Analysis of CO_2 Mass Transfer in Ionic Liquid–Water Mixtures by Thermophysical and Taylor Bubble Studies

Alberto Arici[1], **Felix Kexel[2]**, Friedrich Thießen[2], Michael Schlüter[2], Giorgio Besagni[1,3]

[1] Department of Energy, Politecnico di Milano, Milano, Italy
[2] Institute of Multiphase Flows, Hamburg University of Technology, Hamburg/Germany
[3] Institute of Fluid Dynamics, Helmholtz-Zentrum Dresden – Rossendorf, Dresden, Germany

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Ministero dell'Università e della Ricerca

FIS

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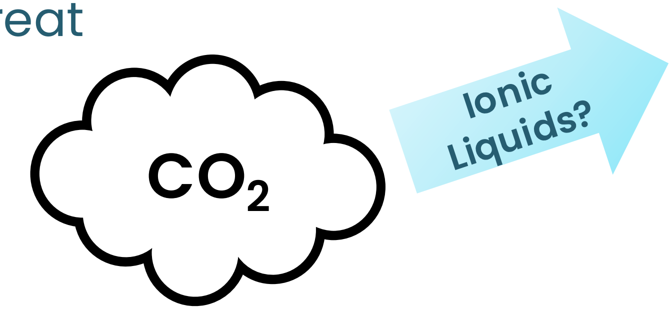
Motivation & Aim

- Large scale gas-liquid processes are of great importance for our everyday life
    - Chemical & Biotec industry
    - CO_2 Absorption & Carbon capture
    - Pharmaceutical industry
- Mass transfer often limits Reactor performance



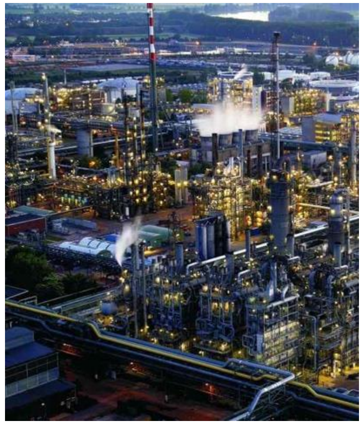
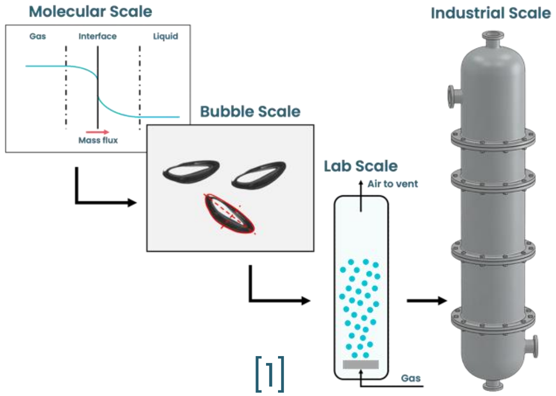


Image of an industrial site showing large scale chemical reactors (image BASF)

- Fluid properties influence bubble shape & velocity
- Bubble dynamics determine the wake structure and flow patterns, controlling local and global mixing
- Mixing directly affects mass transfer and reaction rates
- Chemical reaction effects fluid properties and mass transfer



[1]

[1] Besagni (2018)

Motivation & Aim

Molecular scale

Interaction between CO_2 and ionic species determine solubility, interface concentration and reaction pathways

Bubble scale

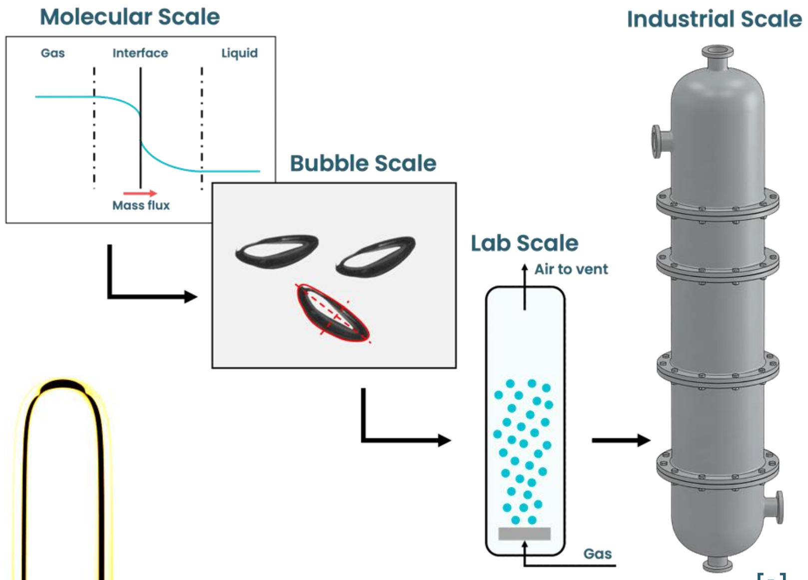
Fluid properties (viscosity, surface tension, density) influence bubble shape, velocity and film thickness

Macroscopic scales

Fluid dynamics control mixing and interfacial renewal determining the overall mass transfer rate

Taylor Bubbles

- Reduced complexity at high predictability and reproducibility
- Good accessibility for optical measurement techniques
- Reliable adjustment of fluid dynamic conditions
- High safety measures and small liquid volumes



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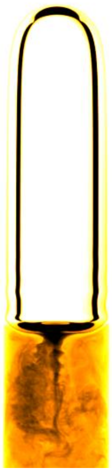

[2]

Taylor bubbles allow us to isolate the fundamental mechanisms governing mass transfer

[1] Besagni (2018)
[2] SPP 1740 (2023)

State of the Art – Taylor Bubbles

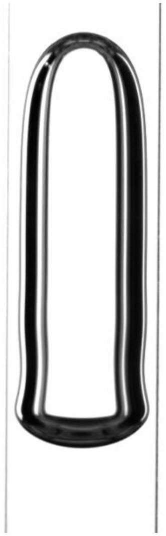
- Large, elongate bubbles in a confined space [3]
- Rise velocity v_b independent of bubble volume once Taylor bubble shape is developed [3, 4, 5, 6]
- Characterized by the Eötvös number

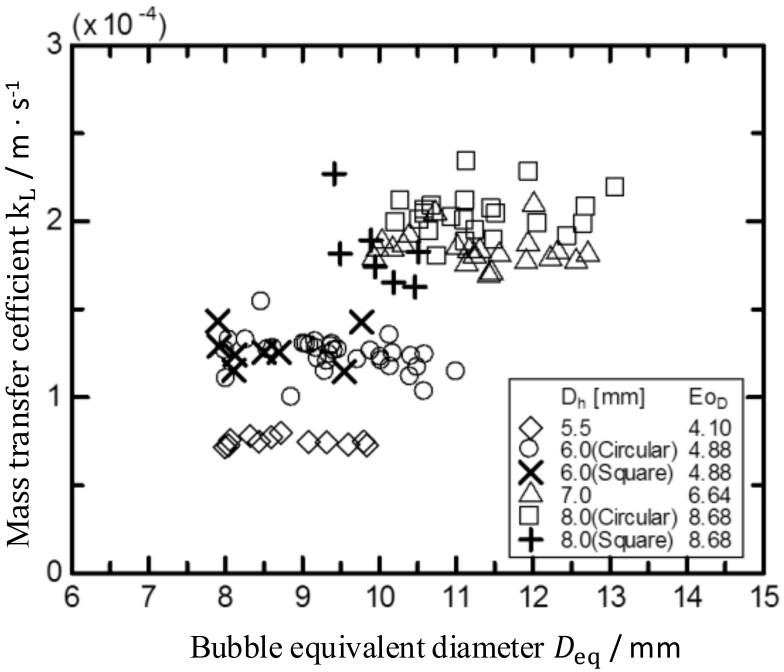
$$Eo = \frac{\Delta\rho g D_c^2}{\sigma} = \frac{\text{Buoyancy forces}}{\text{Surface tension}}$$

- Buoyancy driven if critical Eötvös number is exceeded [3]

$$Eo_{crit} \approx 4$$

- Ideal to study mass transfer over a time interval, enabling initial mass transfer studies





Data from Kastens et al. [6]

D_c	Capillary diameter / mm	
D_{eq}	Equivalent diameter / mm	
σ	Surface tension / N·m^{-1}	
ρ	Density / kg·m^{-3}	
g	Gravitation constant / m·s^{-2}	
η	Dynamic viscosity / Pa·s	
v_b	Rise velocity / m·s^{-1}	

[3] Taylor (1961)
[4] Kexel (2023)
[5] Hayashi (2011)
[6] Kastens (2015)

Why Ionic Liquids?

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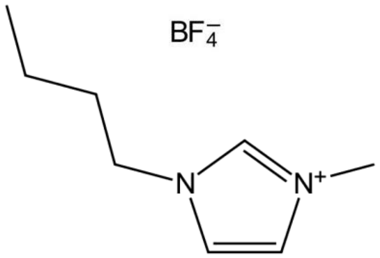
- Negligible vapor pressure and high chemical and thermal stability
  - ➢ prevents **solvent evaporation and loss**
- Tunable and wide physiochemical properties depending on cation-anion combination (about ~10^{14})
  - ➢ Physicochemical properties can be **systematically tailored**
  - ➢ Interesting to study influence on mass transfer

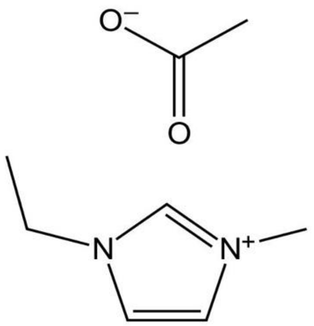
1-butyl-3-methylimidazolium tetrafluoroborate
[BMIM][BF4]

- Physical absorption
- Relatively low viscosity
- Well studied benchmark for CO_2 study



1-ethyl-3-methylimidazolium acetate
[EMIM][OAc]

- Physical/chemical absorption
- High viscosity
- Stronger CO_2 affinity



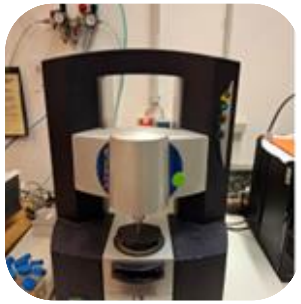
Determination of Thermophysical Properties

Experimental Parameters:

- 14 Binary mixtures of [BMIM][BF_4] and water
- Temperatures T = **293.15**, 298.15 & 303.15 K
- All experiments at atmospheric pressure

Viscosity

- *Kinexus PRO* rheometer
- Concentric cylinder
- Shear rate $\dot{\gamma} \in \{1, 100\}$ s^{-1}



Surface tension

- KRÜSS BP50 tensiometer
- Bubble pressure method
- Surface age 15-7500 ms

Density

- A. Paar DMA35 densimeter
- Mean values reported

Taylor Bubble Experiments

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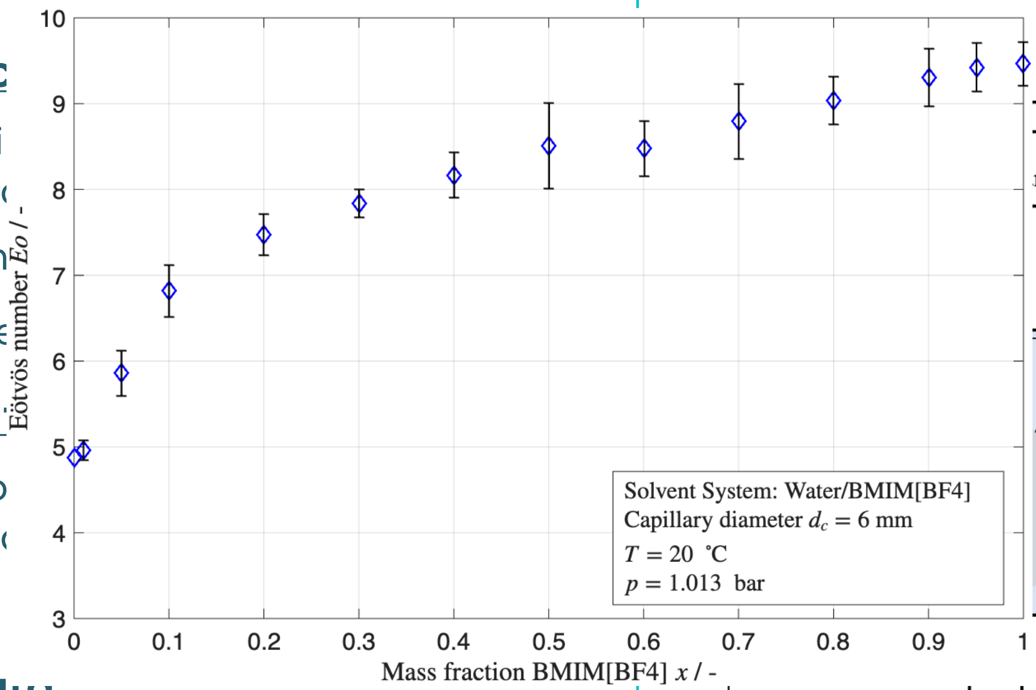
Experimental rac

- Nikon D7500 equi
  Planar 2/50M rec
- Background illum
- Capillary diamete

Fluid dynamic in

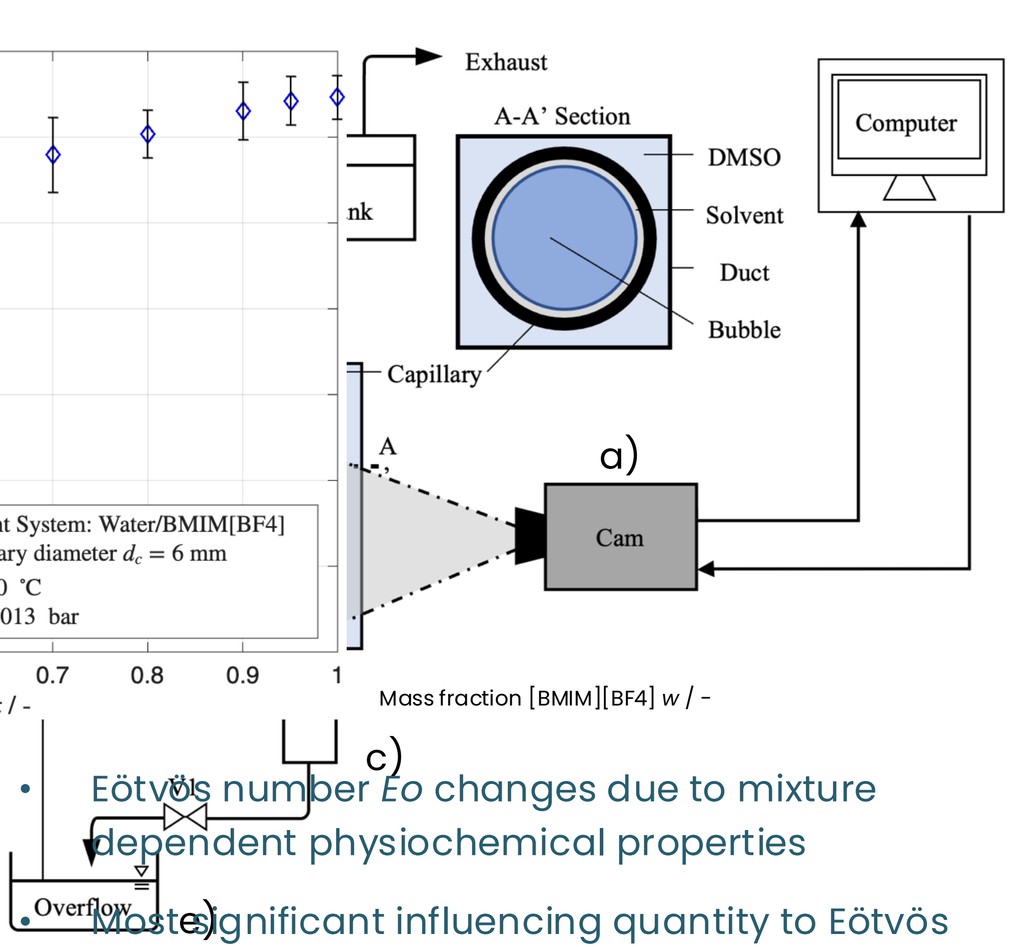
- Terminal rise velo
  centroid in stagn
- Air bubbles in air



Mass fraction [BMIM][BF4] *w* / -

Mass transfer data:

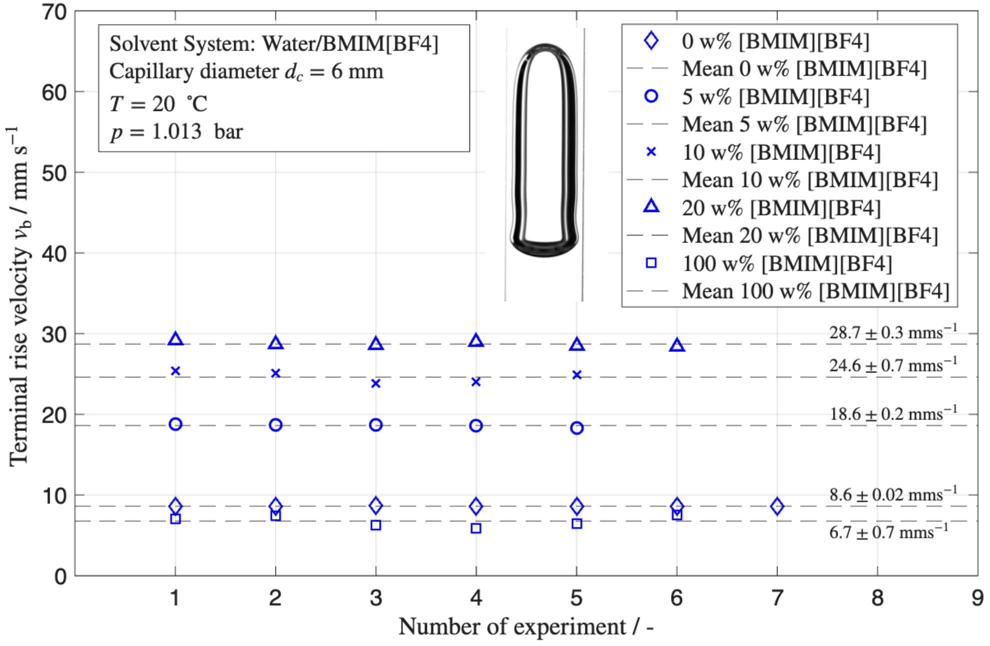
- Counter current flow adjusted by valve V1 to keep Taylor bubble at fixed position
- Mass transfer from the bubble shrinkage
- CO2 bubbles in IL/Water mixtures

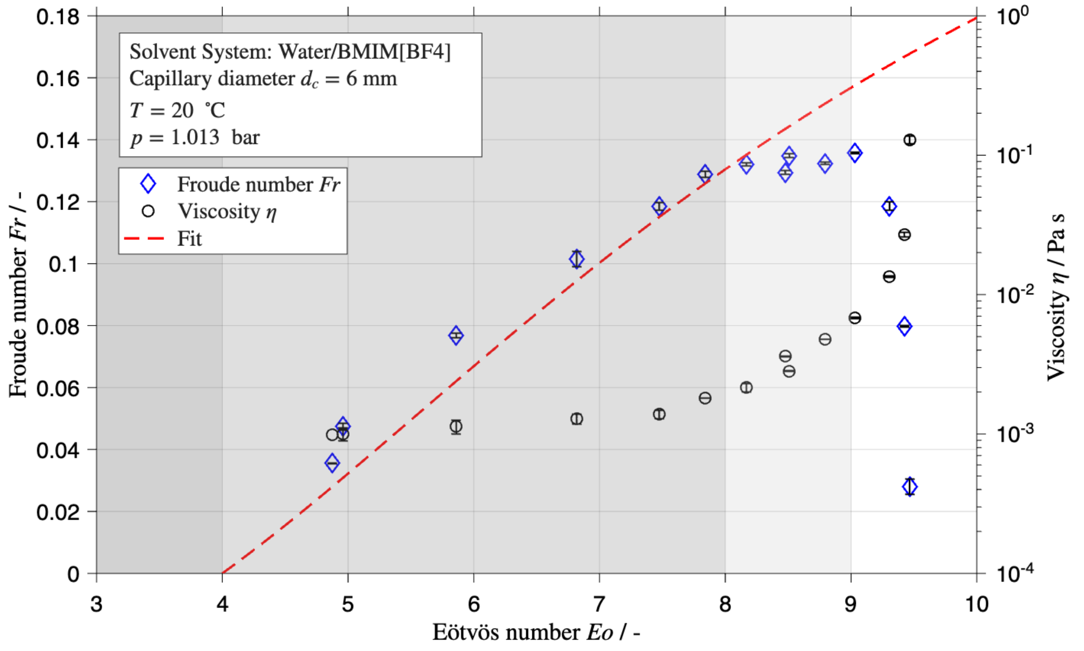


- Eötvös number *Eo* changes due to mixture dependent physiochemical properties
- Most significant influencing quantity to Eötvös number: Surface tension σ

Relevant components: a) SLR camera Nikon D7000; b) Upper glass tank; c) Glass capillary; d) LED backlight; e) Overflow tank

CO_2 Taylor Bubble Motion in IL/Water Mixtures





Dependencies of Bubble terminal rise velocity v_b:

- Independent of bubble volume
- Depending on [BMIM][BF4] mass fraction

Three Taylor bubble terminal rising states can be distinguished:

$$Fr = \frac{v_b}{\sqrt{g D_c}} = \frac{-0.033\left(1/Eo - 0.25\right)}{\left(1/Eo\right)^2 - 0.064/Eo + 0.024} \cdot H(Eo - Eo_{crit})$$ [4]

$Eo \leq 8$ or $w_{BMIM} \leq 0.5$: Froude follows own fit

$8 \leq Eo \leq 9$ or $0.5 \leq w_{BMIM} \leq 0.8$: Froude constant

$Eo > 9$ or $w_{BMIM} > 0.8$: Froude drops strongly

D_c	Capillary diameter / mm
v_b	Rise velocity / mm·s^{-1}
g	Gravitation constant / m·s^{-2}
Eo	Eötvös number /-
Eo_{crit}	Critical Eötvös number /-
Fr	Froude number / -
H	Heaviside step function
w	mass fraction /-

[4] Kexel (2023)

CO_2 Taylor Bubble Motion in IL/Water Mixtures

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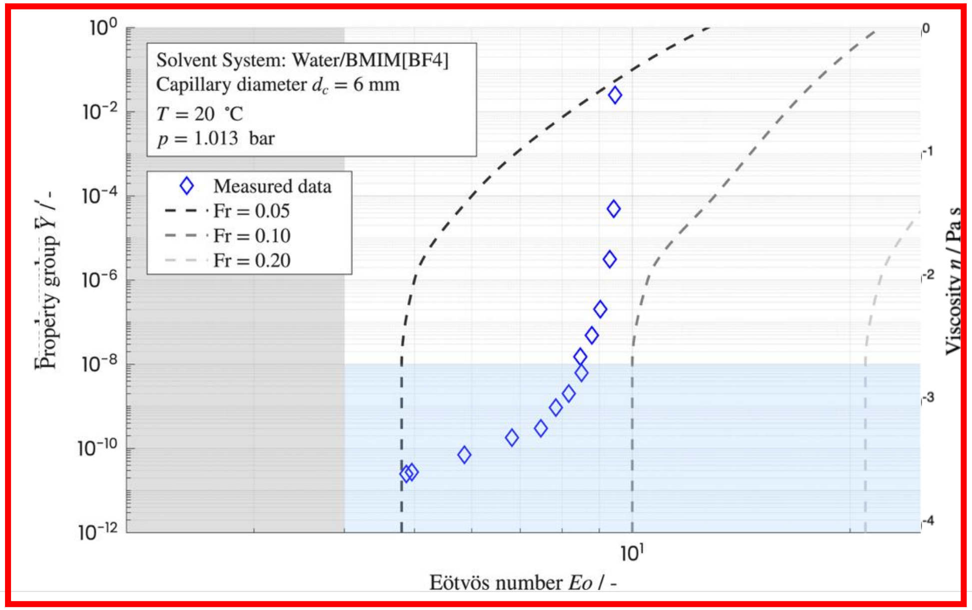
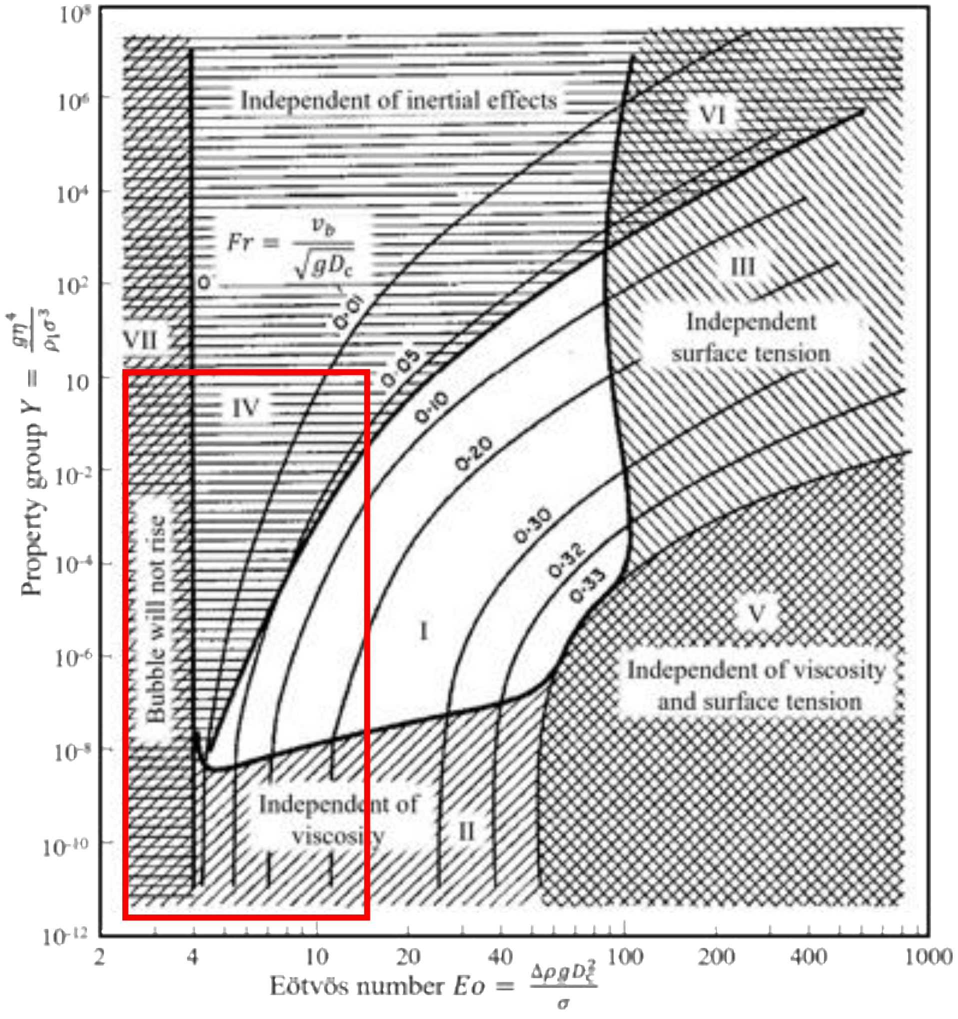


- Similar effects have been observed by White [7]
  - (I) No independencies (white)
  - (II) Independet of viscosity
  - (VII) No bubble rise (dark gray)
- Measured data shows sort of transitional regime (light gray)



[7] White (1962)

Determining Mass Transfer Coefficients from Taylor Bubbles

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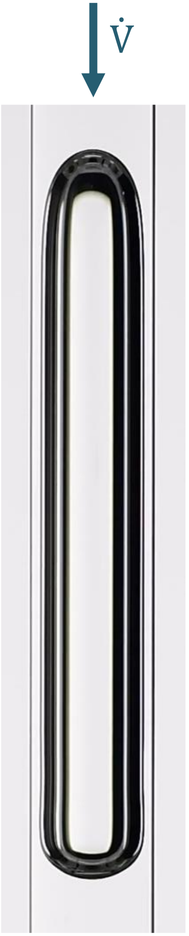


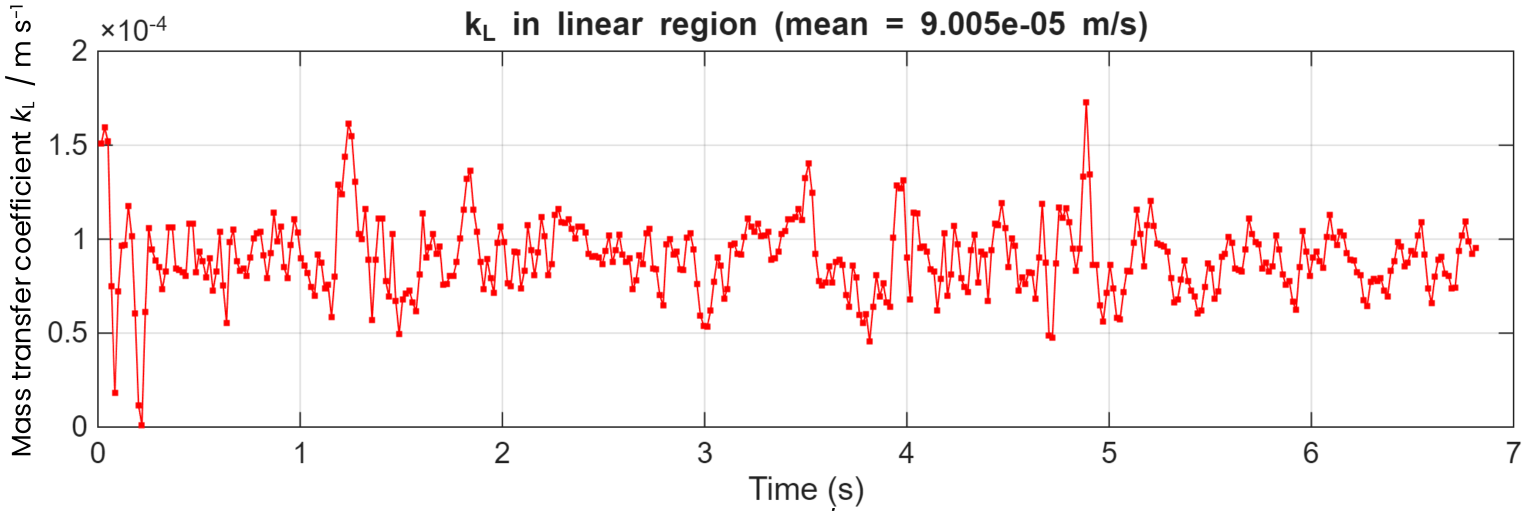
Real time recording showing a time span of Δt = 6.81 s

Determination of the mass transfer coefficient according to Hosoda [8]

$$k_L = \frac{H}{p C_V R T A} \frac{d}{dt}(p_B V_B)$$

Input for calculations:

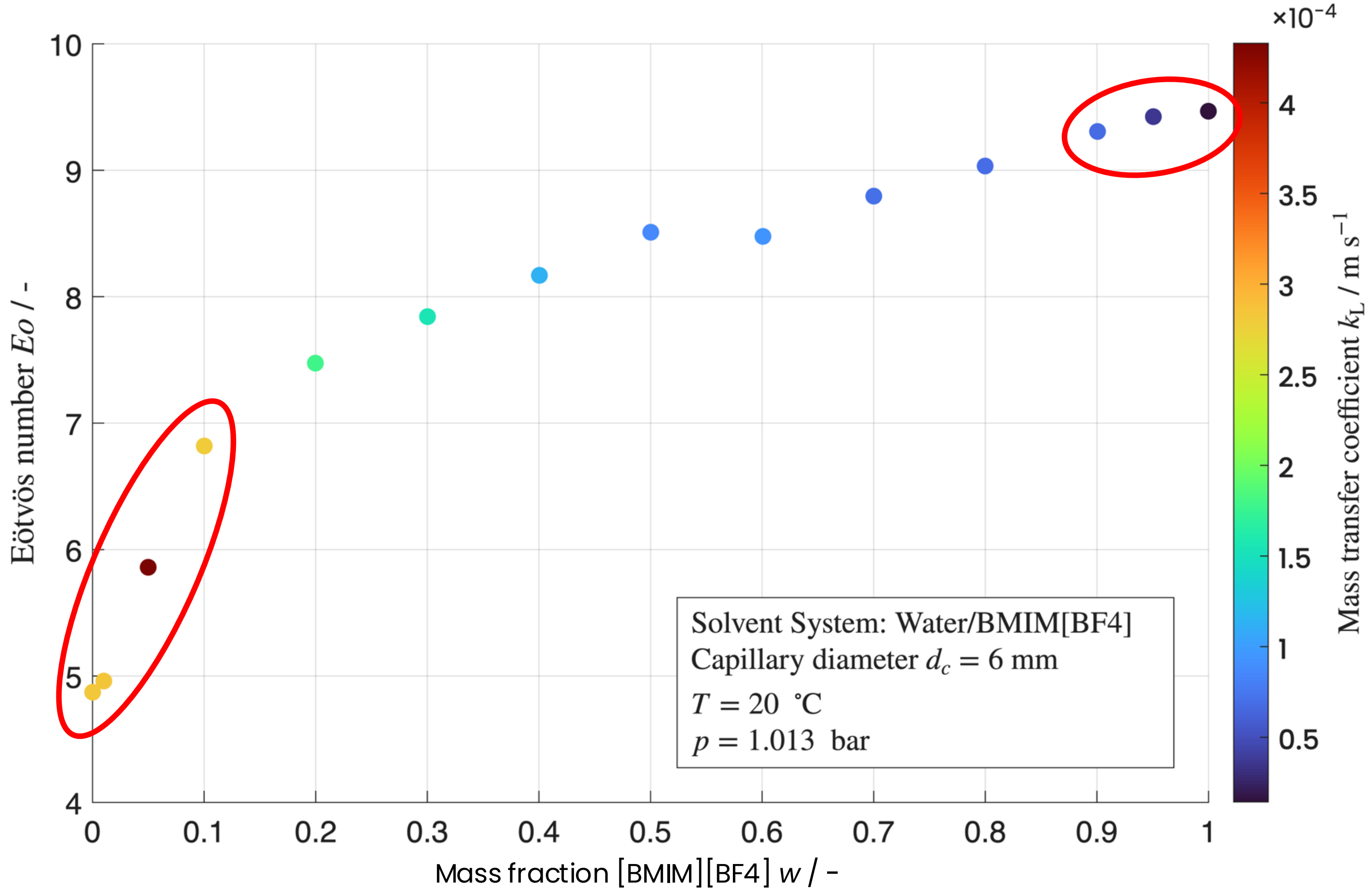
- Temperature T = 293.15 K
- Pressure*Volume (pV) from experiment
- Henry constant H for each mixture from Cosmo
- Analysis within linear time interval of volume change
- Area A of bubble reconstructed from images



A	Surface area / m²
p	Pressure / Pa
p_b	Bubble pressure / Pa
C_V	Mixture Molar concentration / mol L⁻¹
R	gas constant / J mol⁻¹ K⁻¹
V_B	Bubble volume / m³
T	Temperature / K
H	Henry constant /
k_L	Mass transfer coefficient / m s⁻¹

[8] Hosoda (2014)

CO2 Mass Transfer Coefficient in [BMIM][BF4]



CO2 Mass Transfer Coefficient in [BMIM][BF4]

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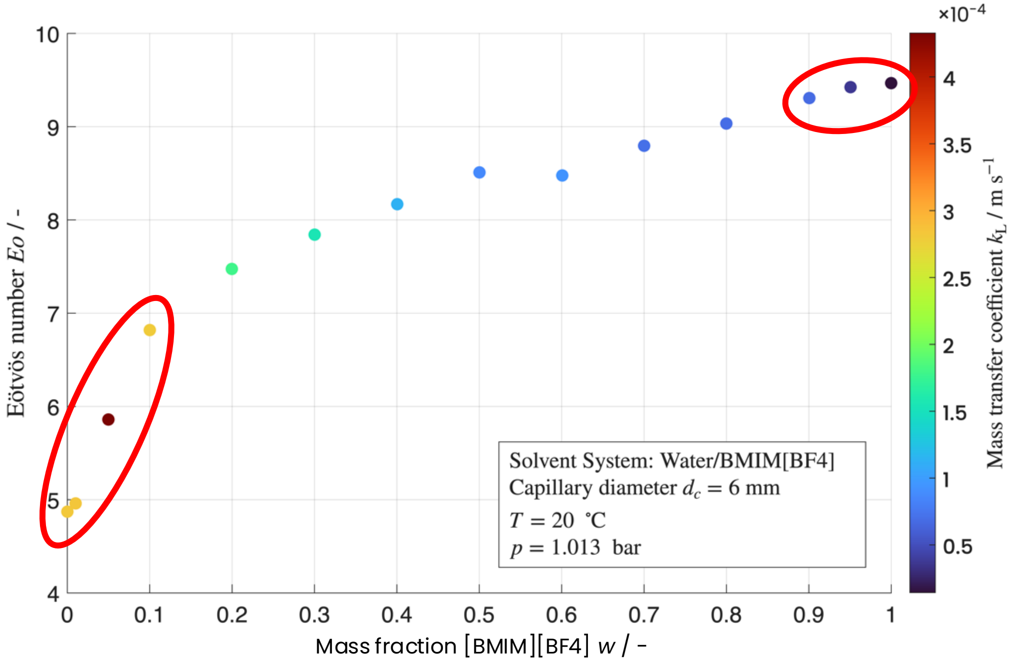
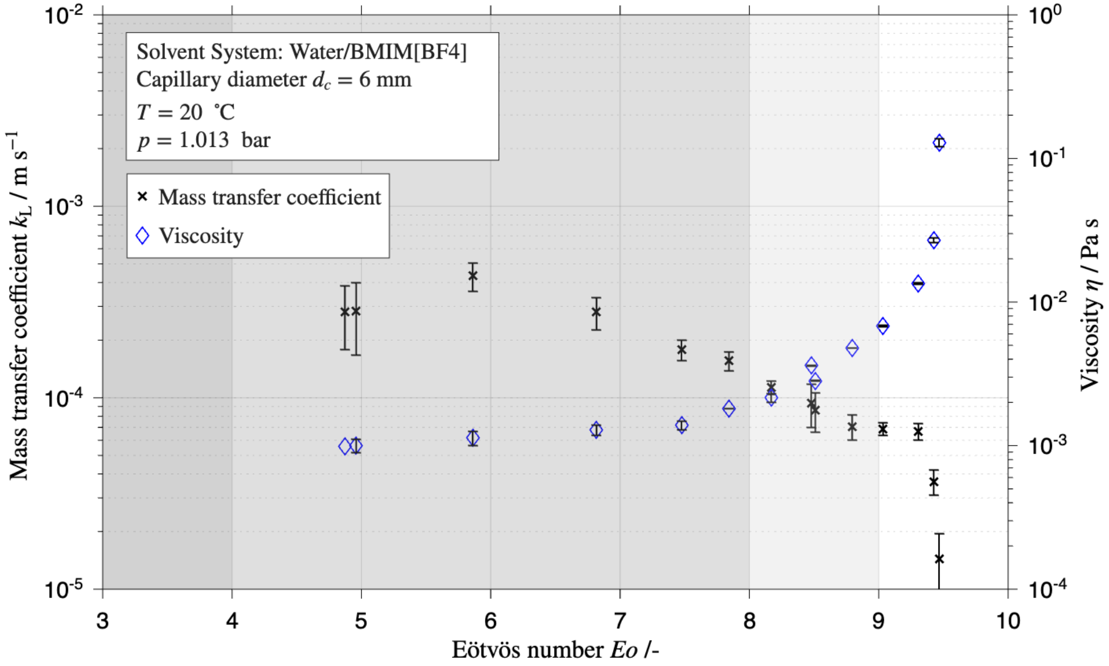


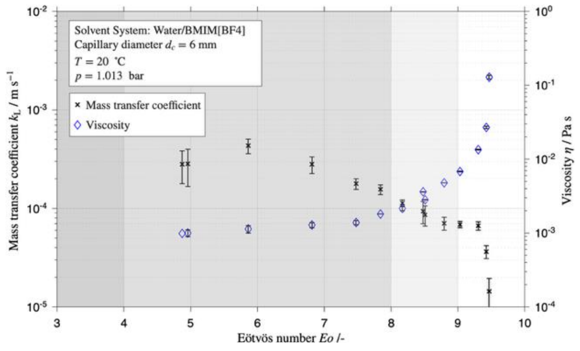


- Dependency of mass transfer to Eötvös number is limited to the individual liquid phase!
- Value of mass transfer seems to be closely connected to viscosity
- Low ionic liquid concentrations seem beneficial for higher k_L

Summary & Outlook

Physiochemical Properties of Ionic Liquids and bubble motion

- ✓ Besides CO_2 solubility (ongoing) all relevant parameters have been measured
- ✓ Surface tension & viscosity are identified as most influential parameters

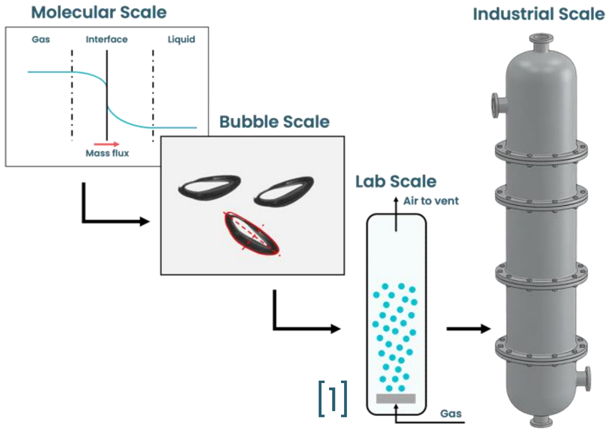


CO2 Mass Transfer of Taylor Bubbles in Ionic Liquids

- ✓ Mass transfer behavior is highly depending on mixture properties, especially viscosity
- ✓ There seems to be a local optima in mass transfer performance at low mass fractions of Ionic Liquid in water ($w_{\mathrm{BMIM}} < 10\ \%$)

Outlook & Pending Work

- ➢ Obtaining data for both Ionic Liquids (BMIM/EMIM)
- ➢ Determination of the Diffusion coefficient for all mixtures
- ➢ Connect CO_2 solubility in relevant IL/Water mixtures to the mass transfer
- ➢ Define an ideal operation point e.g., by defining a suitable cost function
- ➢ Transfer to larger scale experiments following the multiscale approach



[1] Besagni (2018)

Analysis of CO_2 Mass Transfer in Ionic Liquid–Water Mixtures by Thermophysical and Taylor Bubble Studies

Vielen Dank!

Fragen?

Dr.-Ing. Felix Kexel
felix.kexel@tuhh.de
040 42878 4663

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Research Foundati … rs)

This work has been … rsity and Research
(Ministero dell'Univ … . In particular, This
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Einladung und Aufruf zum Einreichen von Beiträgen

33. Fachtagung

Experimentelle Strömungsmechanik

Forschung, Entwicklung und Anwendung

8.-10 September 2026
Technische Universität Hamburg

Veranstalter

GALA

Deutsche Gesellschaft für Laser-Anemometrie GALA e.V.

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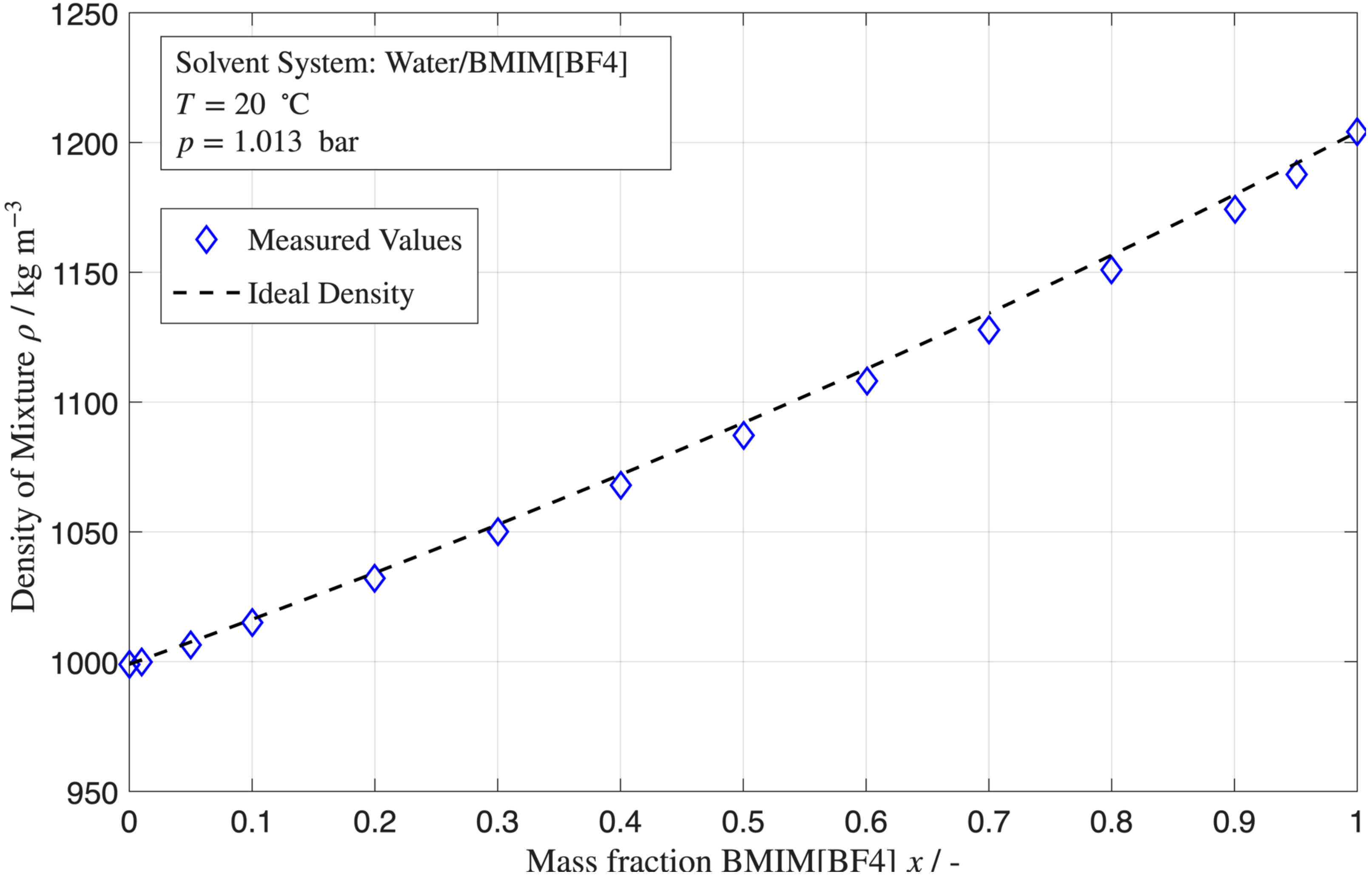
Density


Solvent System: Water/BMIM[BF4]
T = 20 °C
p = 1.013 bar
Measured Values
Ideal Density
Density of Mixture ρ / kg m−3
Mass fraction BMIM[BF4] x / -
1250
1200
1150
1100
1050
1000
950
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1