[a] www.keller-druck.com
[b] www.bronkhorst.com
[c] http://ssihplc.com/hplc-pumps/supercritical-24-pump-cf/

mini CORI-FLOW[b] (18) with the range from 2 to 200 g/h and accuracy better than 0.2% of reading. The flow meter operates at pressures from 3 up to 140 bar and can measure both the gases and the liquids. The pressure drop on the flow meter is between 1.7 and 24 bar depending on the flow rate and the fluid properties, i.e. mostly compressible/incompressible fluid. The testing CO_2 branch is terminated by an exhaust capillary tube CAP 5 where the supercritical CO_2 is adiabatically throttled to the atmospheric pressure. The capillary tube has inner diameter of 0.1 mm and length of 0.77 m.

The supercritical fluid pump requires approximately 30 minutes of flushing with pure CO_2 before its operation in order to remove air humidity and other impurities from its internal volumes. The CO_2 branch is therefore equipped with a flushing capillary tube CAP 1 with inner diameter 0.1 mm and a length of 0.33 cm. In CAP 1, the saturated liquid CO_2 is throttled from the vapor pressure of 63 bar to the atmospheric pressure by the maximum proposed flow rate below 150 g/h. Fig. 2 shows photo of the main part of the CO_2 branch in the laboratory.

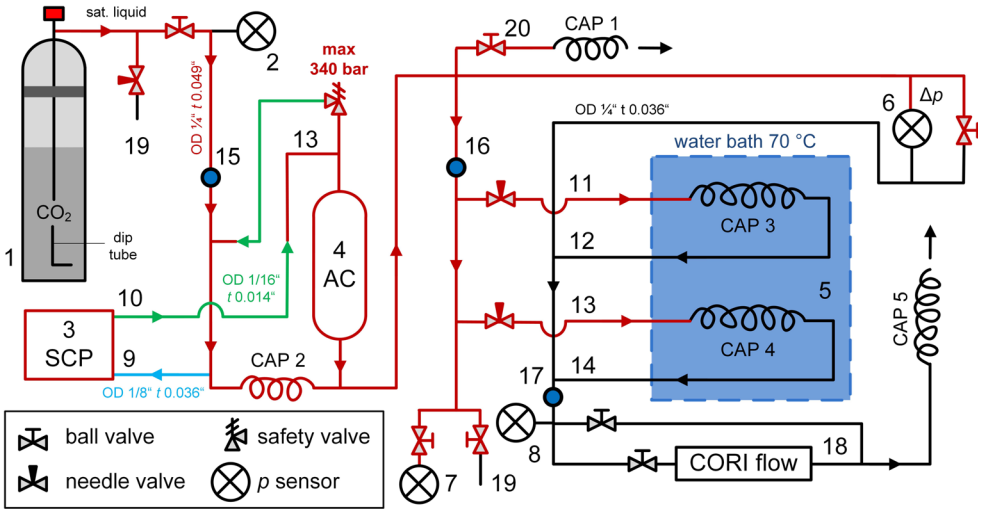

Fig. 1. Scheme of the CO_2 branch of the MPD system. Description: 1 – liquid CO_2 cylinder, 2 – pressure gauge, 3 – supercritical fluid pump (SCP), 4 – accumulator (AC), 5 – thermostatic bath, 6 – differential pressure transducer, 7 – absolute pressure transducer – high p, 8 – absolute pressure transducer – low p, 9 – SCP inlet, 10 – SCP outlet, 11 – CAP 3 inlet, 12 – CAP 3 outlet, 13 – CAP 4 inlet, 14 – CAP 4 outlet, 15 – T sensor (from the CO_2 cylinder), 16 – T sensor (after AC), 17 – T sensor (after bath), 18 – mass flow meter, 19 – connection for vacuum pump, 20 – flushing valve.

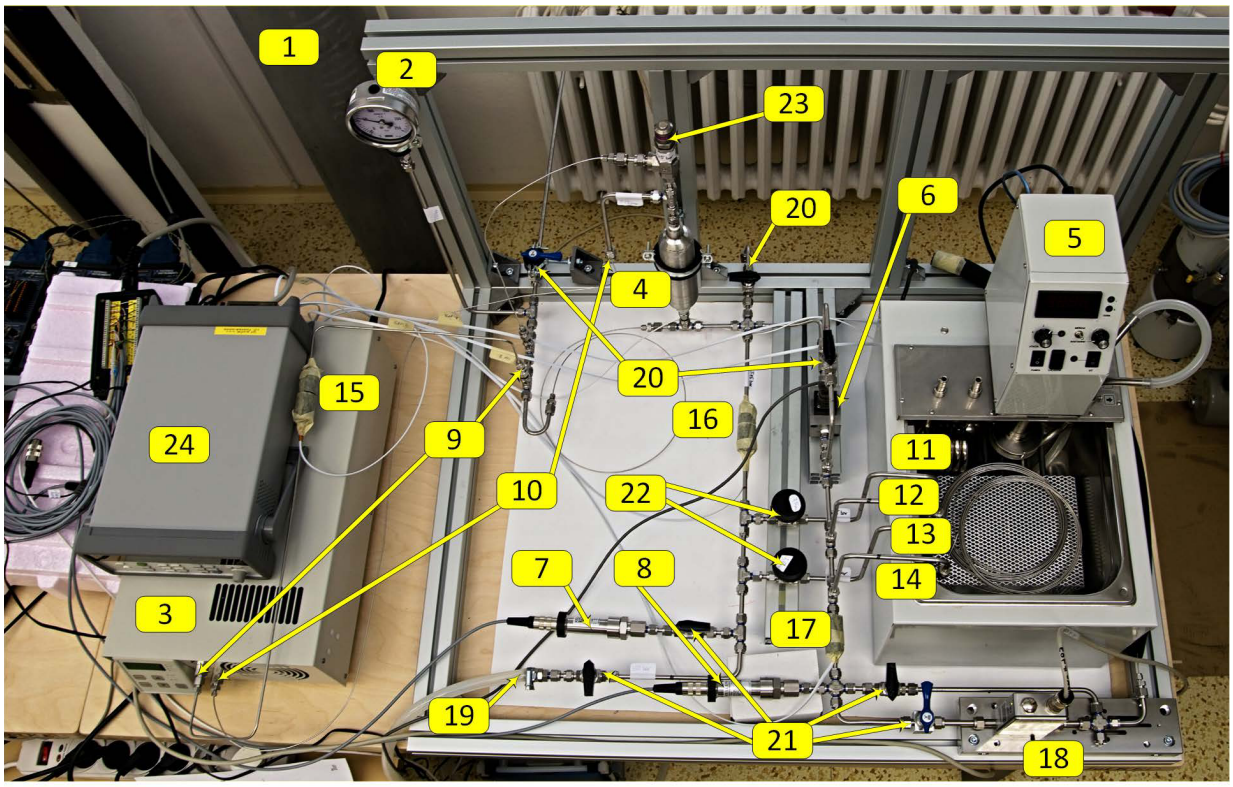

Fig. 2. Photo of the CO_2 branch of the MPD system. Description: points from 1 to 20 are same as in Fig. 1; 21 – ball valves, 22 – needle valves, 23 – safety valve, 24 – DAQ system.

3.2.1 Capillary tubes on CO_2 branch

A set of various capillary tubes CAP 1 to CAP 5 operating at different flow regimes is used on the CO_2 branch of MPD; see Fig. 1. Table 1 summarizes parameters of all capillary tubes and their designed flow regimes. Capillary tubes CAP 1, CAP 2 and CAP 5 operate at adiabatic flow conditions, while the flow in CAP 3 and CAP 4 placed inside the thermostatic bath can be considered as isothermal. In the exhaust capillary tubes CAP 1 and CAP 5, the two-phase flow of vapor-liquid mixture occurs as CO_2 is throttled from pressures above 60 bar down to the atmospheric pressure. In case of remaining three capillary tubes, the CO_2 fluid stays under the supercritical conditions along the entire length of the capillary tube.

Table 1. List of capillary tubes (CAP) employed on the CO_2 branch and their operating flow conditions.

CAP	**1**	**2**	**3**	**4**	**5**
***d* [mm]**	0.1	0.1	0.1	0.1	0.1
***L* [cm]**	33	85	350	750	75
T_{in} [°C]	25	25	40	40	38
p_{in} [bar]	64	220	220	220	100
p_{out} [bar]	1	64	100	100	1
flow condition[1]	A	A	iT	iT	A
CO_2 state[2]	TP	S	S	S	S + TP

[1] A – adiabatic, iT – isothermal
[2] TP – two-phase vapor + liquid, S – supercritical

The flow performance of the isothermal capillary tubes CAP 3 and CAP 4 was predicted with a simple one-dimensional numerical model. The defined pressure drop over the capillary tube from the inlet pressure p_{in} to the outlet pressure p_{out} was divided in N segments, i.e. $\Delta p_{i-1} = p_{i-1} - p_i$ with $i = 1 \div N$. Length of the i-th segment of the capillary tube Δz_i could be obtained from the following equation

$$\Delta z_i = \frac{p_{i-1} - p_i - \frac{\dot{m}^2}{A^2}\left(v_i - v_{i-1}\right)}{\frac{4 f_{i-1} \dot{m}^2 \left(v_i + v_{i-1}\right)}{\pi^2 d^5}}, \qquad (1)$$

where $\dot{m}$ is the mass flow in kg/s, d is the capillary tube inner diameter, A is the cross section area $A = \pi d^2/4$, v is the specific volume in m³/kg and f is the friction factor calculated, e.g., from the Colebrook equation [5]

$$\frac{1}{\sqrt{f}} = -2\log\left(\frac{\varepsilon}{3.7d} + \frac{2.51}{Re\sqrt{f}}\right). \qquad (2)$$

In equation (2), Re is the Reynolds number and ε is the inner wall roughness of the capillary tube. In all our calculations, a relative wall roughness $\varepsilon/d = 0.003$ was considered. Thermophysical properties of CO_2, i.e. the density $\rho = 1/v$ and viscosity, were taken as functions of pressure p_i at a constant temperature. The properties at each computational grid point i were calculated using the property packages REFPROP [6] and TREND [7].

The two-phase flow of vapor-liquid mixture within an adiabatic capillary tube was predicted by the numerical model similar to that employed in our previous studies [8-10]. More details about the design of the capillary tubes and the flow behavior of CO_2 at various inlet and outlet conditions will be provided in other article.

4 Preliminary tests

Functionality of the CO_2 branch was successfully tested in this study. Signals from the pressure transducers, the mass flow meter, and the temperature sensors were recorded by the automatic data acquisition system (DAQ) Keysight[d] 34970A (item 24 in Fig. 2).

Fig. 3 shows temperatures along the system recorded during the flushing of SCP. The saturated liquid CO_2 coming from the cylinder flows through SCP and leaves the CO_2 branch via the exhaust capillary tube CAP 1. Three temperature sensors T CAP 1 to 3 were additionally placed on the capillary tube in the distances 140, 60, and 4 mm from the capillary tube outlet. As can be seen in Fig. 3, the temperature drops along the capillary tube from 24 °C to 7 °C due to the evaporation of liquid CO_2 during the adiabatic throttling. We note that the outlet temperature is significantly below the minimum measured temperature of 7 °C. The recorded temperatures along the capillary tube have only informative character as their uncertainties are quite high due to the relatively low flow rates, very small inner diameter of the capillary tube of 0.1 mm compared to its wall thickness around 0.75 mm and poor thermal contact between the temperature probes and the capillary tube with outer diameter of only 1.6 mm. Nevertheless, the measured temperatures show physically correct trend.

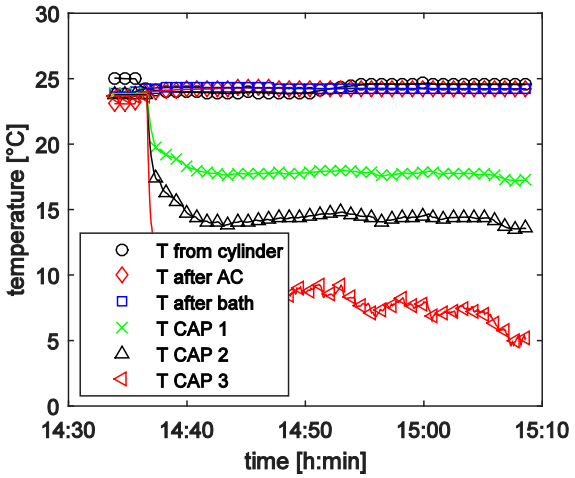

Fig. 3. Temperatures along the system and the exhaust capillary tube CAP 1 during the flushing of SCP with the saturated liquid CO_2.

Fig. 4 shows typical conditions during the startup of SCP. The flow rate through the pump was set to

[d] http://www.keysight.com

7.0 ml/min of the saturated liquid CO_2 in this case. As can be seen, the pressure at the pump outlet, i.e. after the buffer tank AC, is gradually increasing until it stabilizes at a final value of 180 bar. The pressure after the isothermal capillary tubes CAP 3 and CAP 4 together with the mass flow are accordingly increasing to the equilibrated conditions of 77 bar and 105 g/h, respectively. The system got stabilized after approximately 30 minutes from the pump startup due to the relatively low flow rate of 7.0 ml/min set on the pump compared to the proposed operating conditions at 12.0 ml/min.

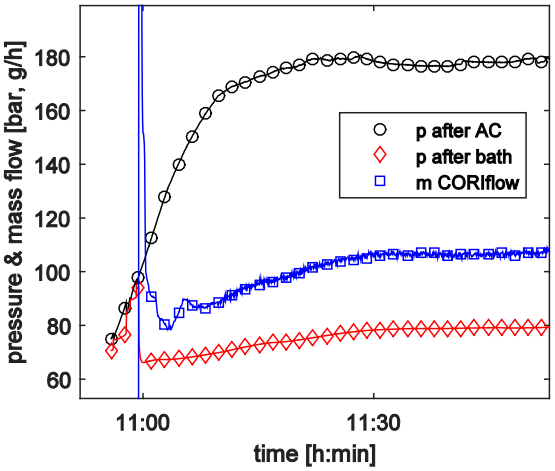

Fig. 4. Pressure in bar after the buffer tank (AC) and after the thermostatic bath with capillary tubes CAP 3 and CAP 4 and the mass flow in g/h measured by mini CORI-FLOW after the startup of SCP with the set point of 7.0 ml/min.

Fig. 5 shows temperatures measured along the CO_2 branch and additionally at the outer surface of the exhaust capillary tube CAP 5 during the startup of the supercritical pump. The flow conditions correspond to the case depicted in Fig. 4. Temperature in the thermostatic bath with capillary tubes CAP 3 and CAP 4 was set to a constant value of 55 °C. It is important to note that the tubing of the CO_2 branch was not insulated during the preliminary tests. Consequently, the temperature after the thermostatic bath varied relatively slowly due to the heat losses into the ambient. The temperature sensors T CAP 1 to 3 were placed on the outer surface of capillary tube CAP 5 in a distance of 229, 74, and 3 mm from its outlet, respectively. Similarly as in case shown in Fig. 3, the temperatures measured along the capillary tube have only informative character. Fluctuations of temperature T CAP 1 are caused by a presence of water ice formed from the condensed air humidity close to the capillary tube outlet.

Performance of the CO_2 branch was investigated at several stabilized flow conditions. Some of these tests are summarized in Table 2. First two cases were measured with SCP turned off. The maximum pressure in the system was given by the vapor pressure of CO_2 at the room temperature. First case represents ordinary flushing of the CO_2 branch before the startup of SCP and corresponds to the results shown in Fig. 3. In the second case and the third case, the exhaust capillary tube for flushing CAP 1 was placed after the mass flow meter instead of the exhaust capillary tube CAP 5 shown in Fig. 1. The third case was measured with the flow rate of the saturated liquid CO_2 through SCP of 5.0 ml/min by both isothermal capillary tubes CAP 3 and CAP 4 open. The thermostatic bath (5) was set to 40 °C in this case. The mass flow through the system and the pressure after SCP stabilized at 87.6 g/h and 137 bar, respectively.

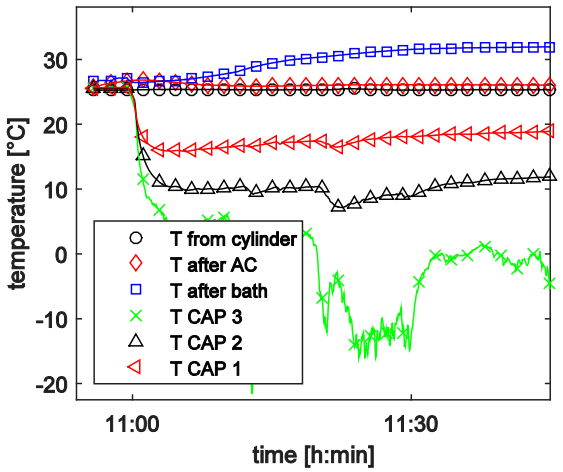

Fig. 5. Temperatures in the system measured after the startup of SCP with the flow rate of the saturated liquid CO_2 set to 7.0 ml/min. Temperatures T CAP 1 to 3 were measured at the outer surface of the exhaust throttling capillary tube CAP 5.

Last two cases in Table 2 were measured at the proposed operating conditions, i.e. at the flow rate set to 12.0 ml/min on SCP and the thermostatic bath set to 70 °C. The capillary tube CAP 5 was employed as the exhaust tube after the mass flow meter in both cases. In the first case, the isothermal capillary tube CAP 3 with the length of 3.5 m was open while in the second case the capillary tube CAP 4 with the length of 7.5 m was active. Results in Table 2 show an interesting phenomenon. The pressure drop over the shorter capillary tube CAP 3 equal to 184 bar_d was higher than in case of the longer capillary tube CAP 4 equal to 158 bar_d. Consequently, the absolute pressure before the mass flow meter was 10 bars higher in case of open capillary tube CAP 4 than in case of open capillary tube CAP 3. As a result, the mass flow through the system was higher by 13.7 g/h in case of open capillary tube CAP 4. The result is in contradiction with the numerical model described in section 3.1.1. At the flow conditions provided in Table 2, the numerical model predicts maximum mass flow of 112 g/h and 74 g/h for capillary tube CAP 3 and CAP 4, respectively. Possible explanation for this behavior can be that the capillary tubes do not have a constant inner diameter of 0.10 mm. An increase of the inner diameter to 0.11 mm results in the mass flow predicted by the model of 144 g/h for CAP 3 and 95 g/h for CAP 4. The other explanation could be that the flow performance of the CO_2 branch is strongly affected by the exhaust capillary tube CAP 5, i.e. the last element of the CO_2 system where the pressure drops from approximately 100 bar to the atmospheric pressure. The exhaust capillary tube behaves

Table 2. Description of stabilized flow conditions within the CO_2 branch at different set points on SCP and different configurations of the exhaust tube to atmosphere

Pump set point	iso-*T* CAPS	*p* after AC	*p* after bath	$\dot{m}$ CORI-FLOW	*T* from cylinder	*T* after AC	*T* set on bath	*T* after bath	*T* CAP 1	*T* CAP 2	*T* CAP 3	Exhaust tube
[ml/min]		[bar]	[bar]	[g/h]	[°C]	[°C]	[°C]	[°C]	[°C]	[°C]	[°C]	
off[1]	both	63.21	63.15	0.00	24.6	24.2	–	24.2	17.7	14.3	7.3	
off[2]	both	63.39	38.21	25.05	25.0	24.2	–	24.5	17.7	16.4	10.5	CAP 1
5	both	137.19	70.42	87.55	25.6	25.9	40	28.7	–	–	–	
12	CAP 3	283.45	99.15	135.43	24.7	26.2	70	35.7	23.5	16.2	0.7	CAP 5
12	CAP 4	268.36	110.64	149.12	24.8	26.0	70	36.3	23.7	18.2	1.3	

[1] Flushing of the system – exhaust capillary tube CAP 1 placed after the flushing valve (item 20 in Fig. 1)
[2] Exhaust capillary tube CAP 1 placed after the mini CORI-FLOW mass flow meter instead of CAP 5

similarly as a chocked nozzle at these conditions. The detected performance of the isothermal capillary tubes CAP 3 and CAP 4 combined with the exhaust capillary tube CAP 5 require further investigation. However, in the final design of MPD, the mass flow meter and the exhaust capillary tube will not be present. A new test is therefore planned in the close future. The exhaust capillary tube CAP 5 will be removed from the system. The outlet of the mass flow meter will be connected back to the CO_2 cylinder outlet (item 1 in Fig. 1) after pressure gauge (2). In this case, the pressure after the mass flow meter will be equal to the vapor pressure of CO_2, i.e. approximately 63 bar, which is relatively close to the designed conditions when the outlet pressure from the CO_2 branch shall vary between 74 and 150 bar. Another option is to replace the current capillary tubes CAP 3 and CAP 4 with another capillary tubes having the same dimensions and to verify the results measured in this study. The inner diameter can significantly vary along the capillary tube length [9,10].

5 Conclusions

A CO_2 branch representing the main part of a unique mixture preparation device (MPD) was designed, assembled and successfully tested in this study. The MPD system is intended for preparation of CO_2-rich mixtures in a sense of accurate setting of flow rates of individual components of the gaseous mixture. MPD is part of the larger in-house designed experimental apparatus for investigation of nucleation of droplets in CCS relevant systems.

The CO_2 branch comprises of several high-tech components such as supercritical fluid pump, accurate pressure transducers and Coriolis mass flow meter. A set of various stainless steel capillary tubes are used in the CO_2 branch for fine tuning of the output mass flow of the supercritical CO_2. Parameters of the capillary tubes were determined with the help of a one-dimensional numerical model for isothermal and adiabatic capillary tubes. Preliminary measurements proved functionality of the CO_2 branch. However, some further tests are required, especially, in order to clarify performance of the isothermal capillary tubes placed inside a thermostatic bath.

Acknowledgement

The research leading to these results has received funding from the Norwegian Financial Mechanism 2009–2014 under Project Contract no. 7F14466 and from the institutional support RVO:61388998.

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