Pipeline transport of CO_2 mixtures: Models for transient simulation

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Abstract

This paper reviews current research challenges related to the modelling of transient flow of multiphase CO_2-rich mixtures in pipes. This is relevant not only for events like start-up, shutdown or planned or uncontrolled depressurization of pipelines, but also for normal operation, and therefore needs to be taken into account by simulation tools employed for design and operation of CO_2 pipelines. During transportation, CO_2 will often be in a dense liquid phase, whereas e.g. natural gas is in a dense gaseous phase. This requires special attention to depressurization and the possible propagation of cracks. In addition, we highlight and illustrate research challenges related to thermodynamics, and the modelling of the wave-propagation velocity (speed of sound) for two-phase flows. Further, some relevant currently available simulation tools, and their applicability to CO_2 transport, are briefly discussed.

Keywords: CO_2 transport, pipeline, transient simulation, CFD, fluid dynamics, thermodynamics, transport properties, non-equilibrium, depressurization, crack propagation

1. Introduction

CO_2 capture and storage (CCS) is considered one of the most important technologies for reducing the world's emission of greenhouse gases. In the International Energy Agency's two-degree scenario (2DS), CCS will contribute to reducing the global CO_2 emissions by about seven gigatonnes per year in 2050 (IEA, 2012). This is a much larger amount than what is transported in pipelines today for enhanced oil and gas recovery purposes (about 50 megatonnes per year in the USA (US DOE, 2010)), and a major part will be transported in high-pressure pipelines. Therefore, existing knowledge on models and simulation tools for multiphase flow of CO_2 with relevant impurities should be further developed to help improve safety and cost-efficiency.

Multiphase flow modelling has been an active field of research for the last half century (Slattery, 1967; Ishii, 1975; Drew, 1983; Drew and Passman,

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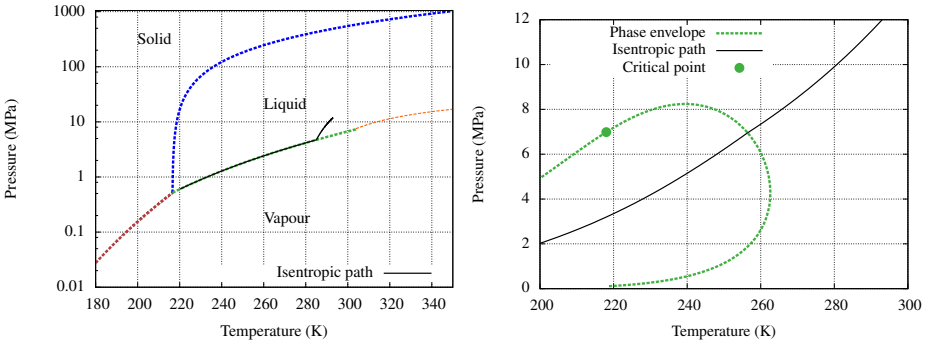
Table 1: Natural gas composition (Aihara and Misawa, 2010).

Component	(mol %)
CH_4	88.9
C_2H_6	6.2
C_3H_8	2.5
iC_4H_{10}	0.4
nC_4H_{10}	0.6
iC_5H_{12}	0.1
nC_5H_{12}	0.1
nC_6H_{14}	0.1
N_2	0.3
CO_2	1.0

1999; Ellul *et al.*, 2004; Ellul, 2010). This development has mainly been driven by the energy sector. In the nuclear industry, two-phase flow is important in reactor cooling systems. Herein, the RELAP model developed by the US Nuclear Regulatory Commission has become the standard tool for simulating transients and accidents in water-cooled reactors (Allison and Hohorst, 2010). In the petroleum industry, there has been a need for pipeline models enabling safe and cost-efficient transport of oil and gas. This research has led to models and tools for dynamic pipeline simulation of three-phase (oil-gas-water) mixtures (Bendiksen *et al.*, 1991; Pauchon *et al.*, 1994; Larsen *et al.*, 1997; Danielson *et al.*, 2011). An example of such a tool is the dynamic multiphase flow simulator OLGA (Bendiksen *et al.*, 1991), which has become industry standard for such applications.

There are a number of specific challenges related to CO_2 transport that makes it, from a modelling point of view, different from the transport of oil and gas. First, the critical point (7.38 MPa at 31.1 °C) and triple point (about 518 kPa at −56.6 °C) are different. This is illustrated in Figure 1, which highlights that CO_2 will normally be transported in a dense liquid state, whereas natural gas is in a dense gaseous state. Second, CO_2 transported in a CCS chain will in general not be pure (de Visser *et al.*, 2008). Depending on the fuel source and capture process, CO_2 might contain nitrogen, oxygen, water, sulphur oxides, methane and other impurities. This will introduce considerable modelling challenges since the presence of even minute quantities of impurities may significantly affect the thermodynamic and transport properties of the mixture (Li *et al.*, 2011a,b). The equation of state by Span and Wagner (1996) (SW EOS) is commonly considered to be the reference for pure CO_2. There are, however, significant gaps in knowledge when it comes to CO_2 with impurities. Furthermore, in pipeline transport of CO_2, it is of interest to predict the minimum water content where hydrates form at a specified pressure, temperature and composition, both for economical and safety reasons (Sloan and Koh, 2008). It is known that even small amounts of impurities can change the equilibrium water content at which hydrates are formed (Song and Kobayashi, 1987, 1990). In case of impurities like water and hydrogen sulphide it is also possible to have multiple liquid phases. When considering tools for simulating multiphase pipe transport, one should distinguish between steady state and transient (time dependent) models. Under normal operation, one scenario for pipeline transport is CO_2



(a) Pure CO_2 (b) Natural gas

Figure 1: Isentropic depressurization from $p = 12$ MPa, $T = 293$ K. CO_2 is in a dense liquid state until it reaches the saturation line and then the triple point, whereas the natural gas is in a dense gaseous state until it reaches the two-phase area. The Span–Wagner EOS has been used for CO_2 and the Peng–Robinson EOS for natural gas. The natural gas composition is given in Table 1.

in a dense or liquid state, since this is the most energy-efficient condition (Zhang *et al.*, 2006; Jung and Nicot, 2010). For this case, pressure-drop predictions for single-phase flow are believed to be satisfactory with the well known correlations for friction factors (see e.g. White, 1994). This is also the case for Nusselt number heat-transfer correlations like the Dittus–Boelter equation (see e.g. Bejan, 1993, Ch. 6). Under such conditions, steady-state analysis to calculate pressure drop, compression work and mass flow might be sufficient for flow assurance. It should be noted, however, that some sources of CO_2, such as coal- or gas-fired powerplants, will be fluctuating, since they are operated in response to external demands. This will cause a transient flow of CO_2 in the pipeline, and moreover, due to the fluctuating mass flow, the pressure will change, and the state in the pipeline may change between single- and two-phase (Klinkby *et al.*, 2011).

There are also other transient events, related to start-up, shutdown and accidents for which the steady-state methodology will be inadequate. One example is pipe depressurization, either accidental or as a part of planned maintenance. The decompression wave associated with such an event will cause the initially dense or liquid CO_2 to undergo phase change. The subsequent cooling might render the pipe material, and any coatings, brittle and vulnerable to cracks. Also, CO_2 has a relatively high triple-point pressure, which means that dry-ice might form during such a depressurization event (Jäger and Span, 2012; Trusler, 2011, 2012). Accurate predictions of the velocity and magnitude of the depressurization and cooling is therefore crucial for assuring safe and reliable operation of a CCS pipeline.

In a transport model, depressurization waves will propagate at the speed of sound of the mixture. In order to accurately resolve transient events, it is therefore essential to model the speed of sound in a physically reasonable way. The multiphase speed of sound is, however, very sensitive to various physical equilibrium assumptions (Flåtten and Lund, 2011). Also, the presence of impurities will affect the propagation velocities of the model (Munkejord *et al.*, 2010). Even in a pure single-phase case, CO_2 mixtures

from different capture technologies will give different dynamic behaviour during pipeline transport. This includes compressor power and hence fuel consumption (Chaczykowski and Osiadacz, 2012).

Widespread implementation of CCS will in some cases require onshore CO_2 transport pipelines running through populated areas. This may require strict safety guidelines due to the pipeline pressure and since CO_2 is toxic at high concentrations. Developing such guidelines will require accurate models for predicting both the occurrence and evolution of pipeline cracks (Nordhagen *et al.*, 2012). Pipelines can then be designed specifically to avoid the significant hazards and financial costs associated with the formation of a running ductile fracture – while reducing the need for safety factors. Existing models for predicting cracks in pipes are semi-empirically-based and were mainly developed for natural gas transport. Such models will need re-calibration when applied to CO_2 with impurities transported in pipes made of modern steel materials.

It should be emphasized that the accuracy of a simulation depends not only on the accuracy of the physical model, but also on the ability of the numerical scheme to correctly resolve the underlying model. It has been shown that numerical diffusion associated with certain numerical methods can adversely affect the resolution of a depressurization wave in a pipeline (Clausen and Munkejord, 2012; Morin *et al.*, 2009). This is, however, outside the scope of this paper.

Race *et al.* (2007) reviewed key technical challenges for anthropogenic CO_2 offshore pipeline transport. Fracture propagation and transient flow were mentioned among the subjects requiring further attention. The purpose of this paper is to review the challenges which should be addressed in the development of models and tools for transient simulation of pipeline flow of CO_2. It should be noted that the subject of this article is composed of several research areas, each with their abundant literature. This is a reflection of the fact that the problem at hand is multifaceted. In particular, in this article, we will focus on leaks and crack propagation as highly relevant examples of transient events for which currently available models may not be sufficient for the application to CO_2 transport.

The outline of this paper is as follows: In Section 2 we discuss the most common approaches for modelling multiphase flow in pipelines. Section 3 is devoted to the modelling of closure relations, thermodynamics and transport properties of CO_2 mixtures, as well as issues associated with the formation of hydrates. In Section 4 we consider the modelling of leaks and crack propagation in pipelines. Different scenarios where such modelling is essential as well as specific challenges related to CO_2 are discussed. In Section 5 we review some common commercially available tools for simulating transient multiphase flow in pipelines, and discuss their applicability to CO_2 transport. Section 6 concludes the paper and highlights topics in which more research is needed.

2. Averaged 1D models for pipeline flow

It is not uncommon to state that two-phase flow should be avoided in CO_2 pipelines (see e.g. Race *et al.*, 2007). However, this requirement may not always be realistic. Klinkby *et al.* (2011) performed a modelling study of the

CO_2 transport chain from a coal-fired power plant, including injection into a reservoir. Due to the transient operation of the power plant, the CO_2 supply will vary. As a result of this, Klinkby *et al.* found that the CO_2 will change phase from dense phase to two-phase gas and liquid in the upper part of the well and in the pipeline. It is also interesting to note that two-phase conditions have been documented in a demonstration well at the Ketzin site in Germany (Henninges *et al.*, 2011). There are also indications of two-phase flow at the wellhead at the Sleipner field in the North Sea (Munkejord *et al.*, 2012). In addition to this, phase change will occur during situations like first fill and depressurization. This motivates the study of transient multi-phase flow of CO_2-rich mixtures.

In this section we discuss some of the most common formulations of the governing dynamics of multi-phase pipeline flow. Note that most of these topics will be generic with regard to the transported medium and impurities. Issues specific to CO_2 transport will be most apparent when introducing equations of state and closure relations for the averaged model, which will be the topic of the subsequent sections.

2.1. The two-fluid model

For a real-scale pipeline, fully resolving the governing equations of the multiphase flow is computationally intractable. The usual way to get around this problem is to consider averaged models (see e.g. Drew and Passman, 1999). For a pipeline, a commonly used approach is to consider transport equations for mass, momentum and energy averaged across the cross section of the pipe. For two-phase flow, the resulting 1D model takes a form often referred to as the two-fluid model. A common formulation is given by

Conservation of mass:

$$\frac{\partial}{\partial t}(\rho_{\mathrm{g}}\alpha_{\mathrm{g}}) + \frac{\partial}{\partial x}(\rho_{\mathrm{g}}\alpha_{\mathrm{g}}u_{\mathrm{g}}) = \Gamma, \tag{1}$$

$$\frac{\partial}{\partial t}(\rho_{\ell}\alpha_{\ell}) + \frac{\partial}{\partial x}(\rho_{\ell}\alpha_{\ell}u_{\ell}) = -\Gamma. \tag{2}$$

Conservation of momentum:

$$\frac{\partial}{\partial t}(\rho_{\mathrm{g}}\alpha_{\mathrm{g}}u_{\mathrm{g}}) + \frac{\partial}{\partial x}(\rho_{\mathrm{g}}\alpha_{\mathrm{g}}u_{\mathrm{g}}^2 + \alpha_{\mathrm{g}}p_{\mathrm{g}}) - p^{\mathrm{i}}\frac{\partial \alpha_{\mathrm{g}}}{\partial x} = \rho_{\mathrm{g}}\alpha_{\mathrm{g}}f_x - M_{\mathrm{w,g}} - M^{\mathrm{i}} + u_{\Gamma}^{\mathrm{i}}\Gamma, \tag{3}$$

$$\frac{\partial}{\partial t}(\rho_{\ell}\alpha_{\ell}u_{\ell}) + \frac{\partial}{\partial x}(\rho_{\ell}\alpha_{\ell}u_{\ell}^2 + \alpha_{\ell}p_{\ell}) - p^{\mathrm{i}}\frac{\partial \alpha_{\ell}}{\partial x} = \rho_{\ell}\alpha_{\ell}f_x - M_{\mathrm{w},\ell} + M^{\mathrm{i}} - u_{\Gamma}^{\mathrm{i}}\Gamma. \tag{4}$$

Conservation of energy:

$$\frac{\partial}{\partial t}(\rho_{\mathrm{g}}\alpha_{\mathrm{g}}E_{\mathrm{g}}) + \frac{\partial}{\partial x}\left(\rho_{\mathrm{g}}\alpha_{\mathrm{g}}u_{\mathrm{g}}\left(E_{\mathrm{g}} + \frac{p_{\mathrm{g}}}{\rho_{\mathrm{g}}}\right)\right) + p^{\mathrm{i}}u_{\tau}^{\mathrm{i}}\frac{\partial \alpha_{\mathrm{g}}}{\partial x} = \rho_{\mathrm{g}}\alpha_{\mathrm{g}}u_{\mathrm{g}}f_x + Q_{\mathrm{w,g}} - Q^{\mathrm{i}} - u_M^{\mathrm{i}}M^{\mathrm{i}} + E^{\mathrm{i}}\Gamma, \tag{5}$$

$$\frac{\partial}{\partial t}(\rho_\ell \alpha_\ell E_\ell) + \frac{\partial}{\partial x}\left(\rho_\ell \alpha_\ell u_\ell \left(E_\ell + \frac{p_\ell}{\rho_\ell}\right)\right) + p^{\mathrm{i}} u_\tau^{\mathrm{i}} \frac{\partial \alpha_\ell}{\partial x} = \rho_\ell \alpha_\ell u_\ell f_x + Q_{\mathrm{w},\ell} + Q^{\mathrm{i}} + u_M^{\mathrm{i}} M^{\mathrm{i}} - E^{\mathrm{i}} \Gamma, \quad (6)$$

where the nomenclature is as follows:

α_k	Volume fraction of phase k
ρ_k	Mass density of phase k
u_k	Velocity of phase k
p_k	Pressure of phase k
E_k	Energy density for fluid k, $E_k = e_k + 1/2\, u_k^2$
Q_k	Heat source for phase k
f_x	x-component of body force

In the cross-section averaged description above, the model does not contain information about the internal moving interfaces between the phases. Also, any information on local gradients along the cross section of the pipe is lost in the averaging procedure. Closure relations are thus needed to model the source terms representing transfer of heat, Q, mass, Γ, and momentum, M, between the fields (denoted by the index i) and between the fields and the pipe wall (denoted by the subscript w). In general, these closure relations will depend on the detailed description of the flow, and they cannot be derived from first principles based on averaged quantities (Stewart and Wendroff, 1984). The modelling of such terms is further discussed in Section 3.

2.2. The drift-flux model

In multiphase pipe flow, there are flow regimes where the velocities of the individual phases are highly correlated. For two-phase flow, the relative velocity between the phases can be expressed as a slip relation

$$u_1 - u_2 = \Phi(\alpha_1, p, T, u_1), \quad (7)$$

see the work of e.g. Zuber and Findlay (1965), Ishii (1977) and Hibiki and Ishii (2002).

A slip relation in the form (7) can be used to reduce the complexity of the two-fluid model (1)–(6). In particular, if the pressures in both phases are assumed to be equal, $p_1 = p_2 = p$, the momentum equations (3)–(4) can be combined into a single mixture momentum equation. Likewise, with the assumption of equal phasic temperatures, $T_1 = T_2 = T$, the energy equations (5)–(6) can also be combined. The resulting *drift-flux* model is given by

Conservation of mass:

$$\frac{\partial}{\partial t}(\rho_{\mathrm{g}} \alpha_{\mathrm{g}}) + \frac{\partial}{\partial x}(\rho_{\mathrm{g}} \alpha_{\mathrm{g}} u_{\mathrm{g}}) = \Gamma, \quad (8)$$

$$\frac{\partial}{\partial t}(\rho_\ell \alpha_\ell) + \frac{\partial}{\partial x}(\rho_\ell \alpha_\ell u_\ell) = -\Gamma. \quad (9)$$

Conservation of momentum:

$$\frac{\partial}{\partial t}((\rho_{\mathrm{g}} \alpha_{\mathrm{g}} u_{\mathrm{g}} + \rho_\ell \alpha_\ell u_\ell) + \frac{\partial}{\partial x}(\rho_{\mathrm{g}} \alpha_{\mathrm{g}} u_{\mathrm{g}}^2 + \rho_\ell \alpha_\ell u_\ell^2 + p) = (\rho_{\mathrm{g}} \alpha_{\mathrm{g}} + \rho_\ell \alpha_\ell) f_x - M_{\mathrm{w}}. \quad (10)$$

Conservation of energy:

$$\frac{\partial}{\partial t}\left(\rho_{\mathrm{g}}\alpha_{\mathrm{g}}E_{\mathrm{g}}+\rho_{\ell}\alpha_{\ell}E_{\ell}\right) + \frac{\partial}{\partial x}\Big(\left(\rho_{\mathrm{g}}\alpha_{\mathrm{g}}u_{\mathrm{g}}(E_{\mathrm{g}}+p/\rho_{\mathrm{g}})\right)+\left(\rho_{\ell}\alpha_{\ell}u_{\ell}(E_{\ell}+p/\rho_{\ell})\right)\Big) = \left(\rho_{\mathrm{g}}\alpha_{\mathrm{g}}u_{\mathrm{g}}+\rho_{\ell}\alpha_{\ell}u_{\ell}\right)f_x+Q_{\mathrm{w}}. \quad (11)$$

Besides being simpler and in conservation form, the drift-flux model also, as discussed by Munkejord (2005), has some advantages over the two-fluid model when it comes to stability and well-posedness. However, it may not be appropriate to model all relevant flow regimes with a slip relation of the form (7). The drift-flux model (8)–(11) with the additional assumptions of no slip ($u_{\mathrm{g}} = u_{\ell}$) and equal chemical potential in the two phases is often referred to as the *homogeneous equilibrium* model.

For two-phase mixtures, the composition of the gas and the liquid will in general differ. Hence, if there is slip between the phases, the flow model needs to include a mass-conservation equation for each component.

2.3. Wave speeds in multifluid models

When studying transient events in CO_2 pipelines, the speed with which disturbances propagate along the pipe is an important factor. In any fluid, pressure waves travel at the speed of sound relative to the local velocity. It is therefore essential to include a realistic speed of sound to be able to correctly simulate many transient events in pipes.

For the basic two-fluid model (1)–(6), the eigenvalues of the flux Jacobian are not guaranteed to be real (Gidaspow, 1974). When this occurs, the equation system is no longer hyperbolic, which causes problems related to stability and well-posedness (Stuhmiller, 1977). To remedy this, regularization terms are often introduced, forcing the eigenvalues to be real. In the opposite case, robustness issues are typically encountered, unless the solver has a high-enough numerical smearing.

2.3.1. Non-equilibrium fluid-dynamical models

In general, the wave speeds of a set of conservation laws are also influenced by various source terms. Local source terms will not influence the characteristics of the system but will introduce dispersion, i.e. wave-number dependent sound velocities (Aursand and Flåtten, 2012).

Relaxation terms represent a class of local source terms that are of particular relevance to multiphase flow modelling (Baer and Nunziato, 1986; Saurel *et al.*, 2008; Flåtten and Lund, 2011). Chemical, thermal and mechanical non-equilibrium are examples of processes that can be described with relaxation terms. A hyperbolic relaxation model can be written in the form

$$\frac{\partial}{\partial t}\boldsymbol{Q}+\frac{\partial}{\partial x}\boldsymbol{F}(\boldsymbol{Q}) = \frac{1}{\varepsilon}\boldsymbol{R}(\boldsymbol{Q}), \quad (12)$$

where $\boldsymbol{R}(\boldsymbol{Q})$ is a relaxation term representing the driving-force pulling the system towards local equilibrium, characterized by $\boldsymbol{R}(\boldsymbol{Q}) = 0$. The relaxation time ε can be seen as a characteristic time scale of the relaxation process.

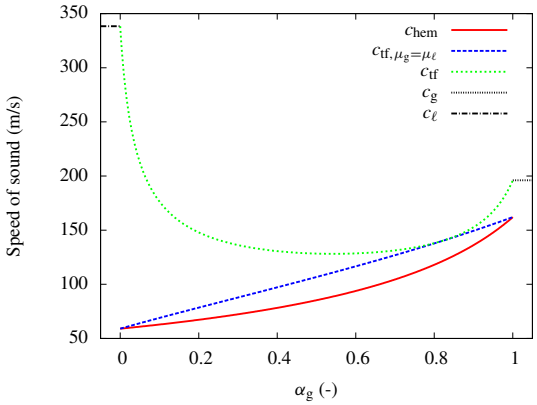


Figure 2: Speed of sound of pure CO_2 in the gas-liquid two-phase area as a function of gas volume fraction for various two-phase flow models. $T = 250\,\mathrm{K}$, SW EOS. 'hem' denotes the homogeneous equilibrium model, 'tf' denotes the two-fluid model with no phase change and no slip, 'tf $\mu_g = \mu_\ell$' is the two-fluid model with full chemical equilibrium and no slip, 'g' is gas and 'ℓ' is liquid.

For a given relaxation process there is a corresponding local equilibrium approximation. The characteristic velocities of the equilibrium model are in general different from those of the relaxation model. Flåtten and Lund (2011) analysed two-phase drift-flux models with and without thermal, mechanical and chemical equilibrium. They showed that imposing equilibrium will always reduce the speed of sound for such models, i.e., the characteristic velocities of the local equilibrium model are smaller than those of the non-equilibrium (relaxation) model. In general, this concept is known as the sub-characteristic condition and is closely related to the stability and well-posedness of the model (Chen *et al.*, 1994).

In the modelling of multiphase flow, the assumption of thermal, mechanical or chemical equilibrium is ubiquitous. While these assumptions often simplify the model in question, it is important to be aware that they will directly influence the wave dynamics of the model. For example, assuming chemical, thermal and mechanical equilibrium may lead to a significant underestimation of the rate of which disturbances will propagate in a pipeline, compared to a non-equilibrium model. This is illustrated in Figure 2, where the speed of sound of pure CO_2 is calculated for different two-phase flow models as a function of gas volume fraction. The graphs are plotted for a temperature of $T = 250\,\mathrm{K}$ using the Span–Wagner equation of state (SW EOS). It can be seen that models with the assumption of full chemical equilibrium (instantaneous phase transfer) have the artifact of a discontinuous speed of sound in the limit of single-phase flow. This is not believed to be physical. Further, it can be seen from the figure that allowing phase transfer lowers the predicted speed of sound in almost the entire volume-fraction range. This highlights how tightly intertwined thermo and fluid dynamics are for two-phase flow.

In Figure 2, we have plotted analytical expressions for the speed of sound. The speed of sound in the homogeneous equilibrium model is also referred to as the 'Wood speed of sound', and it can e.g. be found in Martínez Ferrer

et al. (2012, eq. (3.7)). The speed of sound in the two-fluid model with no phase change and no slip can be found in Martínez Ferrer *et al.* (2012, eq. (3.74)). Finally, the speed of sound in the two-fluid model with full chemical equilibrium and no slip can be found in Morin and Flåtten (2012), see also Morin (2012).

Since decompressions of CO_2 will often pass through the triple point, it is interesting to note that at the triple point, for full equilibrium, the speed of sound is zero (Henderson, 2000, Sec. 2.8.1).

3. Closure relations and thermophysical models

For an averaged multifluid model such as (1)–(6), closure relations are needed for terms depending on transversal gradients and the detailed phase configuration. Since these relations cannot be derived from the same first principles as the averaged flow model, they need to be modelled. Moreover, thermodynamic relations are needed for calculating the pressures, temperatures and compositions, as a function of the variables of the fluid-dynamic transport model.

3.1. Closure relations for CO_2

While the field of multiphase flow modelling is mature, there exists no general way of modelling closures valid for all fluids. Flow maps and correlations must be validated, adjusted or developed for each new working fluid or composition of fluids. This presents one of the main challenges in the modelling of CO_2 flow in pipelines. Existing correlations and models used by research and industry for oil-gas-water mixtures cannot necessarily be assumed to be valid for CO_2 with impurities. These models need to be adapted to these new applications, a process needing experimental input for validation.

For CO_2, there exist flow maps and pressure-drop measurements for tubes and channels with a hydraulic diameter in the millimetre range. Most of them are developed for heat exchanger applications, see e.g. (Bredesen *et al.*, 1997; Pettersen, 2002; Yun and Kim, 2003; Cheng *et al.*, 2008).

Aakenes (2012) compared experimental data for frictional pressure-drop for steady-state two-phase flow of pure CO_2 (see also de Koeijer *et al.*, 2011) to data calculated using the model of Friedel (1979) and that of Cheng *et al.* (2008). Although the latter was developed specifically for CO_2, the former fitted the data better, most likely to its broader experimental base.

Since the existing small-scale data may not be representative for real pipelines, there is a need for medium and large-scale data. Presently, there exist some initiatives towards this end, such as the OXYCFB300 Compostilla Project (CIUDEN, 2012) and the multiphase CO_2 lab at the Institute for Energy Technology (IFE) (SPT Group, 2012).

3.2. Thermophysical models for pure CO_2

For pure CO_2, a large amount of experiments have been conducted for thermodynamic properties such as densities, heat capacities and liquid-vapour coexisting curves, as well as for transport properties. The accurate single-component equation of state (EOS) by Span and Wagner (1996) is

considered the reference EOS for pure CO_2. The EOS is valid for temperatures from 216 to 1100 K and pressures up to 800 MPa, which is more than sufficient for pipeline transport of CO_2. Accurate models for the viscosity and the thermal conductivity were developed by Vesovic *et al.* (1990). Fenghour and Wakeman (1998) presented an improved viscosity model. The resulting overall viscosity model for pure CO_2 covers the temperature range of 200 K–1500 K and pressures up to 300 MPa.

3.3. Thermophysical models for CO_2 mixtures

For CO_2 mixtures relevant for CCS, the amount of available data is more scarce than for single-component CO_2. This is true both for the thermodynamic properties (Li *et al.*, 2011a; Hu *et al.*, 2007) and for the transport properties (Li *et al.*, 2011b). Consequently the development of comprehensive reference models has not yet been possible.

Li *et al.* (2011a) argue in their review that there is no equation of state which shows any clear advantages in CCS applications. The cubic equations of state have a simple structure and are capable of giving reasonable results for the thermodynamic properties, but are inaccurate in the dense phase and around the critical point (Wilhelmsen *et al.*, 2012). More complex equations of state such as Lee-Kesler (Lee and Kesler, 1975), SAFT (Wertheim, 1984a,b, 1986a,b) and GERG (Kunz *et al.*, 2007) typically give better results for the density, but not necessarily for the vapour-liquid equilibrium. See also Dauber and Span (2012). Wilhelmsen *et al.* (2012) have recently shown evaluations with the SPUNG EOS (Jørstad, 1993). They found that the SPUNG equation represents a good compromise between accuracy, versatility and computational time-use for calculations with CO_2 mixtures.

It is well known that the EOS must be equipped with suitable interaction parameters to give reliable phase-equilibrium predictions (Wilhelmsen *et al.*, 2012). These are available for cubic EOS'es and several CO_2 mixtures (Li and Yan, 2009), but for other EOS'es, regression of new interaction parameters is needed (Wilhelmsen *et al.*, 2012).

For the viscosities and thermal conductivities of CO_2 mixtures, the gas phase is well investigated for many impurities. Accurate models are available in the literature, for instance through Chapman-Enskog theory or corresponding-state relations (Reid *et al.*, 1987). For the liquid phase, however, no experimental data are available except for mixtures of $CO_2/H_2O/NaCl$, which makes development and validation of models difficult (Li *et al.*, 2011b). One should therefore expect large uncertainties in empirical closure relations which rely heavily on the prediction of viscosities or thermal conductivities in liquid phase CO_2 mixtures.

Currently, some experimental work is being carried out towards obtaining properties for CO_2 mixtures (Sanchez-Vicente *et al.*, 2013; Stang *et al.*, 2012). It should also be noted that pseudo-experimental data of vapour-liquid equilibrium and transport properties for CO_2 mixtures can be calculated using molecular simulations based on Monte Carlo and Molecular Dynamics. CO_2 + N_2O and CO_2 + NO are investigated by Lachet *et al.* (2012).

Water is a common impurity in the CO_2 stream, which is the key component in several undesired phenomena, such as hydrate formation, ice formation and corrosion. The CO_2 will have a significant solubility in the

water phase, which changes its properties. In addition, water and CO_2 can form mixtures with more than two phases, which necessitates more than two phases in the fluid-dynamical model formulation. Extensive reviews have been presented in the literature on the mutual solubility of water, CO_2 and other impurities (Chapoy *et al.*, 2004; Austegard *et al.*, 2006; Hu *et al.*, 2007).

3.4. Implementation in fluid-dynamic pipeline models

Equations of state are usually not written in a form suitable for fluid-dynamic simulations. For example, a pressure-temperature state function cannot be directly employed in model formulations of the form presented in Section 2. Rather, a density-energy function is more appropriate. This necessitates the development of fast and robust numerical algorithms for solution phase-equilibrium equations with specification of energy and density (Michelsen and Mollerup, 2007). Giljarhus *et al.* (2012) studied such a method for the Span–Wagner EOS for pure CO_2. With CO_2 containing impurities, robust and time efficient solution of the phase equilibrium is a considerable challenge (Wilhelmsen *et al.*, 2013).

3.5. Hydrate formation, solid CO_2 and non-equilibrium effects

For economic and safety reasons, it is of interest to predict the minimum water content where hydrates form at a specified pressure, temperature and composition (Sloan and Koh, 2008). The equilibrium of hydrate formation is a well investigated issue for natural gas mixtures, but few data are available for pure CO_2 (Tohidi *et al.*, 2010), and even fewer for CO_2 mixtures. Song and Kobayashi (1987, 1990) show that even small amounts of impurities can change the equilibrium water content at which hydrates are formed. Reliable prediction of the hydrate equilibrium depends on equations of state which are able to provide accurate estimates of the chemical potential in CO_2 mixtures with small water concentrations. This is not trivial, and often requires tailored EOS'es and interaction parameters, such as the CPA equation, or SRK with Huron–Vidal mixing rules (Austegard *et al.*, 2006). See also Chapoy *et al.* (2004). Jäger *et al.* (2013) employed accurate equations of state to predict hydrate formation in pure CO_2 with water.

Several commercial codes predict hydrate equilibrium properties also for CO_2 with impurities. However, without an EOS tailormade to provide an accurate estimate of the chemical potentials of water in CO_2 mixtures, the results may not be reliable. During depressurization events or formation of cracks in pipelines, there is a risk of formation of solid CO_2. Zhang (2012) shows models which are capable of providing accurate predictions of the CO_2 freeze-out temperature of several CO_2-CH_4 mixtures, and experimental data are also available for systems with N_2 (Argwal and Laverman, 1974). A comprehensive evaluation solid-phase equilibria for CO_2 mixtures with impurities is currently not available. Uncertainties in the models should be expected for CO_2-rich mixtures with other impurities than CH_4 and N_2 (Zhang, 2012).

In fluid-dynamical simulations, it is common to assume mechanical, thermal and chemical equilibrium between the coexisting phases. Flåtten and Lund (2011) argue that this is insufficient in many applications. In

dynamic simulations of depressurization of pipelines for instance, the transients in the systems will be so fast that the coexisting phases are not in equilibrium. The metastable sections of an equations of state where subcooling or overheating occurs, are well defined mathematically and may be used to a certain extent to account for situations away from equilibrium. However, the rate at which transfer of heat, mass and momentum between the phases occurs is not easily described by thermodynamics alone, since it is about how the transport across the interface separating the gas and liquid evolves over time. Theories for this are currently being developed (Kjelstrup and Bedeaux, 2008), but these theories have yet to be used in existing fluid-dynamical simulations of CO_2 transport.

The presence of free water is the principal influence on corrosion rate in pipes (see e.g. Cole *et al.*, 2011). However, since the present subject is transient effects, this will not be further discussed here.

4. Flow through valves and cracks

Simulating transient events related to depressurization or crack formation in CO_2 pipelines requires modelling of multiphase critical flow through an orifice. For homogeneous flows, critical flow occurs at the sonic point. By assuming isentropic flow, we can integrate the differential relations

$$\mathrm{d}\left(\rho u A\right) = 0 \tag{13}$$

$$\mathrm{d}\left(h + \frac{1}{2}u^2\right) = 0 \tag{14}$$

$$\mathrm{d}s = 0, \tag{15}$$

along a streamline going through the valve or crack. In the above, A is the cross-section area, h is the specific enthalpy and s is the specific entropy.

For multiphase flow, phase transfer needs to be taken into account when integrating (13)–(15). Herein, there are two different assumption in common use, each representing an extreme case:

Homogeneous equilibrium model The choke flow is assumed to remain in equilibrium. Equations (13)–(15) are integrated along a path of chemical equilibrium.

Frozen model The phase composition is assumed to remain constant through the choke. Equations (13)–(15) are integrated along a path where the mass fractions are constant and where the chemical potentials of the phases are not equal.

In addition to the two extreme cases described above, there exists a number of empirical correlations in common use (Auria and Vigni, 1980). One of the most cited is the Henry–Fauske model (Henry and Fauske, 1971), which can be seen as a correction to the frozen approximation.

In general, different assumptions of phase equilibrium will lead to different choke pressures, and consequently different mass-flow rates. A typical situation is illustrated in Figure 3. A homogeneous equilibrium model will give choked flow at a lower pressure difference than a non-equilibrium model. For many cases the resulting difference in predicted mass flow will

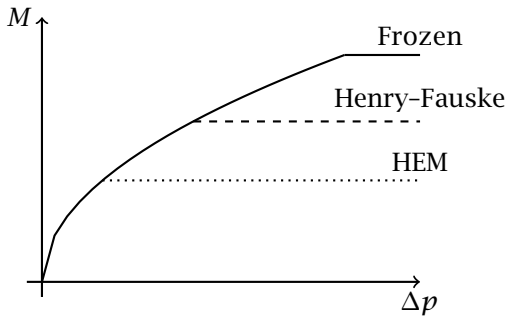


Figure 3: Illustration of the two-phase mass-flow rate M through an orifice as a function of the pressure difference Δp, for different equilibrium assumptions.

be significant. The assumption of phase equilibrium in valves and cracks can therefore strongly influence transient multiphase pipeline simulations.

For multiphase flow, the assumption of homogeneous flow though a valve or crack might not be valid. Depending on the flow regime, the acceleration of the denser phases might be significantly lower than that of the less dense phases.

4.1. Running ductile fractures in CO_2 pipelines

For CO_2 transport, pipeline crack modelling represents a particularly relevant example of an application of critical flow. CO_2 is toxic at high concentrations; predicting the occurrence and evolution of cracks is therefore essential for designing and operating a safe CCS pipeline. For high-pressure pipelines, including CO_2 lines (Maxey, 1986), a concern is also the formation of running ductile fractures. In order to prevent hazardous situations and potentially significant costs, high-pressure pipelines must be designed both to avoid the formation of cracks and to ensure the quick arrest of any cracks that might still form.

Running ductile fracture is commonly assessed using semi-empirical methods like the Battelle method (Maxey, 1974). Herein, the fluid decompression and the fracture propagation in the pipeline are assumed to be uncoupled processes. The fracture velocity is correlated to the fracture energy (e.g. Charpy energy). As long as the fracture velocity is smaller than the decompression wave velocity, crack arrest is assured. In the HLP approach (Sugie *et al.*, 1982), the final crack length is also predicted. There exists a large body of work in the field, see e.g. Ives *et al.* (1974); Parks and Freund (1978); Picard and Bishnoi (1988); Leis and Eiber (1998); Makino *et al.* (2001); Hashemi (2009). Recalibration is needed for new fluids and new material qualities. In particular, for modern steel types with high toughness, the relationship between fracture velocity and Charpy energy is less certain (Leis *et al.*, 2005). Thus it is challenging to predict the pressure at which a running fracture will arrest.

Although the saturation pressure and arrest pressure are key parameters (Cosham and Eiber, 2008), the evolution of a pipeline crack is a coupled material-fluid problem (Mahgerefteh and Atti, 2006). The fracture speed depends on the forces caused by the pressure difference through the crack,

while the pressure in the pipe depends on the rate of escaping mass flow which again depends on the crack size. The arrest or continued propagation of a crack will depend on the difference between the speed of the depressurization wave in the fluid and the speed of the crack tip. If the depressurization propagates faster than the crack, the driving forces maintaining the crack propagation will vanish and the crack will arrest; if not, the crack might form a running fracture. The crack arrest length will therefore also depend on the fluid inside the pipe (Aihara and Misawa, 2010; Mahgerefteh *et al.*, 2012a). This is important because the existing semi-empirical models for evaluating running fractures in pipes were mainly developed for natural gas transport. Such models will need costly recalibration before they can be applied to CO_2 transported in pipes made of modern steel materials (Nordhagen *et al.*, 2012).

Running ductile fracture in gas-transport pipelines consists of three main phenomena, namely, the large-scale elasto-plastic deformation of pipe walls, the three-dimensional nonsteady fluid dynamics and the inelastic dynamic crack-extension process (O'Donoghue *et al.*, 1991). Due to the complexity of these factors, and their interaction, there exist relatively few fully coupled models for the prediction running ductile fracture.

O'Donoghue *et al.* (1991, 1997) developed a fluid-structure interaction model in which a three-dimensional finite-difference fluid-dynamics code was linked with a shell finite-element code. O'Donoghue *et al.* (1997) considered crack arrestors, which are steel rings employed to prevent long running axial cracks. The effect of dissipation of plastic work for high-toughness steels was studied by You *et al.* (2003). Greenshields *et al.* (2000) investigated fast brittle fracture in plastic pipes, employing a finite-volume discretization both for the pipe and the fluid. Herein, the pipe material was represented in 3D, while the fluid flow was calculated in 1D.

Several authors have considered the behaviour of a gas escaping through a crack or nozzle, but few have coupled the structural failure with the fluid behaviour. In the work by Rabczuk *et al.* (2010), a meshfree method for treating fluid-structure interaction of fracturing structures under impulsive loads was described. Terenzi (2005) emphasized that it is necessary to take care of real fluid behaviour when analyzing the decompression properties of dense natural gas mixtures. It was found that friction hinders crack propagation, while condensation promotes it. Mahgerefteh *et al.* (2006) simulated outflow after rupture in pipeline networks. It was found that bends, branches and couplings could have significant effects on the fluid flow. Cumber (2007) described a methodology for predicting outflow from a rupture in a pipeline transporting supercritical ethylene. The flow was modelled without solving a full two-phase flow model, but phase change was accounted for.

Berstad *et al.* (2011); Nordhagen *et al.* (2012) used a coupled material-fluid methodology in order to predict crack arrest for natural gas and hydrogen. Good agreement with full-scale tests (Aihara *et al.*, 2008) was obtained. A similar modelling approach was used by Misawa *et al.* (2010). In an experimental and computational study, Yang *et al.* (2008) found that as the amount of heavier hydrocarbons increased in the natural gas, steels of higher toughness were required. Mahgerefteh *et al.* (2012a) evaluated the effect of some stream impurities on ductile fractures in CO_2 pipelines,

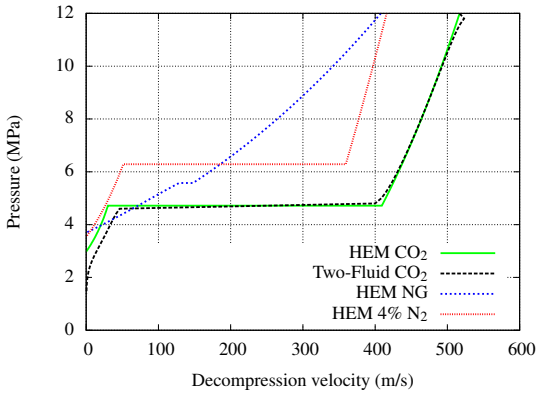


Figure 4: Fluid pressure versus decompression velocity for the homogeneous equilibrium model (HEM) and the two-fluid model with full chemical equilibrium. NG denotes the natural gas from Table 1.

while Aursand *et al.* (2012) took into account dry-ice formation in pure CO_2. Both of the two latter studies found that CO_2 pipelines might be more susceptible to running ductile fracture than natural gas pipelines. Regarding the validation of these predictions, to our knowledge, no experimental data for running fractures in CO_2 pipelines have been published, but work is under way, see e.g. Lucci *et al.* (2011). It can therefore be said that the development of coupled fluid-structure models for crack behaviour in CO_2 pipelines is at an early stage.

To illustrate the effect of fluid flow modelling and fluid properties, we have plotted pressure versus decompression velocity in Figure 4. The decompression velocity is the speed of sound minus the flow velocity ($c - u$) as the decompression wave travels through a 'long' pipe. In the figure, we have plotted the decompression velocity using the homogeneous equilibrium model for pure CO_2 (using the SW EOS), for CO_2 with 4 % N_2 (using the EOS by Peng and Robinson (1976) (PR)) and for a natural gas (using the PR EOS with the composition given in Table 1). The plots have been made for an initial state of $p = 12$ MPa and $T = 293$ K. In e.g. the Battelle method, similar plots are generated, and a curve for the arrest pressure of the pipe is added. In the left region, the CO_2 curves lie above the one for the natural gas. This indicates that CO_2 gives a lower decompression speed in this region, which means that the pipe filled with CO_2 may be more vulnerable to running ductile fracture, see e.g. Cosham and Eiber (2008); Aihara and Misawa (2010). It is clear from the figure that the addition of N_2 to the CO_2 stream aggravates the situation.

Figure 4 also shows a curve calculated using the two-fluid model with full chemical equilibrium. In contrast to the case in Figure 2, here, there is slip between the phases. Hence the decompression speed has been calculated numerically. For cases like the emptying of a pipe, it is quite clear that the assumption of slip or no slip has a large influence. On the other hand, the present plot indicates that for the fast process of crack propagation, the slip modelling may be of less importance. However, it is interesting to note that in this case, the homogeneous equilibrium model would prescribe a

more conservative design than would the two-fluid model.

4.2. Depressurization through valves

For planned maintenance, or in case of emergency shutdown, a CO_2 pipeline might need to be quickly depressurized through one or more valves. If this depressurization is performed too fast, the pipeline might be cooled to the point where the material becomes brittle and cracks might occur. Moreover, if the CO_2 reaches it triple point (518 kPa and −56.6 °C) dry ice will be formed, potentially causing blockages.

The development of reliable simulation tools requires validation of models using experimental data. There is, however, a limited amount of publicly available experimental data for the depressurization of CO_2 pipelines. As a consequence, there is also a limited amount of work along the lines of validating standard models for such applications. Clausen *et al.* (2012) considered the depressurization of a 50 km onshore CO_2 pipeline and compared it to a simulation performed using OLGA®. The results showed reasonable agreement for the pressure, while there were significant discrepancies in the predicted cooling of the pipe. A similar conclusion was reached by de Koeijer *et al.* (2011).

Mahgerefteh *et al.* (2012b) simulated depressurizations of a pipe employing the homogeneous equilibrium model and comparing with experimental data. It was found that for depressurizations from the gaseous phase, the addition of impurities lowered the phase transition pressure plateau, as opposed to depressurizations from the dense phase, where the effect was the opposite.

5. Available simulation tools

The industrial relevance of oil and gas transport has lead to the development of commercial tools for the simulation of pipeline transport. From the point of view of CCS, it is of interest to establish if some of these tools might be applicable and sufficiently accurate for simulating the transport of CO_2 with impurities.

Detailed information on commercial simulation tools is usually not public information. However, the underlying transport model if often published and can be put in context with the technical topics of this paper. In the following, we consider some of the most common commercial tools and briefly discuss their potential for simulating pipeline transport of CO_2.

5.1. OLGA

The development of the dynamic two-fluid model OLGA® was started in the early 80s by Statoil in order to meet the two-phase modelling challenges specific to pipelines (Bendiksen *et al.*, 1991). The tool has since then been under continuous development supported by the oil industry, and is today considered an industry standard for such applications.

Today, the standard OLGA tool solves for a three-phase mixture of gas, oil and water (Håvelsrud, 2012b). The model contains nine conservation equations: Five equations describe conservation of mass in the bulk of the phases as well as oil droplets immersed in gas and gas bubbles immersed in

oil. There are three momentum equations and one mixture energy equation. Standard OLGA can handle impurities through externally supplied thermodynamic data tables. In this case, the phase envelope must be sufficiently wide.

A recent addition to OLGA which makes it more suitable for CO_2 transport is the single-component two-phase module (Håvelsrud, 2012a). This model contains six conservation equations: Three equations describe conservation of mass. There are two momentum equations and one mixture energy equation. For pure CO_2, the Span–Wagner equation of state is used. At present, single-component OLGA cannot take the presence of impurities in CO_2 into account. Future versions might, however, have this capability. The formation of dry-ice is also not supported.

5.2. LedaFlow

LedaFlow® is a transient multiphase flow simulation tool developed in the early 2000s by Total, ConocoPhilips and SINTEF. Today, it is being further developed for the commercial market by Kongsberg Oil & Gas Technologies.

The LedaFlow model is mainly developed for three-phase oil-gas-water mixtures, and the basic model solves 15 transport equations for nine fluids (Danielson *et al.*, 2011; Johansen, 2012): Nine mass equations govern the conservation of the mass in the bulk phases as well as immersed droplets and bubbles in each. Also, three momentum and energy equations are used. For thermodynamics, the model uses the SRK and Peng–Robinson equations of state.

While the standard LedaFlow described above applies to oil-gas-water mixtures, the framework and formulation is generally applicable for multiphase flow, and can in principle be applied to CO_2 transport. This, however, requires the implementation of closure relations relevant to CO_2 and the relevant impurities.

5.3. TACITE/PIPEPHASE

TACITE is a transient multicomponent, multiphase flow simulation tool developed by Elf Aquitaine/Total in the early 1990s. The tool has been developed mainly for simulating natural gas transport. TACITE is currently licensed as an add-on module to PIPEPHASE (Cos, 2012).

The underlying multifluid model of TACITE is described by Pauchon *et al.* (1994). It is a drift-flux model with one mass-conservation equation for each phase, one mixture momentum conservation equation and one mixture energy conservation equation. In addition, the model contains a flow-regime dependent closure law governing the momentum exchange between phases. For thermodynamics, TACITE uses tabulated values for the fluid properties as a function of pressure and temperature.

While the basic formulation of the model in TACITE is quite general, it uses closure relations and thermodynamics based of flow regimes and tabulated properties. TACITE considers eight types of flow regimes: Single-phase liquid, dispersed, slug, annular dispersed, stratified smooth, stratified wavy, annular and single-phase gas. The characterization of – and transition between – these flow regimes is highly dependent on the fluid. The models of TACITE have been developed and validated for natural gas transport, and their validity to CO_2 is not clear.

5.4. PipeTech

PipeTech is a transient multicomponent simulation tool developed and maintained by professor Haroun Mahgerefteh at Interglobe ltd. The main focus of PipeTech is the simulation of transient behaviour related to accidental depressurization and catastrophic failure of pipelines. The tool is used by the petroleum industry for safety assessment.

The PipeTech model employs the homogeneous equilibrium formulation of the transport equations (Mahgerefteh and Atti, 2006; Mahgerefteh *et al.*, 2011). It solves one mass equation, one momentum equation and one energy equation for the homogeneous mixture. A feature of this tool is the ability to model the evolution of pipeline cracks via a coupled fluid-fracture model. This enables the study of running ductile fractures.

PipeTech has a thermodynamics module taking account of CO_2 with impurities (Mahgerefteh *et al.*, 2012a).

6. Conclusion

In this paper, we have reviewed the state of the art for the modelling of transient flow of CO_2 mixtures in pipes. A main point of interest has been the modelling of the depressurization related to running ductile fracture, since this forms an important part of safety and design analyses. Running ductile fracture is a coupled fluid-structure problem, since the pipe influences the fluid flow, and vice versa.

The transport of CO_2 will often take place at a supercritical pressure. Therefore, in most cases, phase transfer will occur during a depressurization. In coupled fluid-structure simulations of running ductile fractures, it is important to correctly capture the wave-propagation speed in the fluid, as well as the crack-propagation speed in the pipe material. In two- or multiphase flow, the wave-propagation speed (speed of sound) is not a purely thermodynamic function, but it is also a function of the flow topology. In particular, the predicted two-phase speed of sound is a function of the assumptions regarding equilibrium in velocity, pressure, temperature and chemical potential. It should be noted that the common assumption of full equilibrium gives a discontinuous speed of sound in the limit of single-phase flow. Experimental data for the two-phase wave-propagation speed of relevant CO_2 mixtures would be useful not only for model validation, but also to gain insight into the applicability of different mathematical formulations of two-phase flow models, such as the homogeneous equilibrium model versus the two-fluid model, etc.

The thermodynamic properties of pure CO_2 at equilibrium are well described e.g. using the Span–Wagner reference EOS. Similar reference EOS'es for CCS-relevant impurities are under development. Further insight into the proper modelling of departure from thermodynamic equilibrium is needed in order to avoid such non-physical model features as a discontinuous speed of sound at phase boundaries.

The gas and liquid in a CO_2 mixture will in general have different compositions. In addition, the gas and liquid are likely to have different velocities during a depressurization. Therefore, flow models intended to describe depressurization of CO_2 mixtures will need to include component tracking.

In some cases, the amount of impurities will be small. Therefore, the flow models should also be able to handle the situation when a phase envelope turns into a line for a vanishing fraction of impurities.

Due to the high triple-point pressure of CO_2 (518 kPa), models intended to accurately simulate depressurization down to atmospheric pressure will need to take into account the formation of dry ice.

Some commonly used commercial tools for simulating transient multiphase pipeline transport have been screened. The tools available today have been developed for natural gas transport. The multifluid transport models used in such tools can in principle be generalized to model any liquid with impurities. However, the closure terms that are employed are often based on empirical models highly adapted to the original oil-gas-water application.

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