



Anomalous nucleation near a fluid phase boundary created by a rapid heat pulse



Romain Demur, Jed Oukmal, Guillaume Luu, Cécile Mousset, Marie Leman, Gustavo Villares, Margaux Kerdraon, Matthew Sullivan, Christopher Harrison*

Sensor Physics Department, Schlumberger-Doll Research, 1 Hampshire Street, Cambridge MA 02139, USA

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ABSTRACT

We report a novel method to rapidly nucleate vapor in a liquid with an electrically pulsed wire that temporarily raises the temperature of nearby liquid. This temperature increase is sufficient to overcome the nucleation barrier to vapor formation by locally putting the system in a non-equilibrium superheated state. By observing the subsequent behavior of bubbles after nucleation and thermal relaxation, we can determine the stability of the vapor phase. We map out a phase envelope by adjustment of the ambient temperature and pressure of the fluid around the wire. We find good agreement between measurements with this method and those made using a conventional apparatus. Surprisingly, we find that rapid heating of the liquid around the wire is able to produce nucleation in certain fluid mixtures for which the temperature increase is not expected to create an unstable or even metastable liquid. Evidence suggests a temporary enrichment of the region around the wire by lighter components due to the Soret effect is responsible for nucleation in such cases.

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1. Introduction

Knowledge of the phase behavior of reservoir hydrocarbon fluids is important to their efficient economic exploitation [1]. Hydrocarbons are found in reservoirs over a wide range of pressures and temperatures with complex geochemical development paths; the resulting distribution of molecular weights and architectures creates a complex phase behavior [2–4]. Phase behavior of reservoir fluids is typically determined by acquiring downhole samples, transporting them to a surface laboratory, and measuring, among other properties, the bubble or dew points (P_b or P_d , respectively). [5,6] Rapid and accurate measurement of the saturation pressure of multi-component fluids is challenging in part due to the nucleation barrier that must be overcome to form a second phase [7–12]. In response, we have developed a non-mechanical thermal nucleation method that uses millijoule heat pulses produced by a fine wire to temporarily overcome the nucleation barrier [13]. In the course of doing so, we have discovered that a second (vapor) phase can be anomalously nucleated at pressures and temperatures far outside of the phase envelope.

Calculations suggest that anomalous nucleation is a consequence of the Soret effect (thermodiffusion) which creates a temporary compositional inhomogeneity.

There is a rich literature on nucleation and boiling resulting from fast heat pulses emanating from thin wires immersed in single-component fluids such as water, but little is known about similar phenomena in multi-component fluids [14–17]. Rapid heating is used in a variety of microfluidic applications, such as for bubble-based pumps or for ink-jet printer heads [18]. In these cases, the heat pulse sufficiently superheats the fluid such that nucleation becomes kinetically favorable and occurs on short timescales. However, almost no work has been performed on multi-component systems above their critical pressures, where thermal pulses would not supersaturate the fluid and the formation of a separate phase would not be thermodynamically favorable, one of the topics of this manuscript.

2. Experimental

Hydrocarbon mixtures of liquid n-alkanes, methane and ethane were procured from conventional sources such as Fisher Scientific and American Gas Products, respectively, and prepared in high pressure cylinders by rocking at high pressure (see Table 1). Several modeling programs were used, including GERG-2008 [19],

* Corresponding author.

E-mail address: ckh.prince@gmail.com (C. Harrison).

Table 1

Provenance table of materials used in this manuscript and their sources.

Compound	Source	Supplier purity	Supplier method
methane	American Gas Products	99.999% (Grade 5)	GC
ethane	American Gas Products	99% (Grade 2)	GC
<i>n</i> -pentane	Fischer Scientific	>99%	GC
<i>n</i> -hexane	Fischer Scientific	98%	GC
<i>n</i> -octane	Sigma–Aldrich	>99%	GC
<i>n</i> -decane	Acros Organics	99%	GC

proprietary software by Schlumberger, and REFPROP from the National Institute of Standards and Technology. Equation of State (EOS) modeling was used as a guide to formulating the appropriate mixing ratios of the two components such that the cricondenbar (highest pressure of two-phase coexistence) was accessible for our apparatus, but the calculated bubble point pressures were often insufficiently accurate to be used a reference for the fluid mixtures we investigated. Hence when evaluating the accuracy of our measurements, a conventional, stirred high pressure JEFRI PVT view cell was used (DBR Technology Center in Edmonton, CA), referred to as the Conventional View Cell (CVC) [20]. As indicated in the text, critical points were taken directly from literature. Experiments described here were performed with fluid mixtures below their critical temperature and hence depressurization from a high pressure single-phase state created bubbles of vapor and not droplets of condensate. A complete list of the composition of the mixtures used can be found in Table 2 in this article.

Mixtures were charged into a custom Phase Transition Cell (PTC) with two high pressure spherical sapphire windows of diameter 2 mm, two fluidic connections, and two electrical feedthroughs, the latter connecting to a wire made of Nichrome or platinum of diameter *c.a.* 25 μm (Fig. 1A); both were used during different stages of our research. The wire is straight and centered in the optical and fluidic path. Broadband illumination was directed through one window via an optical fiber and measured via a similar fiber through the second window. Further details of the PTC, which has an internal volume of 5 μL , can be found in United States patents 8910514, 8950246, references therein [21]. To initiate nucleation, a current source of *c.a.* 10 A was connected to the wire with a duration that could be varied from 4 to 100 μs ; the voltage could be varied from 10 to 25 V (Fig. 1B). We typically found that 10 μs was sufficient, but for fluids with a low amount of dissolved gas (less than 10% by mole), 20 μs was found to create a more easily detectable signal. This allows ohmic heating in the wire to be adjusted from a few Kelvin to over one thousand Kelvin, though due to the short duration and low duty cycle, measurements showed that the average temperature of the PTC itself never increased more

Table 2Footnote: Expected uncertainty in molar percentage is $\pm 3\%$.

Sample	Mole % (1)	Mole % (2)
Methane (1) + <i>n</i> -Octane (2) Mixture 1	72	28
Methane (1) + <i>n</i> -Decane (2) Mixture 2	86	14
Ethane (1) + <i>n</i> -Pentane (2) EP1	22	78
EP2	26	74
EP3	42	58
EP4	56	44
EP5	75	25
EP6	83	17
EP7	93	7
EP8	97	3
EP9	98	2

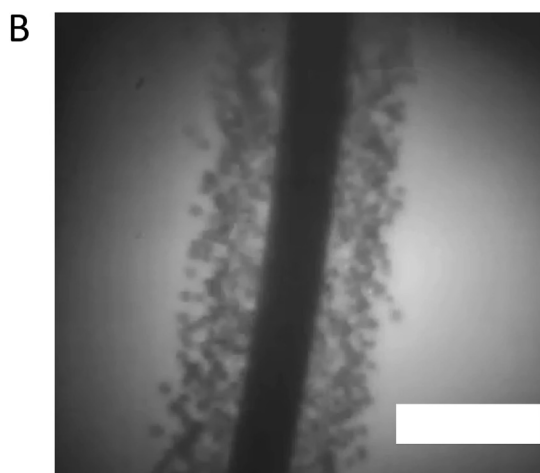
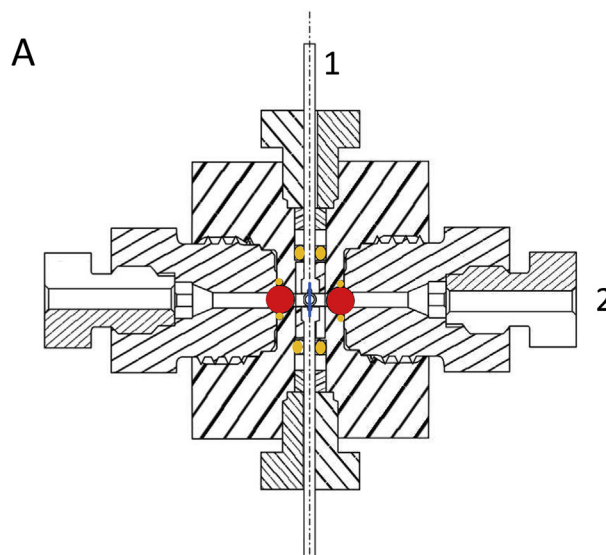
100 μm

Fig. 1. A) Schematic of phase transition cell (PTC) used to measure bubblepoint pressure. High pressure fluid is charged into cell through flowpath represented by unfilled circle (oriented normal to page and centered in between red sapphire lenses). Vertical blue line represents the nucleation wire. Two sapphire windows (drawn here in red) allow light to pass through the cell from the source on the right to a photodiode or for direct observation on the left. O-rings are represented in orange. Electrical feedthroughs (indicated by number 1) direct current through fine wire centered in flowline. Glands (indicated by number 2) secure optical fibers. B) Image of bubbles in an alkane mixture nucleated by black wire, *c.a.* 100 ms after the 10 μs thermal pulse.

than few millikelvin. A full discussion and characterization of the magnitude of the temperature pulse will be discussed in a separate manuscript [22]. A Quartzdyne pressure gauge (model QMB102-16-177) was used to measure the fluid pressure during depressurization and a small piston was used to control pressure. The heat pulses and resulting bubbles are sufficiently small that the thermal pulse did not cause the pressure of the system to change. The system was typically depressurized at 10 psi/second (0.069 MPa/s), starting with the hydrocarbon mixture in single phase. Results obtained with the PTC were compared with those obtained with a CVC.

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