



Kent Academic Repository

Shao, Ding, Yan, Yong, Zhang, Wenbiao, Sun, Shijie, Sun, Caiying and Xu, Lijun (2019) *Dynamic measurement of gas volume fraction in a CO2 pipeline through capacitive sensing and data driven modelling*. *International Journal of Greenhouse Gas Control*, 94 . ISSN 1750-5836.

Downloaded from

<https://kar.kent.ac.uk/79507/> The University of Kent's Academic Repository KAR

The version of record is available from

<https://doi.org/10.1016/j.ijggc.2019.102950>

This document version

Author's Accepted Manuscript

DOI for this version

Licence for this version

CC BY-NC-ND (Attribution-NonCommercial-NoDerivatives)

Additional information

Versions of research works

Versions of Record

If this version is the version of record, it is the same as the published version available on the publisher's web site. Cite as the published version.

Author Accepted Manuscripts

If this document is identified as the Author Accepted Manuscript it is the version after peer review but before type setting, copy editing or publisher branding. Cite as Surname, Initial. (Year) 'Title of article'. To be published in *Title of Journal*, Volume and issue numbers [peer-reviewed accepted version]. Available at: DOI or URL (Accessed: date).

Enquiries

If you have questions about this document contact ResearchSupport@kent.ac.uk. Please include the URL of the record in KAR. If you believe that your, or a third party's rights have been compromised through this document please see our [Take Down policy](https://www.kent.ac.uk/guides/kar-the-kent-academic-repository#policies) (available from <https://www.kent.ac.uk/guides/kar-the-kent-academic-repository#policies>).

Dynamic measurement of gas volume fraction in a CO₂ pipeline through capacitive sensing and data driven modelling

Ding Shao¹, Yong Yan^{2*}, Wenbiao Zhang¹, Shijie Sun³, Caiying Sun¹, Lijun Xu³

¹ School of Control and Computer Engineering, North China Electric Power University, Beijing 102206, P.R. China

² School of Engineering and Digital Arts, University of Kent, Canterbury, Kent CT2 7NT, U.K.

³ School of Instrumentation and Optoelectronic Engineering, Beihang University, Beijing 100191, P.R. China.

* Correspondent author: y.yan@kent.ac.uk

Abstract

Gas volume fraction (GVF) measurement of gas-liquid two-phase CO₂ flow is essential in the deployment of carbon capture and storage (CCS) technology. This paper presents a new method to measure the GVF of two-phase CO₂ flow using a 12-electrode capacitive sensor. Three data driven models, based on back-propagation neural network (BPNN), radial basis function neural network (RBFNN) and least-squares support vector machine (LS-SVM), respectively, are established using the capacitance data. In the data pre-processing stage, copula functions are applied to select feature variables and generate training datasets for the data driven models. Experiments were conducted on a CO₂ gas-liquid two-phase flow rig under steady-state flow conditions with the mass flowrate of liquid CO₂ ranging from 200 kg/h to 3100 kg/h and the GVF from 0% to 84%. Due to the flexible operations of the power generation utility with CCS capabilities, dynamic experiments with rapid changes in the GVF were also carried out on the test rig to evaluate the real-time performance of the data driven models. Measurement results under steady-state flow conditions demonstrate that the RBFNN yields relative errors within $\pm 7\%$ and outperforms the other two models. The results under dynamic flow conditions illustrate that the RBFNN can follow the rapid changes in the GVF with an error within $\pm 16\%$.

Keywords: Carbon capture and storage; Gas volume fraction; Two-phase CO₂ flow; Data driven models; Copula functions

Nomenclature			
τ	Kendall's correlation coefficient	h_g	Specific enthalpy of gas CO ₂ before being mixed (kJ/kg)
μ	Spearman's correlation coefficient	h_l	Specific enthalpy of liquid CO ₂ before being mixed (kJ/kg)
r	Pearson's correlation coefficient	h_g'	Specific enthalpy of gas CO ₂ in two-phase flow (kJ/kg)
α_0	Gas volume fraction without correction	h_l'	Specific enthalpy of liquid CO ₂ in two-phase flow (kJ/kg)
χ	Gas mass fraction	ρ_l	Density of liquid CO ₂ (kg/m ³)
α	Reference gas volume fraction	ρ_g	Density of gas CO ₂ (kg/m ³)
q_{mg}	Mass flowrate of gas CO ₂ before being mixed (kg/h)	C_{norm}	Normalized capacitance
q_{ml}	Mass flowrate of liquid CO ₂ before being mixed (kg/h)	C_l	Capacitance measured when the pipe is full of liquid CO ₂ (fF)
\dot{q}_{mg}	Mass flowrate of gas CO ₂ in two-phase flow (kg/h)	C_g	Capacitance measured when the pipe is full of gas CO ₂ (fF)
\dot{q}_{ml}	Mass flowrate of liquid CO ₂ in two-phase flow (kg/h)	C_i	Capacitance measured for two-phase CO ₂ (fF)

33

34 **1. Introduction**

35 Global warming and climate change due to greenhouse gas emissions impede the global economic
36 development. The excessive CO₂ emissions from fossil fuel fire power generation utilities is regarded
37 as the main cause of global warming. Recently, carbon capture and storage (CCS) technology has been
38 proposed and is being deployed as an effective approach to reduce the emissions of CO₂ from the power
39 generation (Leung et al., 2014; Kemper, 2015). Accurate measurement of CO₂ flow in pipelines is
40 crucial to economical and safe operations in the CCS process. However, accidental leakage from CO₂
41 pipelines or small changes of the environmental temperature will lead to a significant change in the
42 phase of CO₂, resulting in gas-liquid two-phase CO₂ flow (Wen et al., 2019; Zhang et al., 2018).
43 Impurities produced using different capture methods may also lead to changes in phase properties of
44 CO₂ flow (Nazeri et al., 2016; Proter et al., 2015). In addition, CCS facilities on fossil fuel fired power
45 plants need to be operated flexibly (Abdilahi et al., 2018, Zhang et al., 2018), such as frequent load
46 changes and rapid start-ups and shutdowns. Due to the complex characteristics of CO₂ flow, accurate
47 measurement of CO₂ fluid parameters is more challenging than other gas-liquid two-phase flows. As an
48 important parameter in two-phase CO₂ flow, the gas volume fraction (GVF) is required to determine
49 the single phase mass flowrate and average density. Therefore, the GVF measurement is essential to

50 monitor and optimize the operation of the CCS system. However, few studies on the GVF measurement
51 of CO₂ flow have been reported to date.

52 Several methods based on capacitance probes (Ji et al., 2014;), wire-mesh sensors (Olerni et al., 2013;
53 Bowden et al., 2017), radiation attenuation (Nazemi et al., 2016), optical fiber sensing (Ursenbacher et
54 al., 2004) and ultrasonic sensing (Chakraborty et al., 2009) have been proposed for the direct
55 measurement of the GVF of gas-liquid two-phase flow. In comparison with other measurement
56 instruments, the capacitive sensors have the advantages of low cost, fast response and non-invasiveness
57 (Sun et al., 2017; Sun et al., 2018). Multi-electrode capacitive sensors are often utilized in process
58 tomography to achieve flow pattern recognition and visual monitoring (Xie et al., 2006; Jiang et al.,
59 2009). However, unlike the flow pattern recognition or phase distribution reconstruction of two-phase
60 flow, reconstructed images are usually not required for GVF measurement. Moreover, due to the
61 complex characteristics of gas-liquid two-phase flow, it is difficult to develop a general method that is
62 suitable for all flow patterns.

63 In recent years, some flow instruments incorporating data driven models, such as artificial neural
64 networks (ANNs) and support vector machine (SVM), have been utilized to achieve GVF measurement
65 under two-phase flow conditions (Figueiredo et al., 2016; Wang et al., 2017; Peyvandi and Rad, 2017;
66 Wang et al., 2018). Data driven models are widely used to represent the hidden relationships in large,
67 complex and multivariate datasets using statistical learning techniques. Wang et al. (2017) proposed
68 several data driven models based on ANNs, SVM and genetic programming to measure both the GVF
69 and liquid mass flowrate of an air-water two-phase flow using Coriolis mass flowmeters (CMFs).
70 Although the mass flow measurement errors are mostly within $\pm 1\%$, the maximum error of the GVF is
71 still larger than 10%. Peyvandi and Rad (2017) developed an approach by combining gamma ray
72 attenuation with ANNs to measure the GVF of an air-oil-water three-phase flow. Figueiredo et al. (2016)
73 analyzed acoustic attenuation data from an air-oil-water three-phase flow and developed ANNs and
74 least squares support vector machine (LS-SVM) for the GVF measurement and flow pattern recognition.
75 For two-phase CO₂ flow, Wang et al. (2018) combined LS-SVM models and CMFs to measure the GVF
76 in horizontal and vertical pipelines. The relative errors of GVFs both in horizontal and vertical pipelines
77 are within $\pm 10\%$ when the GVFs are larger than 5%. Previous studies have demonstrated that data
78 driven models, especially ANNs and LS-SVM, combined with conventional sensors perform well in
79 the GVF measurement of gas-liquid two-phase flow.

80 Variable selection is a necessary pre-processing step in the development of data driven models in order
81 to obtain acceptable measurement accuracy. Properties of datasets, including correlation and
82 monotonicity, should be taken into account during data pre-processing. The Pearson's correlation
83 coefficient, which is commonly used to determine statistical dependence, only describes linear
84 dependence (Mu et al., 2018). Recently, copula functions have been used to measure the non-linear
85 dependence and tendency correlation between variables (Han et al., 2019; Karra and Mili, 2019). A
86 series of copula functions, including normal copula, t-copula and Clayton copula, are common choices
87 in the fields of economics, astronomy and meteorology (Mensi et al., 2016; Navarro, 2018; Kim et al.,
88 2019). In comparison to other linear correlation analysis, copula functions can describe both linear and
89 non-linear correlations. For the measurement of gas-liquid two-phase flow, correlations between the
90 sensor signals and two-phase flow parameters are usually non-linear and non-monotonic. Copula
91 functions are capable of providing a comprehensive description of such correlations.

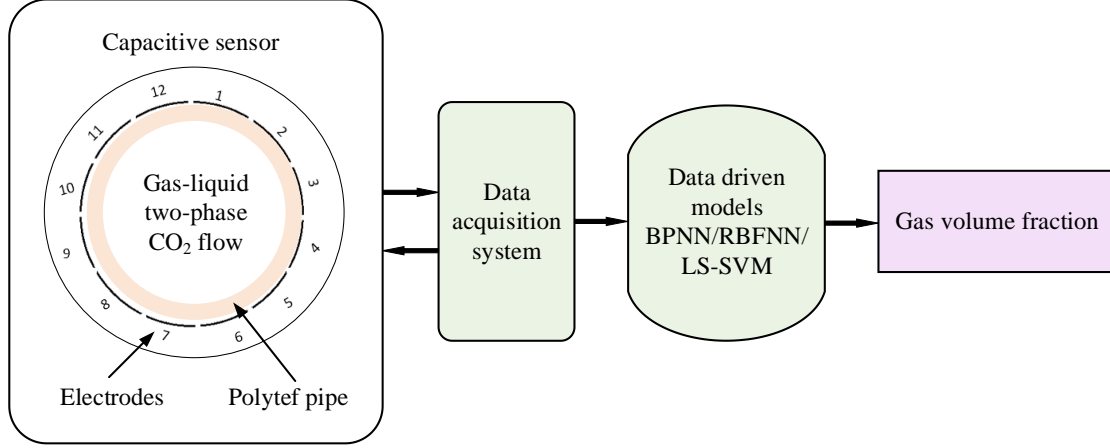
92 This paper presents a method for the GVF measurement of gas-liquid two-phase CO₂ flow by combining
93 a 12-electrode capacitive sensor and data driven models. Signals from the capacitive sensor are used to
94 develop the measurement models without going through a time-consuming image reconstruction
95 process. During the data pre-processing stage, copula functions are used to establish the non-linear
96 relationships and tendency correlations between the measured capacitance data and the GVF. Three data
97 driven models, including back-propagation neural network (BPNN), radial basis function neural
98 network (RBFNN) and LS-SVM, are established to measure the GVF. Experiments under steady-state
99 flow conditions were conducted on a horizontal pipeline on a CO₂ two-phase flow rig. The performance
100 of the proposed measurement models in this study is evaluated in terms of relative errors. In
101 consideration of the flexible operations of a power generation utility with CCS capability, dynamic
102 experiments with rapid changes in flow conditions were also conducted to assess the real-time
103 performance of the data driven models for GVF measurement.

104 **2. Methodology**

105 *2.1. Measurement strategy*

106 The measurement strategy adopted in this study is illustrated in Fig. 1. A high-pressure capacitive sensor
107 is designed and constructed, which consists of 12 identical rectangular electrodes with a length of 40
108 mm and a width of 7 mm. The electrodes are symmetrically mounted on the exterior of a polytef pipe
109 section with an inner diameter of 25 mm and an outer diameter of 31 mm. More details of the capacitive

110 sensor are available in Sun et al. (2018). A data acquisition system is developed to measure the
 111 capacitances between each pair of electrodes in the sensing head, resulting in a total of 66 (C_{12}^2)
 112 independent capacitances. Data driven models, including BPNN, RBFNN and LS-SVM, accept
 113 variables from the data acquisition system to infer the GVF of gas-liquid two-phase CO₂ flow.



114
 115 Fig. 1 Overall strategy for the GVF measurement of gas-liquid two-phase CO₂ flow

116 *2.2 Copula function*

117 Selecting an appropriate set of inputs to the data-driven model is a critical step in the GVF measurement.
 118 The relationship between the input variables and outputs of the data driven model is usually inferred
 119 through statistical analysis. However, the most widely used correlation coefficient in statistics can only
 120 analyze and measure the linear relationship between variables. In recent years, copula functions are
 121 proposed to describe the dependence of random variables more comprehensively (Han et al., 2019;
 122 Karra and Mili, 2019). The copula functions are powerful tools for modeling the non-linear correlation
 123 among multiple variables due to their ability of relating the marginal distribution function of each
 124 variable to their multivariate joint distributions functions. The distribution functions, including marginal
 125 distribution and joint distribution, describe completely the statistical regularity of random variables
 126 (Sun et al., 2019). Sklar theory states that a joint distribution can be divided into multiple marginal
 127 distributions and a copula function (Sklar, 1959), namely,

$$128 \quad H(x_1, x_2, \dots, x_n) = C\{F_1(x_1), F_2(x_2), \dots, F_n(x_n), \theta\} \quad (1)$$

129 where $H(x)$ is the joint distribution function of variables, $F(x)$ is the marginal distribution, θ is the
 130 correlation degree parameter which can be determined by using the non-parametric kernel density
 131 estimation method (Wang et al., 2014), and $C(\cdot)$ is the copula function.

132 Two families of copula functions, *Ellipse-copula* family and *Archimedean-copula* family, have been

133 applied to describe the relationship between variables. The *Ellipse-copula* family includes normal
 134 copula, t-copula and logit copula. Gumbel-copula, Clayton-copula and Frank-copula belong to the
 135 *Archimedean-copula* family. The forms of these copula functions can be found in Nelsen (2006). In this
 136 paper, a two-dimensional normal copula function is employed to take into account the correlation
 137 between each capacitance value and the GVF due to its low computational complexity. For a two-
 138 dimensional normal copula function, it can be described as follows:

$$139 \quad C(u, v) = \frac{1}{2\pi\sqrt{1-r^2}} \int_{-\infty}^{\Phi^{-1}(u)} \int_{-\infty}^{\Phi^{-1}(v)} \exp\left[\frac{-(s^2 - 2rst + t^2)}{2(1-r^2)}\right] ds dt \quad (2)$$

140 where u and v are two random variables, Φ^{-1} denotes the inverse function of a standard normal
 141 distribution function and r denotes the Pearson's correlation coefficient between u and v .

142 Unlike traditional linear correlation analysis, copula functions can describe both the linear and nonlinear
 143 correlations between variables. The most widely used scale-invariant measures of association are the
 144 Kendall's and Spearman's rank correlation coefficients (Fredricks and Nelsen, 2007), both of which
 145 can be calculated from copula functions:

$$146 \quad \tau = 4 \int_0^1 \int_0^1 C(u, v) dC(u, v) - 1 \quad (3)$$

$$147 \quad \mu = 12 \int_0^1 \int_0^1 C(u, v) duv - 3 \quad (4)$$

148 where τ and μ are the Kendall's and Spearman's correlation coefficients, respectively. The Kendall's
 149 and Spearman's coefficients are independent of the marginal distribution of random variables. They
 150 determine the degree of consistency and remain unchanged after a strict monotonic transformation,
 151 which illustrates that these coefficients have wider applicability than linear correlation coefficients.

152 2.3 Data driven models

153 In recent years, data driven modeling techniques were proposed for the GVF and flowrate measurement
 154 of gas-liquid two-phase flow (Wang et al., 2019; Peyvandi and Rad, 2017). Among these data driven
 155 models, the BPNN, RBFNN and LS-SVM have been widely used as alternatives to physical-based and
 156 conceptual models. The structure of each data driven model based on BPNN, RBFNN and LS-SVM is
 157 explained in detail in this section.

158 2.3.1 BPNN

159 As one of the most common neural networks, BPNNs have been applied to achieve the measurement
 160 of gas-liquid two-phase flow due to their strong nonlinear mapping capability, good adaptability and

161 fault tolerance (Azizi et al., 2015). The BPNN is a multilayer neural network consisting of an input
 162 layer, an output layer and a hidden layer, as shown in Fig. 2. The output of the BPNN is calculated from:

$$163 \quad y_{BP}(t) = \sum_{j=1}^L \omega_j H_j(t) + b \quad (5)$$

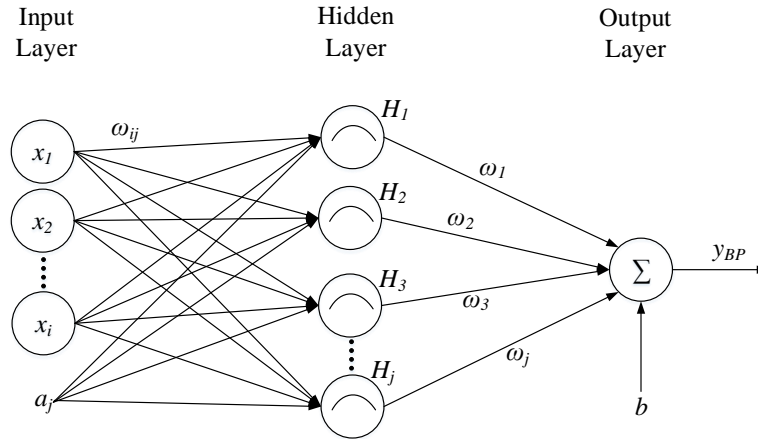
164 where ω_j and b are the connection weight and bias between the j th hidden neuron and the output layer,
 165 respectively. H_j is the output of the j th hidden neurons and is determined from:

$$166 \quad H_j(t) = f\left(\sum_{i=1}^n \omega_{ij} x_i(t) + a_j\right) \quad (6)$$

167 where ω_{ij} is the connection weight between the i th input neuron and the j th hidden neuron. x_i is the i th
 168 input variable and a_j is the bias of j th hidden neuron. $f(x)$ is the activation function of hidden neurons.
 169 In this paper, the hyperbolic tangent sigmoid function is used as an activation function of hidden neurons
 170 (Figueiredo et al., 2016) and presented by

$$171 \quad f(x) = \frac{2}{1 + e^{-2x}} - 1 \quad (7)$$

172 Although BPNNs have been widely applied in practice, however, a successful BPNN model depends
 173 significantly on the user-dependent parameters such as an appropriate model structure and training
 174 initialization.



175
 176 Fig. 2 Structure of a typical BPNN

177 2.3.2 RBFNN

178 RBFNN is a feedforward neural network consisting of three layers, as illustrated in Fig. 3, and uses a
 179 type of radial basis function (RBF) as activation to the hidden nodes. The output of the network is a
 180 linear combination of RBFs of the inputs and neuron parameters. The RBF measures the distance
 181 between the input vectors and the weight vectors and is typically taken to be the Gaussian function. The

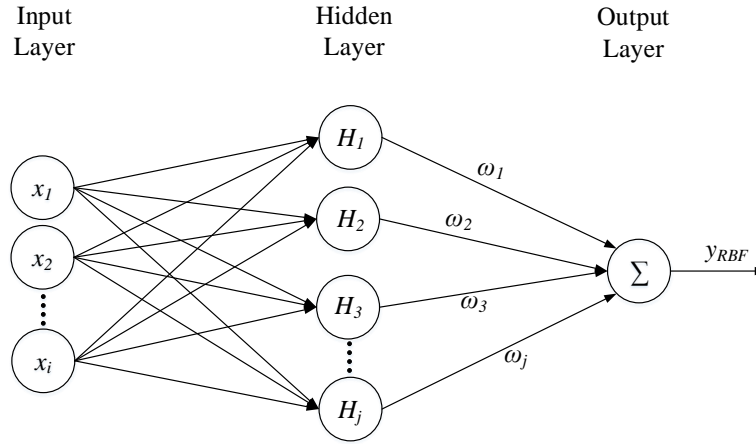
182 output of a RBFNN is calculated from:

$$183 \quad y_{RBF}(t) = \sum_{j=1}^N \omega_j(t) \varphi_j(x(t)) \quad (8)$$

184 where ω_j is the connection weight between the j th hidden neurons and the output layer, $x(t)$ is the input
 185 variables vector, and N is the number of hidden neurons. φ_j is the j th nonlinear mapping between the
 186 input neurons and the j th hidden neuron, respectively, namely,

$$187 \quad \varphi_j(x(t)) = \exp\left(-\left(\|x(t) - C_j\|^2 / 2\sigma_j^2\right)\right) \quad (9)$$

188 where C_j and σ_j^2 is the center vector and the variance for the j th hidden neuron, respectively, and C_j is
 189 determined by the K-means clustering method (Liao, 2010).



190
 191 Fig. 3 Structure of RBFNN

192 2.3.3 LS-SVM

193 The SVM algorithm maps linear inseparable data to a new space, in which these data become linearly
 194 separable. The SVMs have been applied to achieve the phase fraction prediction and flow regime
 195 identification due to the good generalization and the suitability for small sample training (Wang et al.,
 196 2009; Zhang et al., 2011). In order to achieve faster convergence, Suykens (2002) proposed an LS-SVM
 197 model to solve the nonlinear regression problem by mapping the data into a high-dimensional feature
 198 space and then developing a linear regression model in this space. Given training samples x and the
 199 desired output y , the LS-SVM model is defined as

$$200 \quad \min_{\omega, b, e} \frac{1}{2} \omega^T \omega + \frac{1}{2} \gamma \sum_{k=1}^n e \quad (10)$$

s.t.

$$201 \quad y = \omega^T \varphi(x) + b + e$$

201 where ω^T and b are the transposed vector and bias, respectively. $\varphi(x)$ is a nonlinear mapping function.

202 γ refers to penalty parameter. e_k is slack variables. x_k and y_k are the k th input and output elements,
 203 respectively.

204 In order to achieve the optimization solution of Eq. 10, Lagrange function is adopted, i.e.

$$205 \quad L(\omega, b, e, a) = \frac{1}{2} \omega^T \omega + \frac{1}{2} \gamma \sum_{k=1}^n e_k^2 - \sum_{k=1}^n a_k (\omega^T \varphi(x_k) + b - e_k - y_k) \quad (11)$$

206 where a_k is the Lagrange multiplier.

207 Furthermore, the partial derivative of Lagrange function is given by:

$$208 \quad \begin{cases} \frac{\partial L}{\partial \omega} = 0 \rightarrow \omega = \sum_{k=1}^n a_k \varphi(x_k) \\ \frac{\partial L}{\partial b} = 0 \rightarrow \sum_{k=1}^n a_k = 0 \\ \frac{\partial L}{\partial e_k} = 0 \rightarrow a_k = \gamma e_k \\ \frac{\partial L}{\partial a_k} = 0 \rightarrow y_k = \omega^T \varphi(x_k) + b + e_k \end{cases} \quad (12)$$

209 The estimation of the LS-SVM model is obtained by solving the following equation,

$$210 \quad y_k = \sum_{k=1}^n a_k K(x, x_k) + b \quad (13)$$

211 where $K(x, x_k)$ is a kernel function.

212 Samples from the original space are mapped into a higher-dimensional space by the kernel functions,
 213 such as linear, polynomial, RBF, and sigmoid function. In this study, an RBF kernel function is chosen
 214 due to the strong nonlinear mapping abilities. Thus, the output of the LS-SVM model is finally
 215 represented as:

$$216 \quad y_{SVM}(t) = \sum_{k=1}^n a_k \exp\left(-\frac{1}{2\sigma^2} \|x(t) - x_i\|^2\right) + b \quad (14)$$

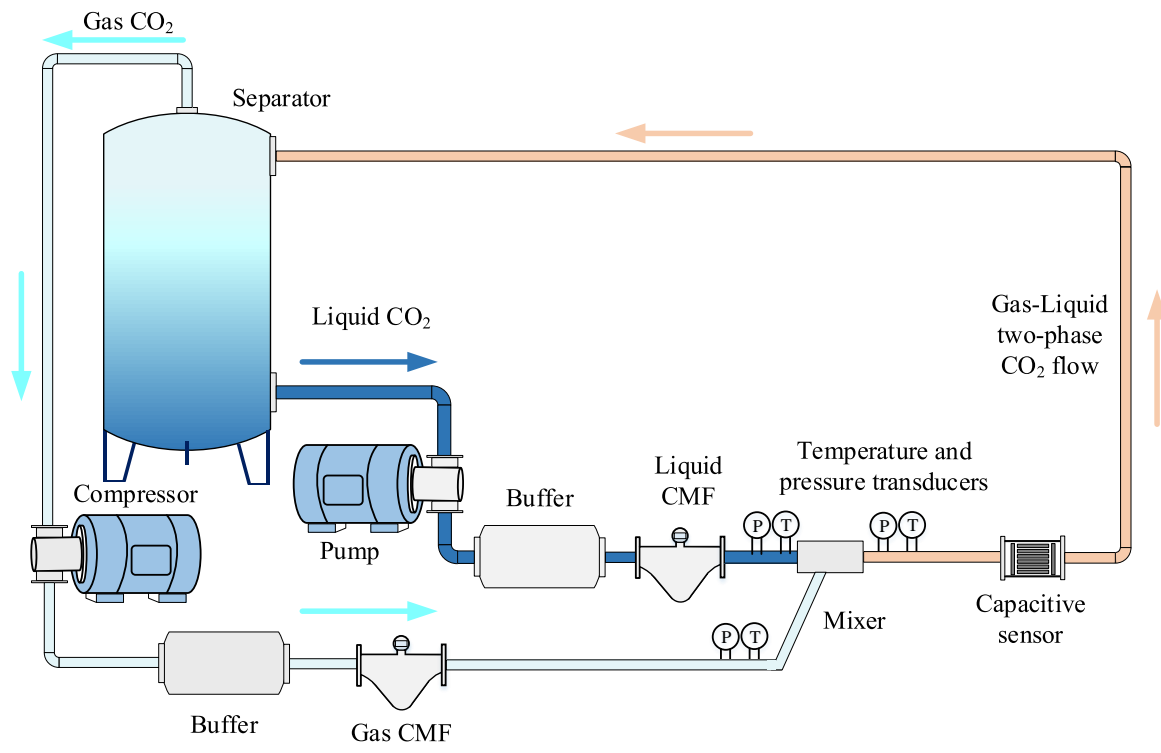
217 3. Experimental conditions

218 3.1 Test rig

219 Experiments were conducted on a 1-in bore gas-liquid two-phase CO₂ flow rig as depicted in Fig. 4.

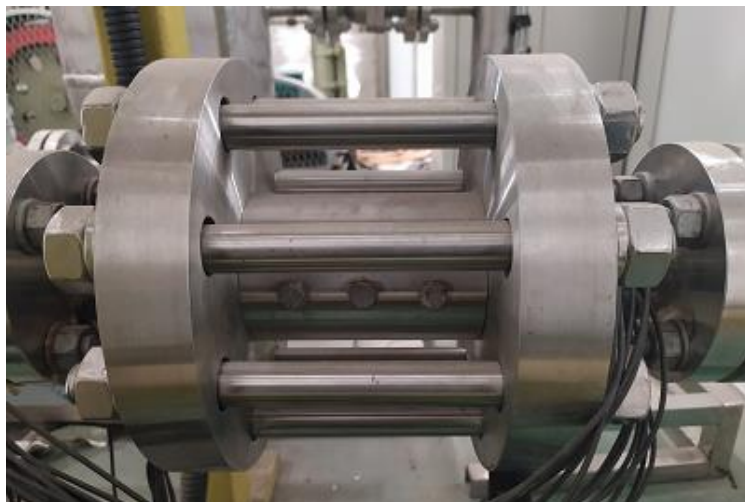
220 The capacitive sensor, as shown in Fig. 5, is installed in the horizontal test section. A stainless steel pipe
 221 is wrapped outside the polytef pipe to withstand the high pressure. For experiments under steady-state
 222 flow conditions, the mass flowrate of liquid CO₂ is set from 200 kg/h to 3100 kg/h, resulting in the
 223 reference GVF from 0% to 84%. Experiments under dynamic conditions were also conducted to
 224 investigate the real-time performance of the established data driven models. The gas phase CO₂ was

225 increased from 120 kg/h to 400 kg/h and then decreased from 400 kg/h to 120 kg/h when the liquid
 226 phase CO₂ was fixed at 1500 kg/h. Meanwhile, the liquid phase CO₂ was increased from 350 kg/h to
 227 750 kg/h and then decreased from 750 kg/h to 350 kg/h when the gas phase CO₂ was fixed at 70 kg/h.
 228 These dynamic flow conditions result in variations in GVF. The material properties and operation
 229 conditions of the CO₂ flow test rig are summarized in Table 1.



230
 231

Fig. 4 Schematic of the gas-liquid two-phase CO₂ flow rig



232
 233
 234
 235

Fig. 5 Photograph of the capacitive sensor

Table 1 Material properties and operation conditions of the CO₂ flow test rig

Parameter	Value	Parameter	Value
Pressure (bar)	57 - 72	Liquid density (kg/m ³)	740 - 800
Temperature (°C)	20 - 30	Gas specific enthalpy (kJ/kg)	403.26 - 437.74
Gas mass flowrate (kg/h)	15 - 400	Liquid specific enthalpy (kJ/kg)	253.25 - 262.93
Liquid mass flowrate (kg/h)	70 - 3100	Gas permittivity	1.0
Gas density (kg/m ³)	190 - 210	Liquid permittivity	1.6

237

238 *3.2 Calculation of reference GVF*

239 Two CMFs are installed on the liquid phase and gas phase sections, respectively, to provide the mass
 240 flowrate and density of single-phase CO₂ flow. The measurement uncertainty of gas phase CO₂ flow is
 241 0.35% while that of liquid phase CO₂ flow is 0.16%. Temperature and pressure transducers are installed
 242 at the entrance and exit of the mixer, respectively, to provide temperature and pressure information of
 243 single-phase and two-phase CO₂ flows. When the gas and liquid CO₂ are mixed without considering
 244 the phase transition between the two, the GVF (α_0) of two-phase CO₂ flow is calculated from,

$$245 \quad \alpha_0 = \frac{q_{mg} \rho_l}{q_{ml} \rho_g + q_{mg} \rho_l} \quad (15)$$

246 where q_{mg} and ρ_g are the mass flowrate and density of gas phase CO₂, respectively. q_{ml} and ρ_l are the
 247 mass flowrate and density of liquid phase CO₂, respectively. These parameters are all obtained from the
 248 reference CMFs.

249 However, phase transition may occur due to the changes in temperature and pressure at the mixer. The
 250 GVF should be corrected by the first law of thermodynamics of an open system as follows:

$$251 \quad \begin{aligned} q_{mg} h_g + q_{ml} h_l &= q'_{mg} h'_g + q'_{ml} h'_l \\ q_{mg} + q_{ml} &= q'_{mg} + q'_{ml} \end{aligned} \quad (16)$$

252 where h_g and h_l are the specific enthalpy values of pure gas and liquid CO₂ before being mixed,
 253 respectively. h'_g and h'_l are the specific enthalpy values of gas and liquid CO₂ in two-phase flow,
 254 respectively. q'_{mg} and q'_{ml} are the mass flowrate of gas and liquid CO₂ in two-phase flow, respectively.

255 Specific enthalpy is a thermodynamic quantity equivalent to internal energy of a system plus the product
 256 of its pressure and volume. Temperatures and pressures at the entrance and exit of the mixer are used to
 257 determine the specific enthalpy of CO₂. The gas mass fraction (χ) and the reference GVF (α) are
 258 calculated as follows,

$$\chi = \frac{(h_l - h_l') - \frac{q_{ml}}{q_{ml} + q_{mg}}(h_l - h_g)}{h_g' - h_l'} \quad (17)$$

$$\alpha = \frac{\chi \rho_l}{(1 - \chi) \rho_g} \quad (18)$$

261 4. Results and Discussion

262 4.1 Correlation analysis by copula functions

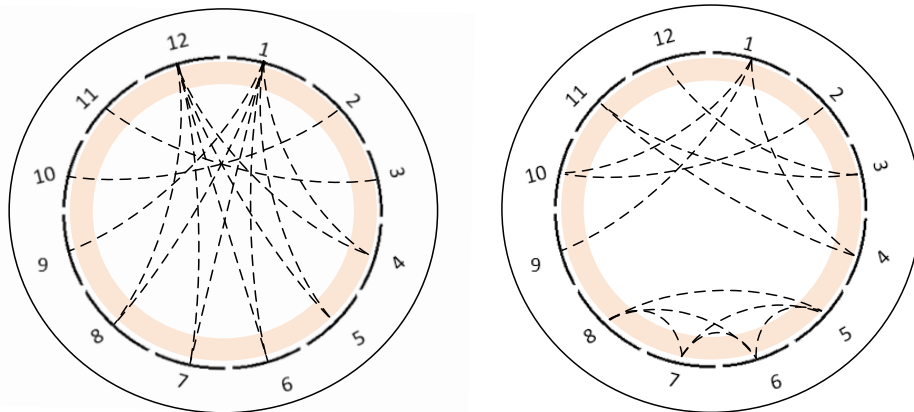
263 Firstly, the measured capacitances are normalized as follows,

$$C_{norm} = \frac{C_i - C_l}{C_g - C_l} \quad (19)$$

265 where C_i is the capacitance measured under two-phase CO₂ flow conditions. C_l and C_g are the
 266 capacitances which are measured during calibration when the pipeline is full of liquid phase CO₂ and
 267 gas phase CO₂, respectively.

268 Secondly, marginal probability density distributions for the capacitances of each pair of electrodes are
 269 estimated using the non-parametric kernel density estimation method. Joint distributions between the
 270 measured capacitances and GVF's are established by using normal copula functions and the semi-
 271 parametric pseudo-maximum-likelihood method. Finally, the Kendall's and Spearman's rank
 272 correlation coefficients between the capacitance values and the GVF's are calculated from Eq. 3 and Eq.
 273 4.

274 Tables 2 and 3 summarize the Kendall's and Spearman's correlation coefficients calculated from copula
 275 functions. The closer these coefficients get to 1 (or -1), the stronger the positive (or negative) correlation
 276 is. The top 20% variables with the strongest correlation are selected and their distributions are depicted
 277 in Fig. 6.



278

279

(a)

(b)

280 Fig. 6 Distributions of electrode pairs selected by different coefficients. (a) Kendall's coefficient (b)

281

Spearman's coefficient.

282 From Fig. 6 (a), electrodes selected by Kendall's coefficient are mainly distributed in the top and bottom

283 of the horizontal pipeline. The sensing area of these electrodes covers the entire interior of the pipe

284 which is good to obtain sufficient information about the flow. However, the sensing segments of the

285 electrode pairs selected by Spearman's coefficients are located at the edge of the pipeline as shown in

286 Fig. 6 (b). To obtain complete assessment for input variables, capacitances selected by both Kendall's

287 and Spearman's coefficients are used to develop data driven models. The performances of these models

288 are evaluated and compared in terms of relative errors.

289

Table 2 Kendall's coefficients between the GVs and capacitances

Number of electrodes	2	3	4	5	6	7	8	9	10	11	12
1	0.32	0.78	0.92	0.92	0.92	0.92	0.93	0.94	0.85	0.38	0.7
2	--	0.63	0.76	0.77	0.76	0.77	0.78	0.89	0.75	0.58	0.34
3	--	--	0.55	0.57	0.58	0.59	0.6	0.61	0.66	0.83	0.82
4	--	--	--	0.87	0.85	0.31	0.27	0.18	0.56	0.88	0.91
5	--	--	--	--	0.82	0.79	0.68	0.31	0.55	0.87	0.9
6	--	--	--	--	--	0.73	0.8	0.35	0.54	0.86	0.89
7	--	--	--	--	--	--	0.81	0.37	0.53	0.86	0.89
8	--	--	--	--	--	--	--	0.4	0.52	0.86	0.89
9	--	--	--	--	--	--	--	--	0.49	0.86	0.89
10	--	--	--	--	--	--	--	--	--	0.67	0.78
11	--	--	--	--	--	--	--	--	--	--	0.23

290

291

Table 3 Spearman's coefficients between the GVs and capacitances

Number of electrodes	2	3	4	5	6	7	8	9	10	11	12
1	0.3	0.69	0.86	0.75	0.75	0.76	0.78	0.79	0.82	0.22	0.77
2	--	0.49	0.72	0.72	0.72	0.73	0.74	0.75	0.78	0.46	0.19
3	--	--	0.27	0.31	0.33	0.35	0.37	0.38	0.47	0.79	0.78
4	--	--	--	0.63	0.6	0.56	0.52	0.42	0.31	0.78	0.72
5	--	--	--	--	0.88	0.87	0.84	0.56	0.29	0.77	0.7
6	--	--	--	--	--	0.88	0.87	0.6	0.27	0.76	0.69
7	--	--	--	--	--	--	0.86	0.64	0.25	0.76	0.68
8	--	--	--	--	--	--	--	0.67	0.22	0.76	0.68
9	--	--	--	--	--	--	--	--	0.17	0.76	0.69
10	--	--	--	--	--	--	--	--	--	0.52	0.68
11	--	--	--	--	--	--	--	--	--	--	0.15

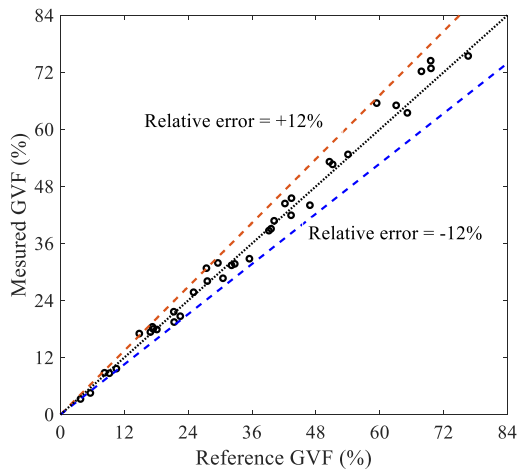
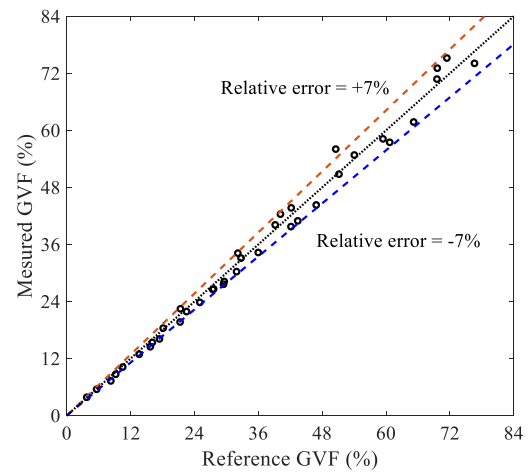
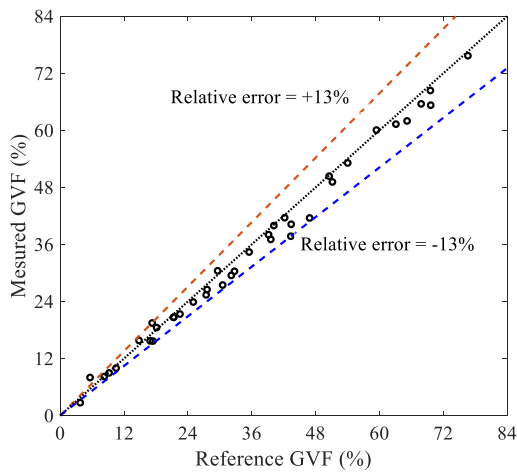
292

293 4.2 Performance of data driven models

294 4.2.1 Steady-State flow conditions

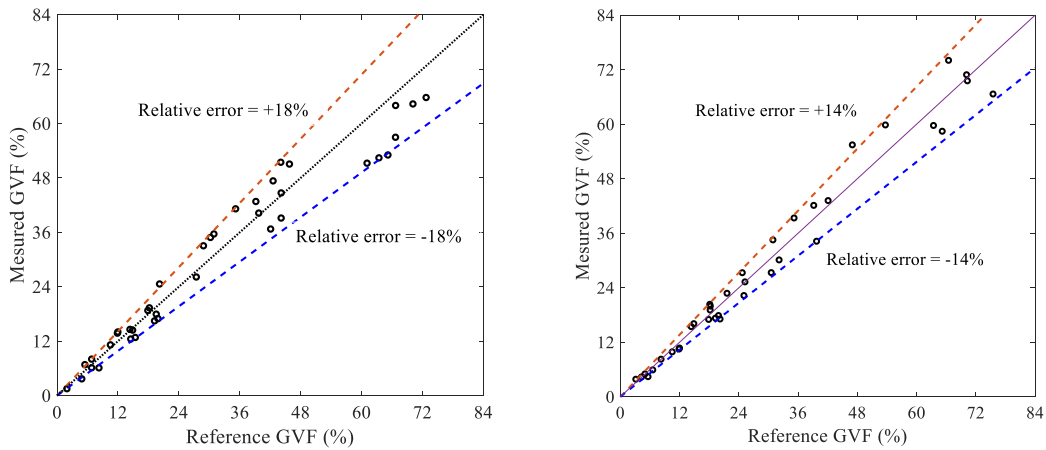
295 A total of 197 sets of the capacitance data under steady-state flow conditions were acquired as sample
296 data for training BPNN, RBFNN and LS-SVM, among which 158 sets (80% of the data) are adopted
297 as training data. The remaining 39 sets (20% of the data) are used as testing data. The three data driven
298 models are compared in terms of measurement accuracy.

299 Fig. 7 shows a comparison between the reference GVF and measured GVF from the data driven models
300 using variables selected using the Kendall's coefficient. The relative errors from the BPNN and LS-
301 SVM models are within $\pm 13\%$ and $\pm 12\%$, respectively, whilst the RBFNN model yields a relative error
302 within $\pm 7\%$. The RBFNN is remarkably more accurate than BPNN and LS-SVM due probably to the
303 fact that the K-means clustering of input variables during the training of the RBFNN model has similar
304 effect on the flow pattern classification of two-phase CO₂ flow.



309 Fig. 7 Comparison between the measured GVF and reference GVF using variables selected via the
310 Kendall's coefficient. (a) BPNN. (b) RBFNN. (c) LS-SVM.

311 Fig. 8 depicts the comparison between the reference GVF and measured GVF using variables selected
312 via the Spearman's coefficient. The relative errors of the BPNN, RBFNN and LS-SVM models are
313 within $\pm 18\%$, $\pm 13.8\%$ and $\pm 17\%$, respectively. The mathematical formulations of Kendall's and
314 Spearman's coefficients are different, resulting in different selected variables and outputs of the data
315 driven models. However, according to Fig. 7 and Fig. 8, both coefficients are effective in selecting input
316 variables for the data driven models. It is because that both coefficients are developed to determine the
317 degree of consistency, which means they are both effective in measuring the nonlinear correlation
318 between the measured capacitance data and GVF. However, the measurement area of electrode pairs
319 selected via Spearman's coefficient is mostly located at the edge of the pipeline, some information about
320 the fluid in the center of the pipeline may be lost, resulting in lower accuracy than that in Fig. 7 for all
321 three models.

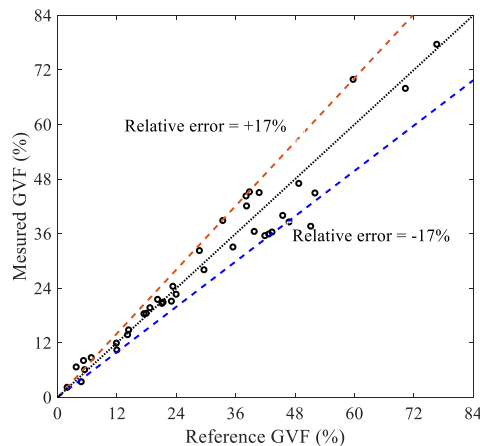


322

323

(a)

(b)



324

325

(c)

326

Fig. 8 Comparison between the measured GVF and reference GVF using variables selected via the

327

Spearman's coefficient. (a) BPNN. (b) RBFNN. (c) LS-SVM.

328

Fig. 9 and Fig. 10 illustrate the relative error histograms from the data driven models using the two

329

different variable selection methods. It is clear that the error distributions of the BPNN and RBF models

330

are much wider and dispersive than those of RBFNN. By comparing the relative errors and error

331

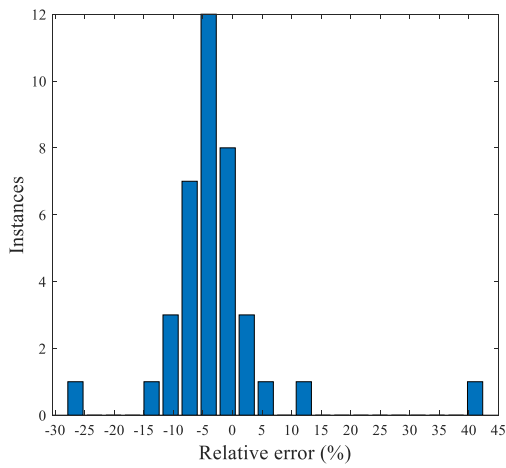
distributions from data driven models, we can conclude that the measurement results from RBFNN

332

incorporating copula functions produce the lowest relative errors and the highest concentration of the

333

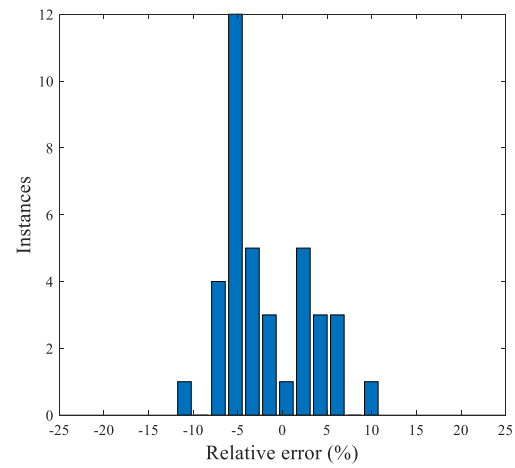
error distributions.



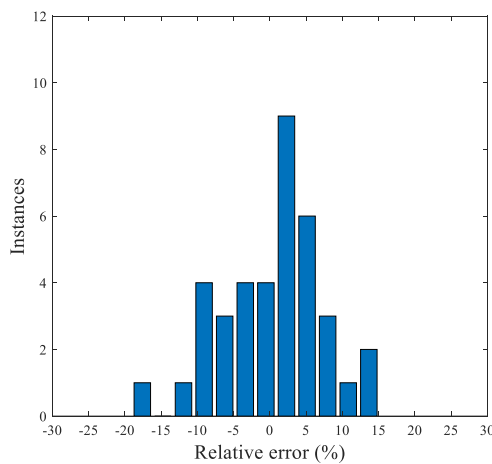
334

(a)

335



(b)



(c)

336

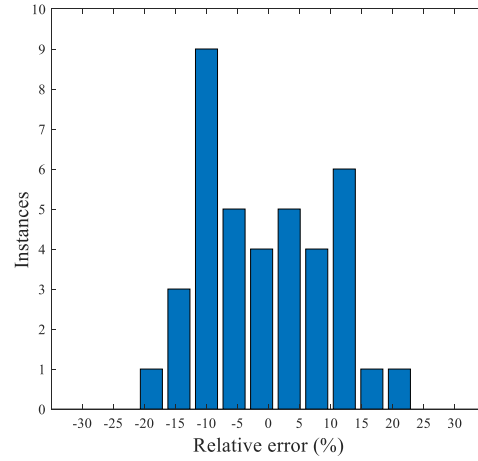
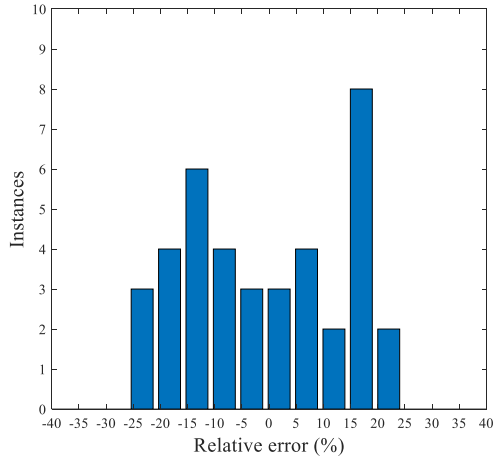
337

338

Fig. 9 Relative error histograms of BPNN, RBFNN and LS-SVM using variables selected by the

339

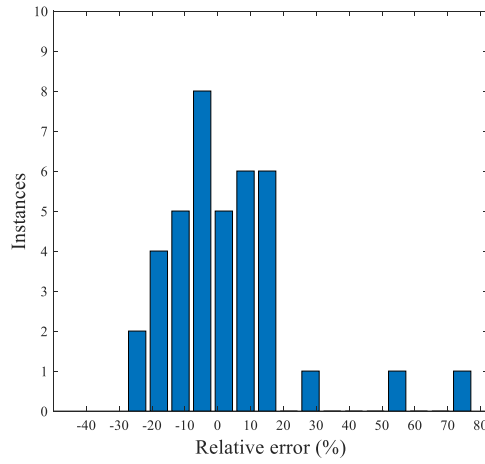
Kendall's coefficient. (a) BPNN. (b) RBFNN. (c) LS-SVM.



340
341

(a)

(b)



342
343

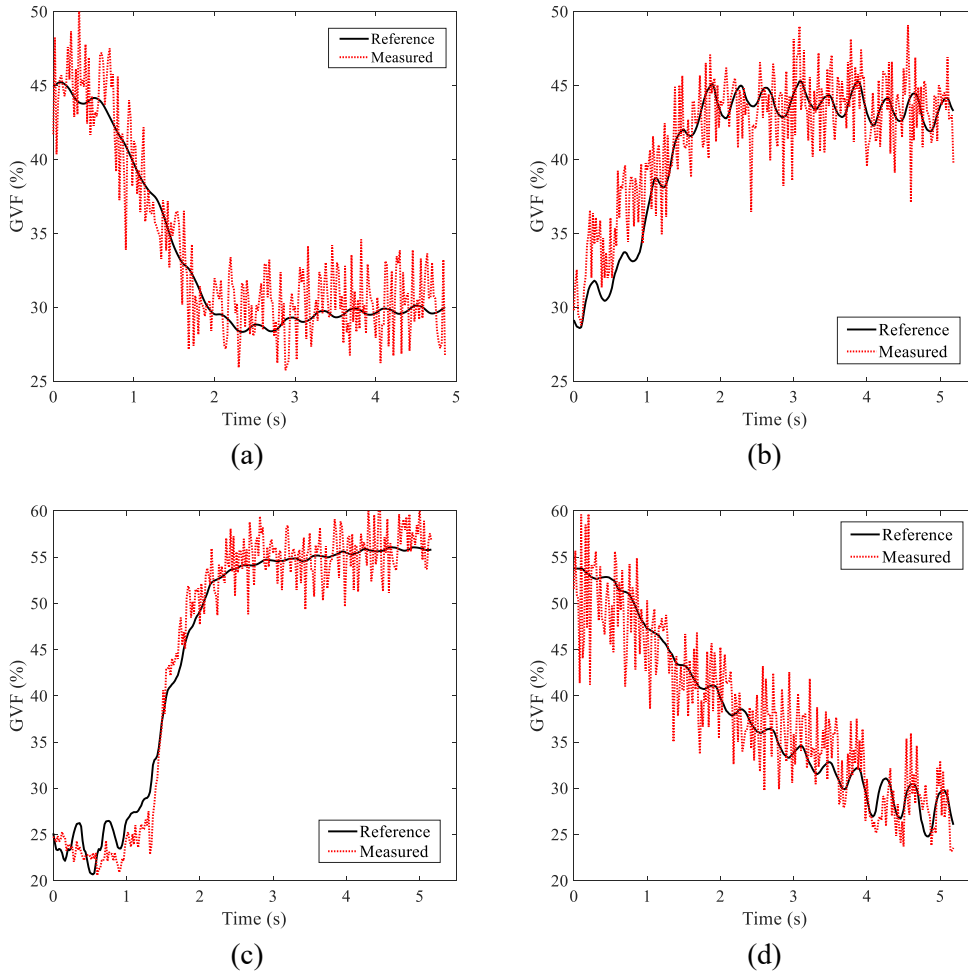
(c)

344 Fig. 10 Relative error histograms of BPNN, RBFNN and LS-SVM using variables selected by the
345 Spearman's coefficient. (a) BPNN. (b) RBFNN. (c) LS-SVM.

346 *4.2.2 Dynamic conditions*

347 The RBFNN, which outperforms the BPNN and LS-SVM under steady-state flow conditions, is applied
348 to achieve the GVF measurement under dynamic conditions. Fig. 11 shows the measurement
349 performance of the RBFNN during the dynamic operations in the horizontal test section. As shown in
350 Fig. 11 (a) and (b), the mass flowrate of gas phase was fixed at 70 kg/h while the mass flowrate of liquid
351 phase experienced the step increase and decrease. In comparison with the reference GVF, the measured
352 GVF can follow the transient changes. Fig. 11 (c) and (d) show the transient behaviours with increasing
353 and decreasing gas phase CO₂ while the liquid phase was fixed at 1500 kg/h. The measurement results
354 can also follow the trend of reference GVF. However, as shown in Fig. 11 (d), the reduction in GVF is
355 gradual because the inertia of the compressor at a reduced frequency is larger than that at an increased

356 frequency, which prevents step reduction in the mass flowrate of gas phase CO₂. Meanwhile, the buffer
 357 installed behind the compressor serves to keep the mass flowrate of gas CO₂ stable in the pipeline,
 358 which also prevents step change in the gas phase CO₂ under dynamic conditions. A comparison between
 359 the reference GVF and measured GVF during dynamic conditions is plotted in Fig. 12. It is clear that
 360 the relative errors are within $\pm 16\%$ in all four cases.



361
362

363
364

365 Fig. 11 Measurement performance of RBFNN under dynamic conditions. (a) Increasing liquid CO₂
 366 with fixed gas CO₂. (b) Decreasing liquid CO₂ with fixed gas CO₂. (c) Increasing gas CO₂ with fixed
 367 liquid CO₂. (d) Decreasing gas CO₂ with fixed liquid CO₂.

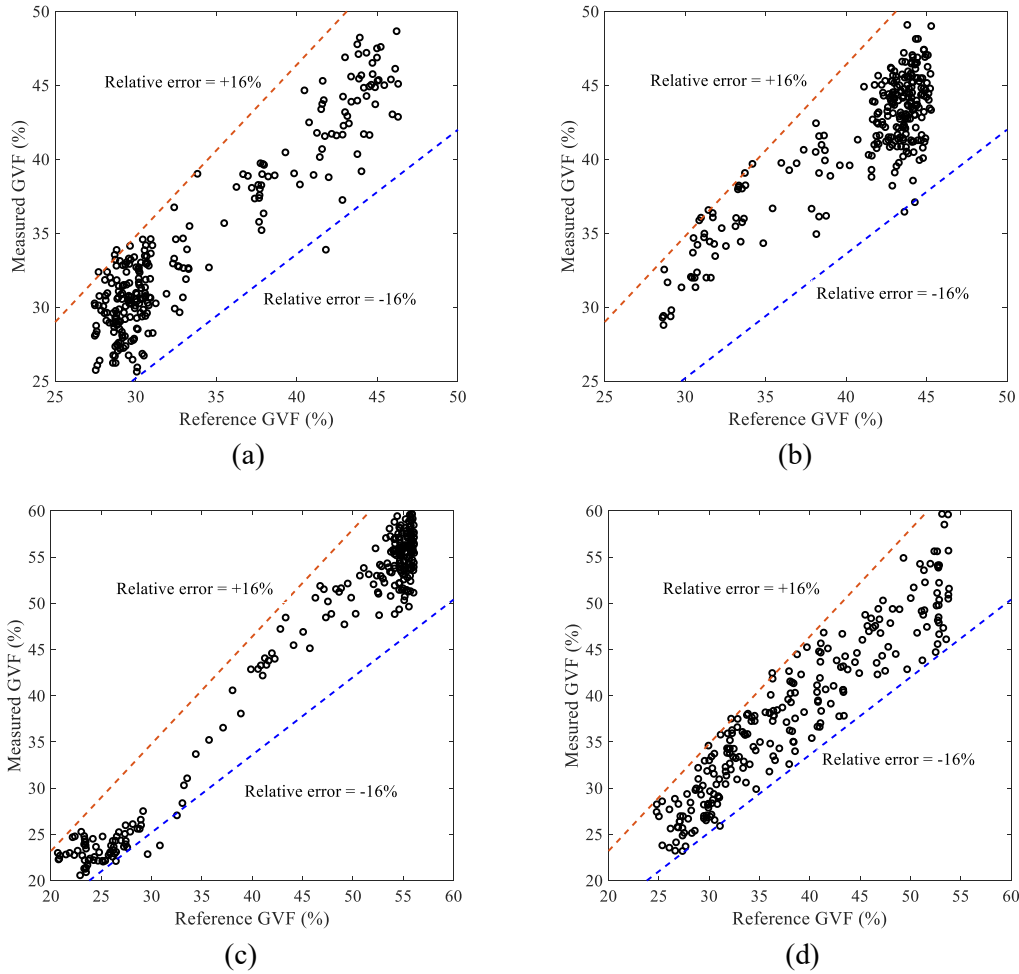


Fig. 12 Comparison between the measured GVF and reference GVF under dynamic conditions. (a) Increasing liquid CO₂ with fixed gas CO₂. (b) Decreasing liquid CO₂ with fixed gas CO₂. (c) Increasing gas CO₂ with fixed liquid CO₂. (d) Decreasing gas CO₂ with fixed liquid CO₂.

5. Conclusions

In this paper analytical and experimental investigations have been carried out to achieve the GVF measurement of gas-liquid two-phase CO₂ flow using a 12-electrode capacitive sensor and data driven models. The results have shown that the RBFNN model produces more accurate GVF measurement than the BPNN and LS-SVM models. During the data pre-processing stage, copula functions and two rank correlation coefficients, i.e. Kendall's and Spearman's coefficients, have been used to select input variables for the data driven models. Under steady-state flow conditions the RBFNN yields a relative error within $\pm 7\%$ in the horizontal pipeline for the GVF ranging from 0% to 84%, whilst the BPNN and LS-SVM models give relative errors within $\pm 13\%$ and $\pm 12\%$, respectively. The results under dynamic flow conditions have verified the real-time performance of the RBFNN with a relative error within $\pm 16\%$. It should be stressed that the GVF measurement of the two-phase CO₂ flow using the capacitance

386 signals and data driven models is achieved without the time-consuming image reconstruction algorithms.

387

388 **Acknowledgements**

389 The authors wish to acknowledge the National Natural Science Foundation of China (No.61973113)
390 and the financial support of the UK CCS Research Centre (www.ukccsrc.ac.uk). The UK CCSRC is
391 funded by the EPSRC as part of the RCUK Energy Programme.

392 **References**

393 Abdilahi, A.M., Mustafa, M.W., Abujarad, S.Y., Mustapha, M., 2018. Harnessing flexibility potential
394 of flexible carbon capture power plants for future low carbon power systems: Review. *Renew. Sust.*
395 *Energ. Rev.* 81, 3101-3110.

396 Azizi S., Awad M. M., Ahmadloo E., 2015. Prediction of water holdup in vertical and inclined oil-water
397 two-phase flow using artificial neural network. *Int. J. Multiph. Flow* 80:181-187.

398 Bowden, R.C., Lessard, étienne, Yang, S.K., 2017. Void fraction measurements of bubbly flow in a
399 horizontal 90 degree bend using wire mesh sensors. *Int. J. Multiph. Flow* 99, 30-47.

400 Chakraborty, S., Keller, E., Talley, J., Srivastav, A., Ray, A., Kim, S., 2009. Void fraction measurement
401 in two-phase flow processes via symbolic dynamic filtering of ultrasonic signals. *Meas. Sci.*
402 *Technol.* 20(2), 023001.

403 Figueiredo, M. M. F., Goncalves, J. L., Nakashima, A. M. V., Fileti, A. M. F., Carvalho, R. D. M., 2016.
404 The use of an ultrasonic technique and neural networks for identification of the flow pattern and
405 measurement of the gas volume fraction in multiphase flows. *Exp. Therm. Fluid Sci.* 70, 29-50.

406 Fredricks, G. A., Nelsen, R. B., 2007. On the relationship between Spearman's rho and Kendall's tau
407 for pairs of continuous random variables. *J. Stat. Plan. Infer.* 137(7), 2143-2150.

408 Han, S., Qiao, Y., Yan, J., Liu, Y., Li, L., Wang, Z., 2019. Mid-to-long term wind and photovoltaic power
409 generation prediction based on copula function and long short term memory network. *Appl. Energy*
410 239, 181-191.

411 Ji, H., Chang, Y., Huang, Z., Wang, B., Li, H., 2014. Voidage measurement of gas-liquid two-phase
412 flow based on capacitively coupled contactless conductivity detection. *Flow Meas. Instrum.* 40,
413 199-205.

414 Jiang, F., Liu, S., Liu, J., Wang, X., 2009. Measurement of Ice Movement in Water Using Electrical

415 Capacitance Tomography. *J. Therm. Sci.* 18(1), 8-12.

416 Karra, K., Mili, L., 2019. Copula index for detecting dependence and monotonicity between stochastic
417 signals. *Inf. Sci.* 485, 18-41.

418 Kemper, J., 2015. Biomass and carbon dioxide capture and storage: A review. *Int. J. of Greenh. Gas*
419 *Control* 40, 401-430.

420 Kim, G., Silvapulle, M.J., Silvapulle, P., 2007. Comparison of semiparametric and parametric methods
421 for estimating copulas. *Comput. Stat. Data Anal.* 51(6): 2836-2850.

422 Kim, J.M.; Lee, N., Xiao, X.Y., 2019. Directional dependence between major cities in China based on
423 copula regression on air pollution measurements. *PLoS One* 14(3), e0213148.

424 Leung, D.Y.C., Caramanna, G., Maroto-Valer, M.M., 2014. An overview of current status of carbon
425 dioxide capture and storage technologies. *Renew. Sust. Energ. Rev.* 39, 426–443.

426 Liao, C., 2010. Genetic k-means algorithm based RBF network for photovoltaic MPP prediction.
427 *Energy* 35(2), 529-536.

428 Mensi, W., Hammoudeh, S., Shahzad, S.J.H., Shahbaz, M., 2016. Modelling systemic risk and
429 dependence structure between oil and stock markets using a variational mode decomposition-based
430 copula method. *J. Bank Financ.* 75, 258-279.

431 Mu, Y., Liu, X., Wang, L., 2018. A Pearson's correlation coefficient based decision tree and its parallel
432 implementation. *Inf. Sci.* 435, 40-58.

433 Navarro, J., 2018. Distribution-free comparisons of residual lifetimes of coherent systems based on
434 copula properties. *Stat. Pap.* 59(2), 781-800.

435 Nazemi, E., Feghhi, S.A.H., Roshani, G.H., Peyvandi, R.G., Setayeshi, S., 2016. Precise void fraction
436 measurement in two-phase flows independent of the flow regime using gamma-ray attenuation.
437 *Nucl. Eng. Technol.* 48(1), 64-71.

438 Nazeri, M., Maroto-Valer, M.M., Jukes, E., 2016. Performance of coriolis flowmeters in CO₂ pipelines
439 with pre-combustion, post-combustion and oxyfuel gas mixtures in carbon capture and storage. *Int.*
440 *J. of Greenh. Gas Control* 54, 297-308.

441 Nelsen, R.B., 2006. *An introduction to Copulas*, second ed. Springer Verlag, New York.

442 Olni, C., Jia, J., Wang, M., 2013. Measurement of air distribution and void fraction of an upwards air–
443 water flow using electrical resistance tomography and a wire-mesh sensor. *Meas. Sci. Technol.*

444 24(3), 035403.

445 Peyvandi, R.G., Rad, S.Z.I., 2017. Application of artificial neural networks for the prediction of volume
446 fraction using spectra of gamma rays backscattered by three-phase flows. *Eur. Phys. J. Plus* 132(12),
447 511.

448 Porter, R.T.J., Fairweather, M., Pourkashanian, M., Woolley, R.M., 2015. The range and level of
449 impurities in CO₂ streams from different carbon capture sources. *Int. J. of Greenh. Gas Control* 36,
450 161-174.

451 Ren, C., An, N., Wang, J., Li, L., Hu, B., & Shang, D., 2014. Optimal parameters selection for BPNN
452 based on particle swarm optimization: a case study of wind speed forecasting. *Knowledge-Based*
453 *Syst.* 56, 226-239.

454 Schweizer, B., Sklar, A., 1983. *Probabilistic metric spaces*. New York, North-Holland.

455 Sklar A., 1959. Fonctions de repartition à n dimensions et leurs marges. *Publ. Inst. Statist. Univ. Paris*
456 8, 229-231.

457 Sun, B., Cao, Y., Feng, Q., Qian, C., Ren, Y., Yang, DZ., 2019. An Archimedean Copula Function-Based
458 Prediction Method for High-Power White LED Considering Multi-Performance. *IEEE Trans.*
459 *Electron. Devices* 66(8), 3405-3410.

460 Sun, S., Cao, Z., Huang, A., Xu, L., Yang, W., 2017. A high-speed digital electrical capacitance
461 tomography system combining digital recursive demodulation and parallel capacitance
462 measurement. *IEEE Sens. j.* 17, 6690-6698.

463 Sun, S.J., Zhang, W.B., Sun, J.T., Cao Z., Xu, L.J. Yan, Y., 2018. Real-time imaging and holdup
464 measurement of carbon dioxide under CCS conditions using electrical capacitance tomography.
465 *IEEE Sens. J.* 18, 7551-7559.

466 Suykens, Johan A. K., 2002. Least squares support vector machines. *Int. J. Circuit Theory Appl.*
467 27(6):605-615.

468 Ursenbacher, T., Wojtan, L., Thome, J.R., 2004. Interfacial measurements in stratified types of flow.
469 part I: new optical measurement technique and dry angle measurements. *Int. J. Multiph. Flow* 30(2),
470 107-124.

471 Wang, H. X., Zhang, L. F., 2009. Identification of two-phase flow regimes based on support vector
472 machine and electrical capacitance tomography. *Meas. Sci. Technol.* 20(11), 114007.

473 Wang, L., Liu, J., Yan, Y., Wang, X., Wang, T., 2017. Gas-liquid two-phase flow measurement using
474 Coriolis flowmeters incorporating artificial neural network, support vector machine, and genetic
475 programming algorithms. *IEEE Trans. Instrum. Meas.* 66(5), 852-868.

476 Wang, L., Yan, Y., Wang, X., Wang, T., Duan, Q., Zhang, W., 2018. Mass flow measurement of gas-
477 liquid two-phase CO₂ in CCS transportation pipelines using Coriolis flowmeters. *Int. J. of Greenh.*
478 *Gas Control* 68, 269-275.

479 Wang, S., Wang, J., Chung, F. L., 2014. Kernel density estimation, kernel methods, and fast learning in
480 large data sets. *IEEE T. Cybern.* 44(1), 1-20.

481 Wang, W., Sefiane, K., Duursma, G., Liang, X., Chen, Y., 2019. Void fraction measurement of gas-
482 liquid two-phase flow based on empirical mode decomposition and artificial neural networks. *Heat*
483 *Transf. Eng.* 40(16), 1403-1416.

484 Wen, C., Karvounis, N., Walther, J.H., Yan, Y.Y., Feng, Y.Q., Yang, Y., 2019. An efficient approach to
485 separate CO₂ using supersonic flows for carbon capture and storage. *Appl. Energ.* 238, 311-319.

486 Xie, D., Huang, Z., Ji, H., Li, H., 2006. An online flow pattern identification system for gas-oil two-
487 phase flow using electrical capacitance tomography. *IEEE Trans. Instrum. Meas.* 55(5): 1833-
488 1838.

489 Zhang, C., Zhang, T., Yuan, C., 2011. Oil holdup prediction of oil–water two phase flow using thermal
490 method based on multiwavelet transform and least squares support vector machine. *Expert Syst.*
491 *Appl.* 38(3), 1602-1610.

492 Zhang, W.B., Shao, D., Yan, Y. Liu, S., Wang, T., 2018. Experimental investigations into the transient
493 behaviours of CO₂ in a horizontal pipeline during flexible CCS operations. *Int. J. of Greenh. Gas*
494 *Control* 79, 193-199.