High Pressure Multicomponent Fluids

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HIGH PRESSURE MULTICOMPONENT FLUIDS

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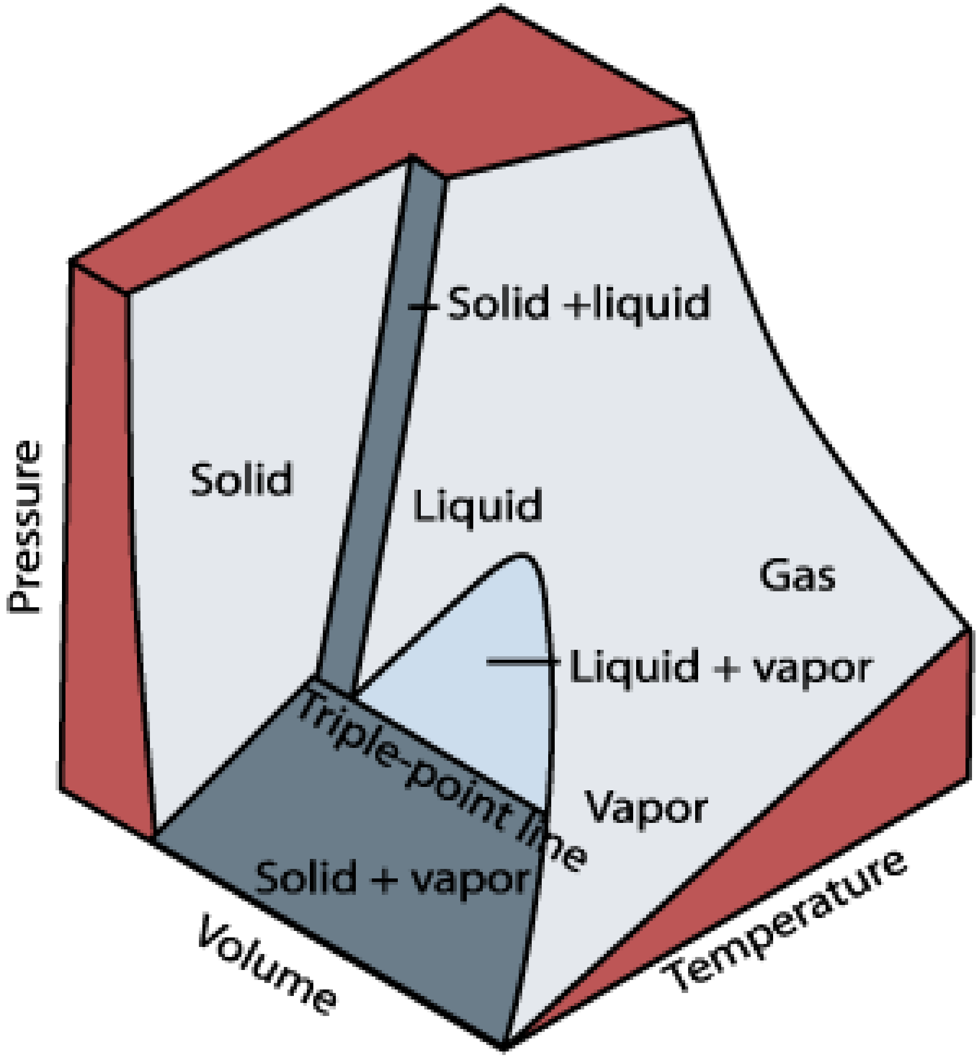
1 Introduction

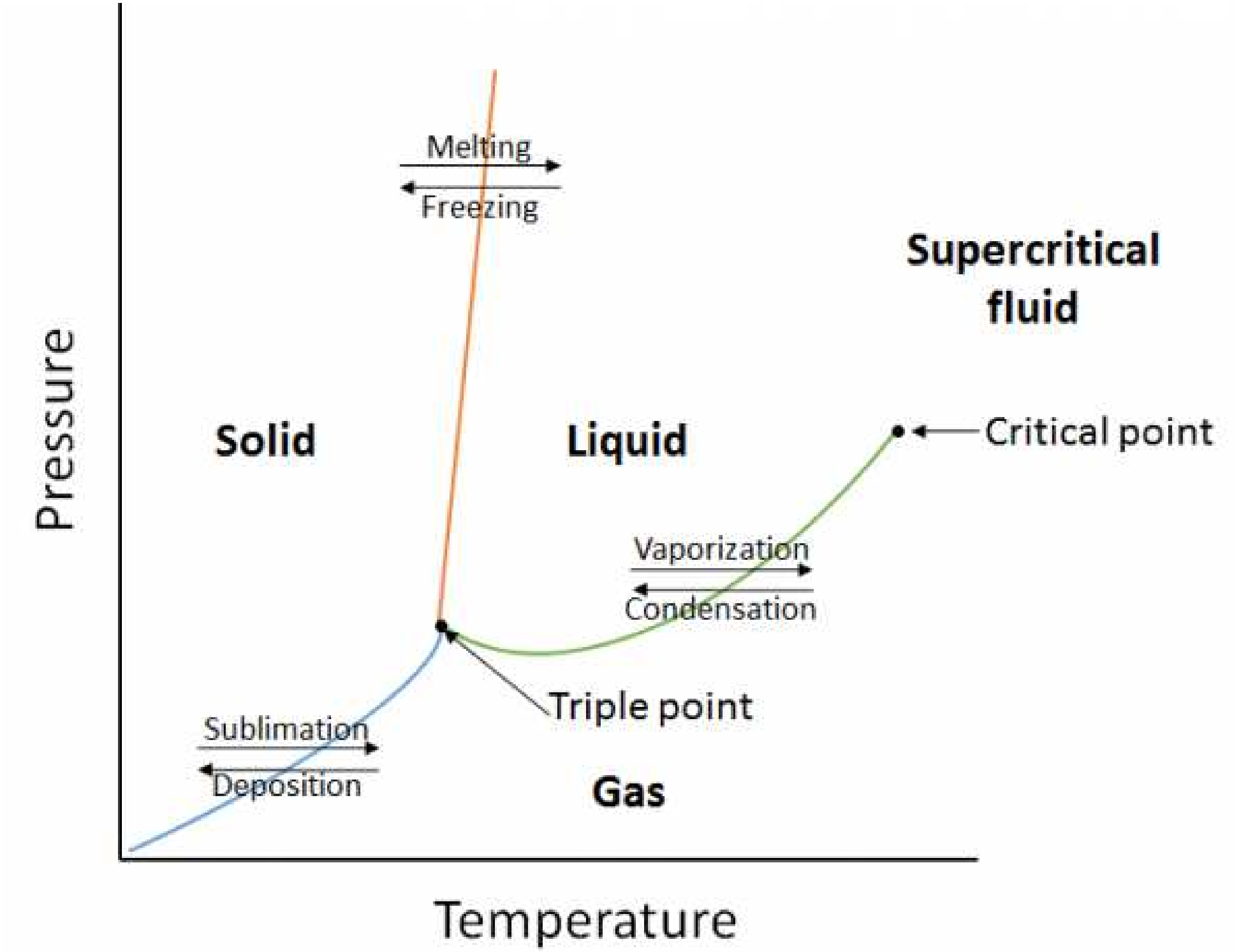
High pressure fluids (1)

- **Thermodynamic equilibrium surface for a single species**



High pressure fluids (2)

- **Projection onto the (T, p) plane**



High pressure fluids (3)

- (T, p) projection for binary mixtures (Van Konynenburg and Scott)

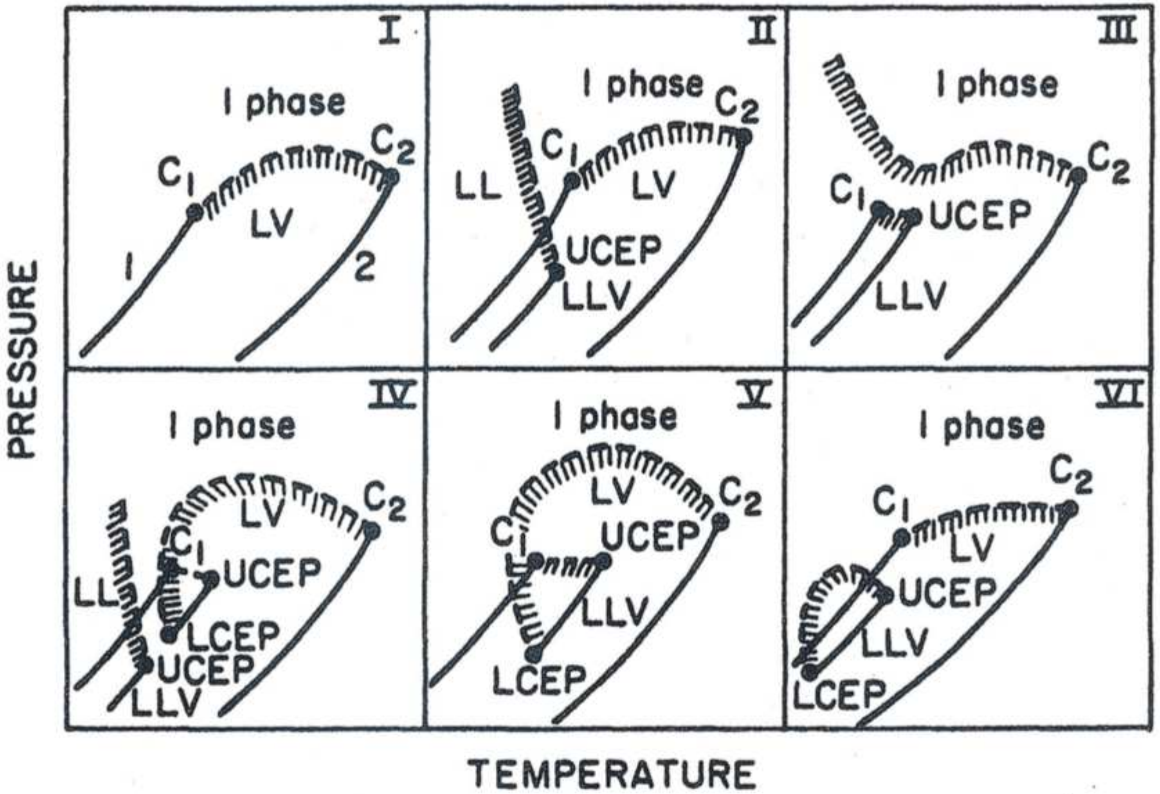


Figure 12-6 Six types of phase behavior in binary fluid systems. C = critical point; L = liquid; V = vapor; UCEP = upper critical end point; LCEP = lower critical end point. Dashed curves are critical lines and hatching marks heterogeneous regions.

High pressure fluids (4)

- **Applications**

  Ariane rocket Vulcain engine

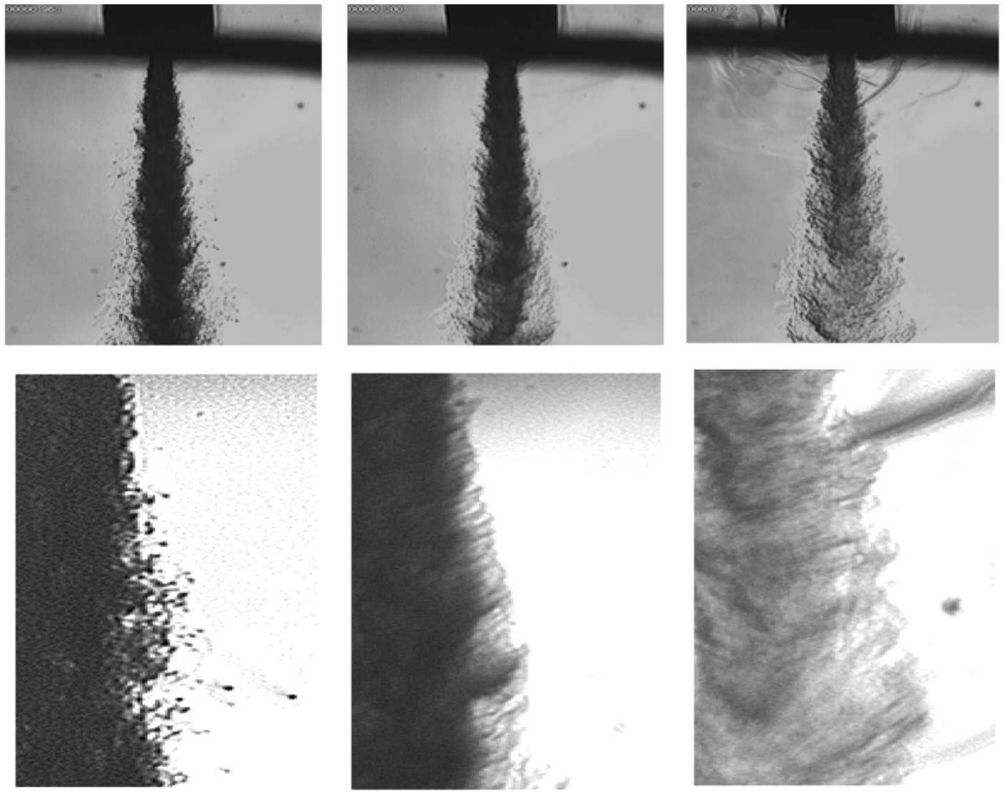
  Coal Power plants

  Extraction (supercritical CO_2)

  Depollution (supercritical CO_2 and H_2O)

High pressure fluids (5)

- **Cryogenic liquid N_2 at 99-110 K jets into gaseous N_2 at 300 K**



(a) Pr = 0.91 Re = 75,281 | (b) Pr = 1.22 Re = 66,609 | (c) Pr = 2.71 Re = 42,830

2 High Pressure Multicomponent Fluid Model

High pressure multicomponent fluid models (0)

- **Kinetic theory of dense gases**

  Enskog (1922), Thorne (Unpublished), Hirschfelder, Curtiss and Bird (1954)

  Chapman and Cowling (1970), Ferziger and Kaper (1972)

  Bajaras, Garcia-Colin and Piña (1973), Van Beijeren and Ernst (1973)

  Kurochkin, Makarenko and Tirskii (1984)

- **Statistical mechanics**

  Irving and Kirkwood (1950), Bearman and Kirkwood (1958), Mori (1958)

- **Thermodynamics of irreversible processes, statistical thermodynamics**

  Marcelin (1910), Meixner (1943), Prigogine (1947), de Groot and Mazur (1984), Keizer (1987)

High pressure multicomponent fluid models (1)

- **Multicomponent diffuse interface fluid model**

$$\partial_t \rho_i + \boldsymbol{\nabla}\cdot(\rho_i \boldsymbol{v}) + \boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{F}}_i = m_i \omega_i \qquad i \in \mathfrak{S} = \{1, \ldots, \mathsf{n}_{\mathrm{s}}\}$$

$$\partial_t(\rho \boldsymbol{v}) + \boldsymbol{\nabla}\cdot(\rho \boldsymbol{v}\otimes\boldsymbol{v}) + \boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{P}} = 0$$

$$\partial_t\big(\mathcal{E} + \tfrac{1}{2}\rho|\boldsymbol{v}|^2\big) + \boldsymbol{\nabla}\cdot\big(\boldsymbol{v}(\mathcal{E} + \tfrac{1}{2}\rho|\boldsymbol{v}|^2)\big) + \boldsymbol{\nabla}\cdot(\boldsymbol{\mathcal{Q}} + \boldsymbol{\mathcal{P}}\cdot\boldsymbol{v}) = 0$$

- **Multicomponent fluxes**

$$\boldsymbol{\mathcal{P}} = p\boldsymbol{I} + \varkappa\boldsymbol{\nabla}\rho\otimes\boldsymbol{\nabla}\rho - \rho\boldsymbol{\nabla}\cdot(\varkappa\boldsymbol{\nabla}\rho)\boldsymbol{I} + \boldsymbol{\mathcal{P}}^{\mathrm{d}}, \qquad \boldsymbol{\mathcal{Q}} = \varkappa\rho\boldsymbol{\nabla}\rho\boldsymbol{\nabla}\cdot\boldsymbol{v} + \boldsymbol{\mathcal{F}}_{\mathrm{e}}$$

$$\boldsymbol{\mathcal{P}}^{\mathrm{d}} = -\mathfrak{v}\boldsymbol{\nabla}\cdot\boldsymbol{v}\,\boldsymbol{I} - \eta\big(\boldsymbol{\nabla}\boldsymbol{v} + \boldsymbol{\nabla}\boldsymbol{v}^t - \tfrac{2}{d}\boldsymbol{\nabla}\cdot\boldsymbol{v}\,\boldsymbol{I}\big)$$

$$\boldsymbol{\mathcal{F}}_i = -\sum_{j\in\mathfrak{S}} L_{ij}\boldsymbol{\nabla}\Big(\frac{g_j}{T}\Big) - L_{i\mathrm{e}}\boldsymbol{\nabla}\Big(\frac{-1}{T}\Big) \qquad i \in \mathfrak{S}$$

$$\boldsymbol{\mathcal{F}}_{\mathrm{e}} = -\sum_{i\in\mathfrak{S}} L_{\mathrm{e}i}\boldsymbol{\nabla}\Big(\frac{g_i}{T}\Big) - L_{\mathrm{ee}}\boldsymbol{\nabla}\Big(\frac{-1}{T}\Big)$$

High pressure multicomponent fluid models (2)

- **Extended thermodynamics**

$$\mathcal{F} = \mathcal{F}^{\rm cl}(\rho_1,\dots,\rho_{\mathsf{n}_{\rm s}},T) + \tfrac{1}{2}\varkappa|\boldsymbol{\nabla}\rho|^2 \qquad \mathcal{S} = \mathcal{S}^{\rm cl}(\rho_1,\dots,\rho_{\mathsf{n}_{\rm s}},T) - \tfrac{1}{2}\partial_T\varkappa|\boldsymbol{\nabla}\rho|^2$$

$$p = p^{\rm cl}(\rho_1,\dots,\rho_{\mathsf{n}_{\rm s}},T) - \tfrac{1}{2}\varkappa|\boldsymbol{\nabla}\rho|^2 \qquad g_i = g_i^{\rm cl}(\rho_1,\dots,\rho_{\mathsf{n}_{\rm s}},T) \qquad i \in \mathfrak{S}$$

$$\mathcal{E} = \mathcal{E}^{\rm cl}(\rho_1,\dots,\rho_{\mathsf{n}_{\rm s}},T) + \tfrac{1}{2}(\varkappa - T\partial_T\varkappa)|\boldsymbol{\nabla}\rho|^2 \qquad \varkappa = \varkappa(T)$$

- **Gibbs relation**

$$T\,d\mathcal{S} = d\mathcal{E} - \sum_{i\in\mathfrak{S}} g_i\,d\rho_i - \varkappa\boldsymbol{\nabla}\rho\boldsymbol{\cdot}\,d\boldsymbol{\nabla}\rho$$

High pressure multicomponent fluid models (3)

- **Thermodynamics : $\mathcal{E}^{\rm cl}$, $p^{\rm cl}$, $\mathcal{S}^{\rm cl}$ functions of $\mathcal{Z} = (\rho_1, \ldots, \rho_{\mathsf{n_s}}, T)^t$ with**

  $(\mathcal{T}_0)$ $\mathcal{E}^{\rm cl}$, $p^{\rm cl}$, $\mathcal{S}^{\rm cl}$ are $C^\gamma(\mathcal{O}_\mathcal{Z})$, $\mathcal{O}_\mathcal{Z} \subset (0,\infty)^{\mathsf{n_s}+1}$ nonempty open connected

  $(\mathcal{T}_1)$ $T\, d\mathcal{S}^{\rm cl} = d\mathcal{E}^{\rm cl} - \sum_{i\in\mathfrak{S}} g_i^{\rm cl}\, d\rho_i$ with $g_k^{\rm cl} = \partial_{\rho_k}\mathcal{E}^{\rm cl} - T\partial_{\rho_k}\mathcal{S}^{\rm cl}$ and $\mathcal{G}^{\rm cl} = \sum_{i\in\mathfrak{S}} \rho_i g_i^{\rm cl}$

  $(\mathcal{T}_2)$ For any $(\mathsf{y}_1, \ldots, \mathsf{y}_{\mathsf{n_s}}, T) \in (0,\infty)^{\mathsf{n_s}+1}$ with $\sum_{i\in\mathfrak{S}} \mathsf{y}_i = 1$ $\quad \exists \rho_{\rm m} > 0$
  $(\rho\mathsf{y}_1, \ldots, \rho\mathsf{y}_{\mathsf{n_s}}, T)^t \in \mathcal{O}_\mathcal{Z}$ for $0 < \rho < \rho_{\rm m}$ and

  $$\lim_{\rho\to 0} (\mathcal{E}^{\rm cl} - \mathcal{E}^{\rm id})/\rho = 0 \qquad \lim_{\rho\to 0} (p^{\rm cl} - p^{\rm id})/\rho = 0 \qquad \lim_{\rho\to 0} (\mathcal{S}^{\rm cl} - \mathcal{S}^{\rm id})/\rho = 0$$

  $(\mathcal{T}_3)$ $\mathcal{O}_\mathcal{Z}$ is increasing with T and $\partial_T \mathcal{E}^{\rm cl} > 0$

- **The matrix $\boldsymbol{\Lambda} = (\boldsymbol{\Lambda}_{ij})_{i,j\in\mathfrak{S}}$ associated with stability**

  $\mathcal{Z} \mapsto \mathcal{X} = (\rho_1, \ldots, \rho_{\mathsf{n_s}}, \mathcal{E}^{\rm cl})^t$ C^γ diffeomorphism $\qquad \Lambda_{ij} = \Lambda_{ij}^{\rm cl} = \partial_{\rho_j} g_i / T$

  $\partial^2_{\mathcal{X}\mathcal{X}} \mathcal{S}^{\rm cl}$ negative definite $\iff$ $\partial_T \mathcal{E}^{\rm cl} > 0$ and Λ positive definite

High pressure multicomponent fluid models (4)

- **Complex chemistry**

$$\sum_{i\in\mathfrak{S}} \nu^{\mathrm{f}}_{ij}\mathfrak{M}_i \rightleftarrows \sum_{i\in\mathfrak{S}} \nu^{\mathrm{b}}_{ij}\mathfrak{M}_i \qquad j \in \mathfrak{R} = \{1,\dots,\mathsf{n}_{\mathrm{r}}\}$$

- **Reduced chemical potential** $\boldsymbol{\mu_i = m_i g_i / RT}$

$$\nu^{\mathrm{f}}_j = \begin{pmatrix} \nu^{\mathrm{f}}_{1j} \\ \vdots \\ \nu^{\mathrm{f}}_{\mathsf{n}_{\mathrm{s}}j} \end{pmatrix} \qquad \nu^{\mathrm{b}}_j = \begin{pmatrix} \nu^{\mathrm{b}}_{1j} \\ \vdots \\ \nu^{\mathrm{b}}_{\mathsf{n}_{\mathrm{s}}j} \end{pmatrix} \qquad \mu = \begin{pmatrix} \mu_1 \\ \vdots \\ \mu_{\mathsf{n}_{\mathrm{s}}} \end{pmatrix} \qquad \omega = \begin{pmatrix} \omega_1 \\ \vdots \\ \omega_{\mathsf{n}_{\mathrm{s}}} \end{pmatrix} \qquad \mathfrak{a}_l = \begin{pmatrix} \mathfrak{a}_{1l} \\ \vdots \\ \mathfrak{a}_{\mathsf{n}_{\mathrm{s}}l} \end{pmatrix}$$

- **Atom and mass conservation**

$$\mathfrak{A} = \{1,\dots,n^a\} \qquad \mathfrak{m} = (m_1,\dots,m_{\mathsf{n}_{\mathrm{s}}})^t \qquad \mathfrak{m} = \sum_{l\in\mathfrak{A}} \widetilde{m}_l\, \mathfrak{a}_l$$

$$\langle \nu_j, \mathfrak{a}_l \rangle = 0, \qquad j \in \mathfrak{R}, \quad l \in \mathfrak{A}, \qquad \langle \nu_j, \mathfrak{m} \rangle = 0$$

Multicomponent Diffuse Interface Fluids (5)

- **Marcelin's production rates**

$$\nu_j = \nu_j^{\mathrm{b}} - \nu_j^{\mathrm{f}} \qquad \omega = \sum_{j \in \mathfrak{R}} \nu_j \tau_j \qquad \tau_j = \mathcal{K}_j \big(\exp\langle \nu_j^{\mathrm{f}}, \mu \rangle - \exp\langle \nu_j^{\mathrm{b}}, \mu \rangle \big)$$

- **Activitiy and generalized mass action law**

$$\mu_i^{\mathrm{id}} = \mu_i^{\mathrm{u,id}}(T) + \log \gamma_i^{\mathrm{id}} \qquad \gamma_i^{\mathrm{id}} = \frac{\rho_i^{\mathrm{id}}}{m_i} \qquad \mathrm{a}_i = \widetilde{\mathrm{a}}_i \, \gamma_i^{\mathrm{id}}$$

$$\mu_i = \mu_i^{\mathrm{u,id}}(T) + \log \mathrm{a}_i, \qquad \tau_j = \mathcal{K}_j^{\mathrm{f}} \prod_{i \in \mathfrak{S}} \mathrm{a}_i^{\nu_{ij}^{\mathrm{f}}} - \mathcal{K}_j^{\mathrm{b}} \prod_{i \in \mathfrak{S}} \mathrm{a}_i^{\nu_{ij}^{\mathrm{b}}}$$

Multicomponent Diffuse Interface Fluids (6)

- **The Matrix L**

$$L = \begin{pmatrix} L_{11} & \cdots & L_{1\mathsf{n_s}} & L_{1\mathrm{e}} \\ \vdots & \ddots & \vdots & \vdots \\ L_{\mathsf{n_s}1} & \cdots & L_{\mathsf{n_s}\mathsf{n_s}} & L_{\mathsf{n_s}e} \\ L_{\mathrm{e}1} & \cdots & L_{\mathrm{e}\mathsf{n_s}} & L_{\mathrm{ee}} \end{pmatrix} \qquad v = \frac{1}{T}\begin{pmatrix} g_1 \\ \vdots \\ g_{\mathsf{n_s}} \\ -1 \end{pmatrix} \qquad \boldsymbol{\mathcal{F}} = \begin{pmatrix} \boldsymbol{\mathcal{F}}_1 \\ \vdots \\ \boldsymbol{\mathcal{F}}_{\mathsf{n_s}} \\ \boldsymbol{\mathcal{F}}_{\mathrm{e}} \end{pmatrix} \qquad \boldsymbol{\mathcal{F}} = -L\boldsymbol{\nabla} v$$

- **Properties of the transport coefficients**

(Tr$_1$) *L, η, $\mathfrak{v}$ are C^γ functions of $Z \in \mathcal{O}_Z$*

(Tr$_2$) *The matrix $L = (L_{ij})_{1\leq i,j\leq \mathsf{n}_s+1}$ is symmetric positive semi-definite with $N(L) = \mathbb{R}\,(\mathbb{1}, 0)^t$ where $\mathbb{1} = \big(1, \ldots, 1\big)^t \in \mathbb{R}^{\mathsf{n}_s}$*

(Tr$_3$) $\eta > 0 \quad \mathfrak{v} \geq 0 \quad$ *and* $\quad \mathfrak{v} > 0 \quad$ *if* $\quad d = 1$

Multicomponent Diffuse Interface Fluids (7)

- **Entropy balance**

$$\partial_t \mathcal{S} + \boldsymbol{\nabla}\boldsymbol{\cdot}(\mathcal{S}\boldsymbol{v}) + \boldsymbol{\nabla}\boldsymbol{\cdot}\Big(-\sum_{i\in\mathfrak{S}} \frac{g_i}{T}\boldsymbol{\mathcal{F}}_i + \frac{1}{T}\boldsymbol{\mathcal{F}}_{\mathrm{e}}\Big) = \mathfrak{v}_{\nabla} + \mathfrak{v}_{\omega}$$

$$\mathfrak{v}_{\nabla} = -\sum_{i\in\mathfrak{S}} \boldsymbol{\nabla}\Big(\frac{g_i}{T}\Big)\boldsymbol{\cdot}\boldsymbol{\mathcal{F}}_i - \boldsymbol{\nabla}\Big(\frac{1}{T}\Big)\boldsymbol{\cdot}\boldsymbol{\mathcal{F}}_{\mathrm{e}} - \frac{1}{T}\boldsymbol{\nabla}\boldsymbol{v}\boldsymbol{:}\boldsymbol{\mathcal{P}}^{\mathrm{d}} \qquad \mathfrak{v}_{\omega} = -\sum_{i\in\mathfrak{S}} \frac{g_i m_i \omega_i}{T}$$

- **Nonnegative entropy production**

$$\mathfrak{v}_{\nabla} = \langle L\boldsymbol{\nabla} v, \boldsymbol{\nabla} v\rangle + \frac{\kappa}{T}(\boldsymbol{\nabla}\boldsymbol{\cdot}\boldsymbol{v})^2 + \frac{\eta}{2T}\Big|\boldsymbol{\nabla}\boldsymbol{v} + \boldsymbol{\nabla}\boldsymbol{v}^t - \tfrac{2}{d}\boldsymbol{\nabla}\boldsymbol{\cdot}\boldsymbol{v}\boldsymbol{I}\Big|^2,$$

$$\mathfrak{v}_{\omega} = \sum_{j\in\mathfrak{R}} R\mathcal{K}_j\big(\langle\nu_j^{\mathrm{f}},\mu\rangle - \langle\nu_j^{\mathrm{b}},\mu\rangle\big)\big(\exp\langle\nu_j^{\mathrm{f}},\mu\rangle - \exp\langle\nu_j^{\mathrm{b}},\mu\rangle\big)$$

3 Existence of Strong Solutions

Multicomponent Augmented System (1)

- **Extra unknown** $\boldsymbol{w} = \boldsymbol{\nabla}\rho$

$$\partial_t \boldsymbol{w} + \sum_{i\in\mathcal{D}} \partial_i \big(\boldsymbol{w}\, v_i + \rho \boldsymbol{\nabla} v_i \big) = 0 \qquad \mathcal{D} = \{\, 1, \dots, d \,\}$$

- **Augmented unknowns**

$$\mathsf{u} = \big(\rho_1, \dots, \rho_{\mathsf{n_s}}, \boldsymbol{w}, \rho\boldsymbol{v}, \mathcal{E} + \tfrac{1}{2}\rho|\boldsymbol{v}|^2 \big)^t \qquad \mathsf{z} = \big(\rho_1, \dots, \rho_{\mathsf{n_s}}, \boldsymbol{w}, \boldsymbol{v}, T \big)^t$$

- **New thermodynamic functions**

$$\mathcal{E} = \mathcal{E}^{\rm cl} + \tfrac{1}{2}(\varkappa - T\partial_T\varkappa)|\boldsymbol{w}|^2 \qquad \mathcal{S} = \mathcal{S}^{\rm cl} - \tfrac{1}{2}\partial_T\varkappa|\boldsymbol{w}|^2$$

$$p = p^{\rm cl} - \tfrac{1}{2}\varkappa|\boldsymbol{w}|^2 \qquad g_k = g_k^{\rm cl} \qquad k \in \mathfrak{S}$$

Multicomponent Augmented System (2)

- **Thermodynamic functions**

(H₁) *$\mathcal{E}$, p, $\mathcal{S}$ are C^γ functions of* $\mathsf{z} \in \mathcal{O}_{\mathsf{z}} \subset (0,\infty)^{\mathsf{n}_s} \times \mathbb{R}^d \times \mathbb{R}^d \times (0,\infty)$
$\mathcal{O}_{\mathsf{z}}$ *open set,* $\varkappa = \varkappa(T)$ *is* $C^{\gamma+1}$ *function of temperature* T
If $(\rho_1,\dots,\rho_{\mathsf{n}_s},T)^t \in \mathcal{O}_Z$, $(\rho_1,\dots,\rho_{\mathsf{n}_s},0,0,T)^t \in \mathcal{O}_{\mathsf{z}}$ *and If*
$(\rho_1,\dots,\rho_{\mathsf{n}_s},\boldsymbol{w},\boldsymbol{v},T)^t \in \mathcal{O}_{\mathsf{z}}$, $(\rho_1,\dots,\rho_{\mathsf{n}_s},T)^t \in \mathcal{O}_Z$

(H₂) $\mathcal{G} = \mathcal{E} + p - T\mathcal{S} = \sum_{i\in\mathfrak{S}} \rho_i g_i \quad T\, d\mathcal{S} = d\mathcal{E} - \sum_{i\in\mathfrak{S}} g_i\, d\rho_i - \varkappa \boldsymbol{w}\boldsymbol{\cdot} d\boldsymbol{w}$

(H₃) *The open set* $\mathcal{O}_{\mathsf{z}}$ *is increasing with temperature* T *and* $\partial_T \mathcal{E} > 0$

(H₄) *The capillarity coefficient is positive* $\varkappa > 0$ *over* $\mathcal{O}_{\mathsf{z}}$

(H₅) *The coefficients* $\mathfrak{v}$, η, *and the matrix* L *are* C^γ *functions over* $\mathcal{O}_{\mathsf{z}}$
We have $\eta > 0$, $\mathfrak{v} \geq 0$, $\mathfrak{v} + \eta(1 - \frac{2}{d}) > 0$, L *is symmetric positive semi-definite and* $N(L) = \mathbb{R}(1,\dots,1,0,0,0,0)^t$.

Multicomponent Augmented System (3)

- **Thermodynamic functions**

(H₆) $(\rho_1,\ldots,\rho_{\mathsf{n}_s},T) \mapsto \Big(\rho, \dfrac{g_2-g_1}{T},\ldots,\dfrac{g_{\mathsf{n}_s}-g_1}{T},T\Big)^t$ *is globally invertible.*

(H₇) *There exists* $\delta>0$ *such that the eigenvalues* $\lambda_1,\ldots,\lambda_{\mathsf{n}_s}$ *of* Λ *satisfy* $\lambda_i \geq \delta$ *for* $i\geq 2$.

(H₈) *Atom conservation* $\langle \nu_j, \mathfrak{a}_l\rangle = 0$ *for* $j\in\mathfrak{R}, l\in\mathfrak{A}$ *and* $m = \sum_{l\in\mathfrak{A}} \widetilde{m}_l\,\mathfrak{a}_l$.

(H₉) *The rate constants* $\mathcal{K}_j$, $j\in\mathfrak{R}$, *are* C^γ *positive functions of* $T>0$.

Multicomponent Augmented System (4)

Lemma 1. *Assuming* (H_1)-(H_2) *and that* $\mathsf{z} \mapsto \mathsf{u}$ *is locally invertible then*

$$\partial^2_{\mathsf{uu}}\mathcal{S} \text{ negative definite} \iff \partial_T \mathcal{E} > 0 \qquad \det(\Lambda) > 0 \qquad \text{and} \qquad \varkappa > 0$$

Lemma 2. *Assuming* (H_1)-(H_3) *then the map* $\mathsf{z} \mapsto \mathsf{u}$ *is a* C^γ *diffeomorphism from the open set* $\mathcal{O}_\mathsf{z}$ *onto an open set* $\mathcal{O}_\mathsf{u}$.

Lemma 3. *Assuming* (H_1), γ *given smooth positive function, and* $\delta > 0$ *given there exists a* $C^{\gamma-1}$ *function* m *such that* $\mathsf{m} \geq 0$

$$\mathsf{m} + \gamma \det \Lambda > 0$$

and $\mathsf{m} = 0$ *if* $\gamma \det \Lambda \geq \delta$.

Multicomponent Augmented System (5)

- **Augmented entropic variable**

$$\sigma = -\mathcal{S} \qquad \mathsf{v} = (\partial_{\mathsf{u}}\sigma)^t = \frac{1}{T}\Big(g_1 - \tfrac{1}{2}|\boldsymbol{v}|^2, \ldots, g_{\mathsf{n_s}} - \tfrac{1}{2}|\boldsymbol{v}|^2, \varkappa\,\boldsymbol{w}, \boldsymbol{v}, -1\Big)^t$$

- **Stable points**

$$\mathcal{O}_{\mathsf{z}}^{\mathrm{st}} = \{\, \mathsf{z} \in \mathcal{O}_{\mathsf{z}} \;\big|\; \Lambda > 0 \,\} = \{\, \mathsf{z} \in \mathcal{O}_{\mathsf{z}} \;\big|\; \det(\Lambda) > 0 \,\}$$

$\mathsf{u} \mapsto \mathsf{v}$ locally invertible around stable points with $\Lambda > 0$

- **Legendre transform $\mathcal{L}$ of entropy**

$$\mathcal{L} = \langle \mathsf{u}, \mathsf{v} \rangle - \sigma = \frac{1}{T}\big(p + \varkappa|\boldsymbol{w}|^2\big) \qquad \partial_{\mathsf{u}}\sigma = \mathsf{v}^t \qquad \partial_{\mathsf{v}}\mathcal{L} = \mathsf{u}^t$$

- **Convective fluxes**

$$\mathsf{F}_i = \big(\partial_{\mathsf{v}}(\mathcal{L}v_i)\big)^t \qquad \mathcal{L}_i = \mathcal{L}v_i$$

Multicomponent Augmented System (6)

- **New augmented form**

$$\partial_t \mathsf{u} + \sum_{i\in\mathcal{D}} \partial_i\big(\mathsf{F}_i + \mathsf{F}_i^{\mathrm{d}} + \mathsf{F}_i^{\mathrm{c}}\big) = \Omega$$

- **Augmented fluxes in the ith direction**

$$\mathsf{F}_i = \Big(\rho_1 v_i, \dots, \rho_{\mathsf{n}_\mathrm{s}} v_i, \boldsymbol{w} v_i, \rho\boldsymbol{v} v_i + (p + \varkappa|\boldsymbol{w}|^2)\mathbf{e}_i, (\mathcal{E} + \tfrac{1}{2}\rho|\boldsymbol{v}|^2 + p + \varkappa|\boldsymbol{w}|^2) v_i\Big)^t$$

$$\mathsf{F}_i^{\mathrm{d}} = \Big(\mathcal{F}_{1i}, \dots, \mathcal{F}_{\mathsf{n}_\mathrm{s} i}, 0_{d,1},\ \boldsymbol{\mathcal{P}}_i^{\mathrm{d}},\ \mathcal{F}_{\mathrm{e}i} + \sum_{j\in\mathcal{D}} \mathcal{P}_{ij}^{\mathrm{d}} v_j\Big)^t \qquad \boldsymbol{\mathcal{P}}_i^{\mathrm{d}} = (\mathcal{P}_{1i}^{\mathrm{d}}, \dots, \mathcal{P}_{di}^{\mathrm{d}})^t$$

$$\mathsf{F}_i^{\mathrm{c}} = \Big(0, \dots, 0, \rho\boldsymbol{\nabla} v_i, -\rho\boldsymbol{\nabla}(\varkappa w_i), \rho\varkappa\boldsymbol{w}\boldsymbol{\cdot}\boldsymbol{\nabla} v_i - \rho\boldsymbol{v}\boldsymbol{\cdot}\boldsymbol{\nabla}(\varkappa w_i)\Big)^t$$

$$\Omega = \Big(m_1\omega_1 \dots, m_{\mathsf{n}_\mathrm{s}}\omega_{\mathsf{n}_\mathrm{s}}, 0, 0, 0\Big)^t$$

- **Similar equivalence of both formulations**

Multicomponent Augmented System (7)

- **Convective, dissipative and capillary matrices**

$$\mathsf{A}_i = \partial_{\mathsf{u}} \mathsf{F}_i \qquad \mathsf{F}_i^{\mathrm{d}} = -\sum_{j\in\mathcal{D}} \mathsf{B}_{ij}^{\mathrm{d}} \partial_j \mathsf{u} \qquad \mathsf{F}_i^{\mathrm{c}} = -\sum_{j\in\mathcal{D}} \mathsf{B}_{ij}^{\mathrm{c}} \partial_j \mathsf{u}, \qquad i \in \mathcal{D}$$

- **Quasilinear form**

$$\partial_t \mathsf{u} + \sum_{i\in\mathcal{D}} \mathsf{A}_i(\mathsf{u}) \partial_i \mathsf{u} - \sum_{i,j\in\mathcal{D}} \partial_i \big(\mathsf{B}_{ij}^{\mathrm{d}}(\mathsf{u}) \partial_j \mathsf{u} \big) - \sum_{i,j\in\mathcal{D}} \partial_i \big(\mathsf{B}_{ij}^{\mathrm{c}}(\mathsf{u}) \partial_j \mathsf{u} \big) = \Omega(\mathsf{u})$$

A_i, $\mathsf{B}_{ij}^{\mathrm{d}}$, and $\mathsf{B}_{ij}^{\mathrm{c}}$, for $i, j \in \mathcal{D}$, have at least regularity $C^{\gamma-1}$ over $\mathcal{O}_{\mathsf{u}}$

- **Symmetrization**

Structure of the system of equations plus existence results

Symmetrized Multicomponent Augmented System (1)

- **Entropic symmetrization for stable points $\mathbf{u} = \mathbf{u}(\mathbf{v})$**

Entropic variable $(\partial_{\mathsf{u}}\sigma)^t = \frac{1}{T}\Big(g_1 - \frac{1}{2}|\boldsymbol{v}|^2, \ldots, g_{\mathsf{n_s}} - \frac{1}{2}|\boldsymbol{v}|^2, \varkappa\,\boldsymbol{w}, \boldsymbol{v}, -1\Big)^t$

$$\widetilde{\mathsf{A}}_0(\mathsf{v})\partial_t \mathsf{v} + \sum_{i\in\mathcal{D}} \widetilde{\mathsf{A}}_i(\mathsf{v})\partial_i \mathsf{v} - \sum_{i,j\in\mathcal{D}} \partial_i\big(\widetilde{\mathsf{B}}^{\mathrm{d}}_{ij}(\mathsf{v})\partial_j \mathsf{v}\big) - \sum_{i,j\in\mathcal{D}} \partial_i\big(\widetilde{\mathsf{B}}^{\mathrm{c}}_{ij}(\mathsf{v})\partial_j \mathsf{v}\big) = \widetilde{\Omega}(\mathsf{v})$$

$$\widetilde{\mathsf{A}}_0 = \partial_{\mathsf{v}}\mathsf{u} \quad \widetilde{\mathsf{A}}_i = \mathsf{A}_i\partial_{\mathsf{v}}\mathsf{u} \quad \widetilde{\mathsf{B}}^{\mathrm{d}}_{ij} = \mathsf{B}^{\mathrm{d}}_{ij}\partial_{\mathsf{v}}\mathsf{u} \quad \widetilde{\mathsf{B}}^{\mathrm{c}}_{ij} = \mathsf{B}^{\mathrm{c}}_{ij}\partial_{\mathsf{v}}\mathsf{u} \quad \widetilde{\Omega} = \Omega \quad \det \widetilde{\mathsf{A}}_0 = \frac{\rho^3 T^8}{\varkappa^3}\frac{\partial_T \mathcal{E}}{\det \Lambda}$$

- **Structure of entropic symmetrized system**

$\widetilde{\mathsf{A}}_0$ symmetric positive definite for stable points $\qquad \widetilde{\mathsf{A}}_i$ symmetric for $i \in \mathcal{D}$

$(\widetilde{\mathsf{B}}^{\mathrm{d}}_{ij})^t = \widetilde{\mathsf{B}}^{\mathrm{d}}_{ji} \qquad \displaystyle\sum_{i,j\in\mathcal{D}} \xi_i\xi_j \widetilde{\mathsf{B}}^{\mathrm{d}}_{ij}$ positive semi definite $\qquad (\widetilde{\mathsf{B}}^{\mathrm{c}}_{ij})^t = -\widetilde{\mathsf{B}}^{\mathrm{c}}_{ji}$

The map $\mathsf{u} \mapsto \mathsf{v}$ is generally not globally invertible

Symmetrized Multicomponent Augmented System (2)

- **Normal variable**

$$\mathsf{w} = (\mathsf{w}_{\mathrm{I}}, \mathsf{w}_{\mathrm{II}})^t = \left(\rho, \boldsymbol{w}, \frac{g_2 - g_1}{T}, \dots, \frac{g_{\mathsf{n}_{\mathrm{s}}} - g_1}{T}, \boldsymbol{v}, T\right)^t$$

$$\mathsf{w}_{\mathrm{I}} = (\rho, \boldsymbol{w})^t \qquad \mathsf{w}_{\mathrm{II}} = \left(\frac{g_2 - g_1}{T}, \dots, \frac{g_{\mathsf{n}_{\mathrm{s}}} - g_1}{T}, \boldsymbol{v}, T\right)^t$$

$\mathbb{R}^{\mathsf{n}} = \mathbb{R}^{\mathsf{n}_{\mathrm{I}}} \times \mathbb{R}^{\mathsf{n}_{\mathrm{II}}} \qquad \mathsf{n} = \mathsf{n}_{\mathrm{I}} + \mathsf{n}_{\mathrm{II}} \qquad \mathsf{n}_{\mathrm{I}} = d + 1 \qquad \mathsf{n}_{\mathrm{II}} = \mathsf{n}_{\mathrm{s}} + d$

$\mathsf{z} \to \mathsf{w}$ diffeomorphism from $\mathcal{O}_{\mathsf{z}}$ onto $\mathcal{O}_{\mathsf{w}}$ and $\mathsf{u} \to \mathsf{w}$ from $\mathcal{O}_{\mathsf{u}}$ onto $\mathcal{O}_{\mathsf{w}}$

- **Notation for more general systems**

$\mathsf{w}_{\mathrm{I}} = (\mathsf{w}_{\mathrm{I}'}, \mathsf{w}_{\mathrm{I}''})^t \quad \mathsf{w}_{\mathrm{I}'} = \rho \quad \mathsf{w}_{\mathrm{I}''} = \boldsymbol{w} \quad \boldsymbol{\nabla}\mathsf{w}_{\mathrm{I}'} = \mathsf{w}_{\mathrm{I}''} \quad \mathsf{w}_{\mathrm{r}} = (\mathsf{w}_{\mathrm{I}'}, \mathsf{w}_{\mathrm{II}})^t$

- **Normal form**

$\mathsf{u} = \mathsf{u}(\mathsf{w})$ and multiplication on the left by $(\partial_{\mathsf{w}}\mathsf{v})^t$

Add $\big(\partial_t \rho + \boldsymbol{\nabla}\cdot(\rho \boldsymbol{v})\big) \times \mathsf{m}$ to the first equation

Symmetrized Multicomponent Augmented System (3)

- **Normal form**

$$\overline{\mathsf{A}}_0(\mathsf{w})\partial_t \mathsf{w} + \sum_{i\in\mathcal{D}} \overline{\mathsf{A}}_i(\mathsf{w})\partial_i \mathsf{w} - \sum_{i,j\in\mathcal{D}} \overline{\mathsf{B}}^{\mathrm{d}}_{ij}(\mathsf{w})\partial_i\partial_j \mathsf{w} - \sum_{i,j\in\mathcal{D}} \overline{\mathsf{B}}^{\mathrm{c}}_{ij}(\mathsf{w})\partial_i\partial_j \mathsf{w} = \mathsf{h}(\mathsf{w}, \boldsymbol{\nabla}\mathsf{w})$$

- **Properties of the normal form**

$\overline{\mathsf{A}}_0 = \mathrm{diag}(\overline{\mathsf{A}}_0^{\mathrm{I,I}}, \overline{\mathsf{A}}_0^{\mathrm{II,II}})$ symmetric positive definite $\qquad \overline{\mathsf{A}}_i$ symmetric for $i \in \mathcal{D}$

$(\overline{\mathsf{B}}^{\mathrm{d}}_{ij})^t = \overline{\mathsf{B}}^{\mathrm{d}}_{ji} \qquad \overline{\mathsf{B}}^{\mathrm{d}}_{ij} = \mathrm{diag}(0, \overline{\mathsf{B}}^{\mathrm{d\,II,II}}_{ij}) \qquad \overline{\mathsf{B}}^{\mathrm{d\,II,II}} = \sum_{i,j\in\mathcal{D}} \xi_i\xi_j \overline{\mathsf{B}}^{\mathrm{d\,II,II}}_{ij}$ positive definite

$(\overline{\mathsf{B}}^{\mathrm{c}}_{ij})^t = -\overline{\mathsf{B}}^{\mathrm{c}}_{ji} \qquad \overline{\mathsf{B}}^{\mathrm{c\,I,I}}_{ij} = 0 \qquad \overline{\mathsf{B}}^{\mathrm{c\,I,II}}_{ij},\ \overline{\mathsf{B}}^{\mathrm{c\,II,I}}_{ij},\ \overline{\mathsf{A}}_0^{\mathrm{II,II}}$ depend on $\mathsf{w}_{\mathrm{r}} = (\mathsf{w}_{\mathrm{I}'}, \mathsf{w}_{\mathrm{II}})^t$

$$\mathsf{h} = (\mathsf{h}_{\mathrm{I}}, \mathsf{h}_{\mathrm{II}})^t \qquad \mathsf{h}_{\mathrm{I}} = \Big(-\mathsf{m}\rho\boldsymbol{\nabla}\cdot\boldsymbol{v}, -\frac{\varkappa}{T}\sum_{i\in\mathcal{D}} w_i \boldsymbol{\nabla} v_i\Big)^t \qquad \mathsf{h}_{\mathrm{II}} = \mathsf{h}_{\mathrm{II}}(\mathsf{w}, \boldsymbol{\nabla}\mathsf{w})$$

Coefficients are ate least $C^{\gamma-2}$ $\qquad$ Explicit calculations (formally singular)

Chemical terms included in h_{II} for local existence results

Symmetrized Multicomponent Augmented System (4)

- **Gradient constraint for nonlinear equations**

  Natural equation for $\boldsymbol{w} - \boldsymbol{\nabla}\rho$

$$\partial_t(\boldsymbol{w} - \boldsymbol{\nabla}\rho) + \boldsymbol{v}\cdot\boldsymbol{\nabla}(\boldsymbol{w} - \boldsymbol{\nabla}\rho) + (\boldsymbol{w} - \boldsymbol{\nabla}\rho)\boldsymbol{\nabla}\cdot\boldsymbol{v} + (\boldsymbol{\nabla}\boldsymbol{v})^t\cdot(\boldsymbol{w} - \boldsymbol{\nabla}\rho) = 0$$

  If w is smooth enough, $\boldsymbol{w}_0 - \boldsymbol{\nabla}\rho_0 = 0$ and $\boldsymbol{w}^\star = 0$ then $\boldsymbol{w} - \boldsymbol{\nabla}\rho = 0$

- **Linearized equation with gradient constraint**

$$\overline{\mathsf{A}}_0(\mathsf{w})\partial_t\widetilde{\mathsf{w}} + \sum_{i\in\mathcal{D}}\overline{\mathsf{A}}_i(\mathsf{w})\partial_i\widetilde{\mathsf{w}} - \sum_{i,j\in\mathcal{D}}\overline{\mathsf{B}}^{\mathrm{d}}_{ij}(\mathsf{w})\partial_i\partial_j\widetilde{\mathsf{w}} - \sum_{i,j\in\mathcal{D}}\overline{\mathsf{B}}^{\mathrm{c}}_{ij}(\mathsf{w})\partial_i\partial_j\widetilde{\mathsf{w}} =$$

$$\Big(-\mathsf{m}\,\rho\,\boldsymbol{\nabla}\cdot\widetilde{\boldsymbol{v}}, -\sum_{i\in\mathcal{D}}\frac{\varkappa}{T}\widetilde{w}_i\boldsymbol{\nabla}v_i, \mathsf{h}_{\mathrm{II}}(\mathsf{w},\boldsymbol{\nabla}\mathsf{w})\Big)^t$$

- **Similar to the single species situation**

Symmetrized Multicomponent Augmented System (5)

- **Linearized equation with gradient constraint**

$$\overline{\mathsf{A}}_0(\mathsf{w})\partial_t\widetilde{\mathsf{w}} + \sum_{i\in\mathcal{D}}\overline{\mathsf{A}}_i'(\mathsf{w})\partial_i\widetilde{\mathsf{w}} - \sum_{i,j\in\mathcal{D}}\overline{\mathsf{B}}_{ij}^{\mathrm{d}}(\mathsf{w})\partial_i\partial_j\widetilde{\mathsf{w}} - \sum_{i,j\in\mathcal{D}}\overline{\mathsf{B}}_{ij}^{\mathrm{c}}(\mathsf{w})\partial_i\partial_j\widetilde{\mathsf{w}}$$

$$+\overline{\mathsf{L}}(\mathsf{w},\boldsymbol{\nabla}\mathsf{w}_{\mathrm{II}})\widetilde{\mathsf{w}} = \mathsf{h}'(\mathsf{w},\boldsymbol{\nabla}\mathsf{w}) = \big(0,\mathsf{h}_{\mathrm{II}}(\mathsf{w},\boldsymbol{\nabla}\mathsf{w})\big)^t$$

$$\overline{\mathsf{A}}_i'(\mathsf{w}) = \overline{\mathsf{A}}_i(\mathsf{w}) + \mathsf{m}\rho\mathsf{e}_1\otimes\mathsf{e}_{d+1+i} \qquad \overline{\mathsf{L}}(\mathsf{w},\boldsymbol{\nabla}\mathsf{w}_{\mathrm{II}}) = \sum_{i\in\mathcal{D}}\frac{\varkappa}{T}(0,\boldsymbol{\nabla}v_i,0_{1,\mathsf{n}_{\mathrm{I}}},0)^t\otimes\mathsf{e}_{i+1}$$

- **Gradient constraint for linearized equations**

Natural equation for $\widetilde{\boldsymbol{w}} - \boldsymbol{\nabla}\widetilde{\rho}$

$$\partial_t(\widetilde{\boldsymbol{w}} - \boldsymbol{\nabla}\widetilde{\rho}) + \boldsymbol{v}\cdot\boldsymbol{\nabla}(\widetilde{\boldsymbol{w}} - \boldsymbol{\nabla}\widetilde{\rho}) + (\boldsymbol{w} - \boldsymbol{\nabla}\rho)\,\boldsymbol{\nabla}\cdot\widetilde{\boldsymbol{v}} + \boldsymbol{\nabla}\boldsymbol{v}^t\cdot(\widetilde{\boldsymbol{w}} - \boldsymbol{\nabla}\widetilde{\rho}) = 0$$

If w and $\widetilde{\mathsf{w}}$ are regular, $\boldsymbol{w} - \boldsymbol{\nabla}\rho = 0$, $\widetilde{\boldsymbol{w}}_0 - \boldsymbol{\nabla}\widetilde{\rho}_0 = 0$, $\widetilde{\boldsymbol{w}}^\star = 0$ then $\widetilde{\boldsymbol{w}} - \boldsymbol{\nabla}\widetilde{\rho} = 0$

Linearized Equations (1)

- **Linearized equations**

$$\overline{\mathsf{A}}_0(\mathsf{w})\partial_t\widetilde{\mathsf{w}} + \sum_{i\in\mathcal{D}} \overline{\mathsf{A}}_i'(\mathsf{w})\partial_i\widetilde{\mathsf{w}} - \sum_{i,j\in\mathcal{D}} \overline{\mathsf{B}}_{ij}^{\mathrm{d}}(\mathsf{w})\partial_i\partial_j\widetilde{\mathsf{w}} - \sum_{i,j\in\mathcal{D}} \overline{\mathsf{B}}_{ij}^{\mathrm{c}}(\mathsf{w})\partial_i\partial_j\widetilde{\mathsf{w}} + \overline{\mathsf{L}}(\mathsf{w},\nabla\mathsf{w}_{\mathrm{r}})\widetilde{\mathsf{w}} = \mathsf{f}+\mathsf{g}$$

- **Assumptions on the coefficients**

$\overline{\mathsf{A}}_0 = \mathrm{diag}(\overline{\mathsf{A}}_0^{\mathrm{I,I}}, \overline{\mathsf{A}}_0^{\mathrm{II,II}})$ symmetric positive definite block diagonal

$\overline{\mathsf{A}}_i'^{\mathrm{I,I}}$ are symmetric, $(\overline{\mathsf{B}}_{ij}^{\mathrm{d}})^t = \overline{\mathsf{B}}_{ji}^{\mathrm{d}}$, $\overline{\mathsf{B}}_{ij}^{\mathrm{d}} = \mathrm{diag}(0, \overline{\mathsf{B}}_{ij}^{\mathrm{d\,II,II}})$

$\overline{\mathsf{B}}^{\mathrm{d\,II,II}} = \sum_{i,j\in\mathcal{D}} \overline{\mathsf{B}}_{ij}^{\mathrm{d\,II,II}}\xi_i\xi_j$ is positive definite for $\boldsymbol{\xi}\in\Sigma^{d-1}$

$(\overline{\mathsf{B}}_{ij}^{\mathrm{c}})^t = -\overline{\mathsf{B}}_{ji}^{\mathrm{c}}$ $\quad \overline{\mathsf{B}}_{ij}^{\mathrm{c,I,I}} = 0$ $\quad \overline{\mathsf{A}}_0^{\mathrm{II,II}}$, $\overline{\mathsf{B}}_{ij}^{\mathrm{c,I,II}}$, $\overline{\mathsf{B}}_{ij}^{\mathrm{c,II,I}}$ only depend on $\mathsf{w}_{\mathrm{r}} = (\mathsf{w}_{\mathrm{I}'}, \mathsf{w}_{\mathrm{II}})^t$

$\overline{\mathsf{L}} = \mathrm{diag}(\overline{\mathsf{L}}^{\mathrm{I,I}}, \overline{\mathsf{L}}^{\mathrm{II,II}})$ $\quad \overline{\mathsf{L}}^{\mathrm{I,I}} = \mathfrak{L}^{\mathrm{I,I}}(\mathsf{w})\boldsymbol{\nabla}\mathsf{w}_{\mathrm{r}}$ $\quad \overline{\mathsf{L}}^{\mathrm{II,II}} = \mathfrak{L}^{\mathrm{II,II}}(\mathsf{w})\boldsymbol{\nabla}\mathsf{w}_{\mathrm{r}}$

$\overline{\mathsf{A}}_0$, $\overline{\mathsf{A}}_i'$, $\overline{\mathsf{B}}_{ij}^{\mathrm{d}}$, $\overline{\mathsf{B}}_{ij}^{\mathrm{c}}$, $\mathfrak{L}^{\mathrm{I,I}}$, $\mathfrak{L}^{\mathrm{II,II}}$ are C^{l+2} over $\mathcal{O}_{\mathsf{w}}$ $\qquad \overline{\mathsf{L}}(\mathsf{w},\nabla\mathsf{w}_{\mathrm{r}})\,\widetilde{\mathsf{w}}^\star = 0$

Linearized Equations (2)

- **Assumptions on w**

$d \geq 1 \quad l \geq l_0 + 2$ where $l_0 = [d/2] + 1 \quad 1 \leq l' \leq l$

w given function of $(t, \boldsymbol{x})$ over $[0, \bar{\tau}] \times \mathbb{R}^d$ with $\bar{\tau} > 0$

$$\begin{cases} \mathsf{w}_{\mathrm{I}} - \mathsf{w}_{\mathrm{I}}^{\star} \in C^0\big([0,\bar{\tau}], H^l\big) \cap C^1\big([0,\bar{\tau}], H^{l-2}\big) \\ \mathsf{w}_{\mathrm{II}} - \mathsf{w}_{\mathrm{II}}^{\star} \in C^0\big([0,\bar{\tau}], H^l\big) \cap C^1\big([0,\bar{\tau}], H^{l-2}\big) \cap L^2\big((0,\bar{\tau}), H^{l+1}\big) \end{cases}$$

$\mathcal{O}_0 \subset \overline{\mathcal{O}}_0 \subset \mathcal{O}_{\mathsf{w}},\ 0 < a_1 < \mathrm{dist}(\overline{\mathcal{O}}_0, \partial\mathcal{O}_{\mathsf{w}}),\ \ \mathcal{O}_1 = \{\, \mathsf{w} \in \mathcal{O}_{\mathsf{w}};\ \mathrm{dist}(\mathsf{w}, \overline{\mathcal{O}}_0) < a_1 \,\}$

$\mathsf{w}_0(\boldsymbol{x}) = \mathsf{w}(0, \boldsymbol{x}) \in \mathcal{O}_0,\ \mathsf{w}(t, \boldsymbol{x}) \in \mathcal{O}_1,\ (t, \boldsymbol{x}) \in [0, \bar{\tau}] \times \mathbb{R}^d$

- **Assumptions on f and g**

f and g given functions of $(t, \boldsymbol{x})$ over $[0, \bar{\tau}] \times \mathbb{R}^d \qquad 1 \leq l' \leq l$

$\mathsf{f} \in C^0\big([0,\bar{\tau}], H^{l'-1}\big) \cap L^1\big((0,\bar{\tau}), H^{l'}\big) \qquad \mathsf{g} \in C^0\big([0,\bar{\tau}], H^{l'-1}\big) \quad \mathsf{g}_{\mathrm{I}} = 0$

Linearized Equations (3)

- **Assumptions on $\widetilde{\mathsf{w}}$**

$$\widetilde{\mathsf{w}}_{\rm I} - \widetilde{\mathsf{w}}_{\rm I}^\star \in C^0\big([0,\bar\tau], H^{l'}\big) \cap C^1\big([0,\bar\tau], H^{l'-2}\big),$$

$$\widetilde{\mathsf{w}}_{\rm II} - \widetilde{\mathsf{w}}_{\rm II}^\star \in C^0\big([0,\bar\tau], H^{l'}\big) \cap C^1\big([0,\bar\tau], H^{l'-2}\big) \cap L^2\big((0,\bar\tau), H^{l'+1}\big),$$

- **Bounding quantities**

$$M^2 = \sup_{0\le\tau\le\bar\tau} |\mathsf{w}(\tau) - \mathsf{w}^\star|_l^2, \qquad M_1^2 = \int_0^{\bar\tau} |\partial_t \mathsf{w}(\tau)|_{l-2}^2\, d\tau, \qquad M_{\rm r}^2 = \int_0^{\bar\tau} |\nabla \mathsf{w}_{\rm r}(\tau)|_l^2\, d\tau$$

- **Linearized estimates for $1 \le l' \le l$**

There exists constants $\mathsf{c}_1(\mathcal{O}_1) \ge 1$ and $\mathsf{c}_2(\mathcal{O}_1, M) \ge 1$ increasing with M with

$$\sup_{0\le\tau\le t} |\widetilde{\mathsf{w}}(\tau) - \widetilde{\mathsf{w}}^\star|_{l'}^2 + \int_0^t |\widetilde{\mathsf{w}}_{\rm II}(\tau) - \widetilde{\mathsf{w}}_{\rm II}^\star|_{l'+1}^2\, d\tau \le \mathsf{c}_1^2 \exp\Big(\mathsf{c}_2\big(t + M_1\sqrt{t} + M_{\rm r}\sqrt{t}\,\big)\Big)\times$$

$$\Big(|\widetilde{\mathsf{w}}_0 - \widetilde{\mathsf{w}}^\star|_{l'}^2 + \mathsf{c}_2\Big\{\int_0^t |\mathsf{f}|_{l'}\, d\tau\Big\}^2 + \mathsf{c}_2 \int_0^t |\mathsf{g}_{\rm II}|_{l'-1}^2\, d\tau\Big)$$

Existence Results for Diffuse Interface Models

- **Isothermal**

  Hattori and Li (1996) Danchin and Desjardins (2001) Kotschote (2008)
  Bresch et al. (2003) (2019)

- **Euler-Korteweg**

  Bresch et al. (2008) (2019) Benzoni et al. (2005) (2006) (2007)
  Donatelli et al. (2004) (2014) Tzavaras et al. (2018) (2017)

- **Full model**

  Haspot (2009) Kotschote (2012) (2014)

- **Symmetrization for diffuse interface fluids**

  Gavrilyuk and Gouin (2000) Kawashima et al. (2022)

Existence of Strong Solutions (1)

- **Structural assumptions**

  Augmented system in normal form with the gradient constraint

  Linearized equations enforcing the gradient constraint

$$\big(\overline{\mathsf{A}}_i'(\mathsf{w}) - \overline{\mathsf{A}}_i(\mathsf{w})\big)\boldsymbol{\nabla}\mathsf{w} + \overline{\mathsf{L}}(\mathsf{w}, \boldsymbol{\nabla}\mathsf{w}_{\mathrm{r}})\mathsf{w} + \mathsf{h}(\mathsf{w}, \boldsymbol{\nabla}\mathsf{w}) = \mathsf{h}'(\mathsf{w}, \boldsymbol{\nabla}\mathsf{w})$$

  Right hand sides in the form

$$\mathsf{h}_{\mathrm{I}} = \sum_{i\in\mathcal{D}} \overline{\mathrm{M}}_i^{\mathrm{I}}(\mathsf{w})\partial_i \mathsf{w}_{\mathrm{r}} + \sum_{i,j\in\mathcal{D}} \overline{\mathrm{M}}_{ij}^{\mathrm{I},\mathrm{I}}(\mathsf{w})\partial_i \mathsf{w}_{\mathrm{r}} \partial_j \mathsf{w}_{\mathrm{r}}$$

$$\mathsf{h}_{\mathrm{II}} = \sum_{i\in\mathcal{D}} \overline{\mathrm{M}}_i^{\mathrm{II}}(\mathsf{w})\partial_i \mathsf{w} + \sum_{i,j\in\mathcal{D}} \overline{\mathrm{M}}_{ij}^{\mathrm{II},\mathrm{II}}(\mathsf{w})\partial_i \mathsf{w} \partial_j \mathsf{w}$$

  w_{r} is the more regular part $\mathsf{w}_{\mathrm{r}} = (\mathsf{w}_{\mathrm{I}'}, \mathsf{w}_{\mathrm{II}})^t$ of the normal variable

Existence of Strong Solutions (2)

Theorem 1. *Let* $d \geq 1$, $l \geq l_0 + 2$, $l_0 = [d/2] + 1$, *and let* $b > 0$. *Let* $\mathcal{O}_0 \subset \overline{\mathcal{O}}_0 \subset \mathcal{O}_{\mathsf{w}}$, $0 < a_1 < \mathrm{dist}(\overline{\mathcal{O}}_0, \partial\mathcal{O}_{\mathsf{w}})$, $\mathcal{O}_1 = \{\, \mathsf{w} \in \mathcal{O}_{\mathsf{w}} ; \mathrm{dist}(\mathsf{w}, \overline{\mathcal{O}}_0) < a_1 \,\}$. *There exists* $\bar{\tau}(\mathcal{O}_1, b) > 0$ *such that for any* w_0 *with* $\mathsf{w}_0 \in \mathcal{O}_0$, $\mathsf{w}_0 - \mathsf{w}^\star \in H^l$, $\mathsf{w}_{0\mathrm{I}''} = \nabla \mathsf{w}_{0\mathrm{I}'}$ *and*

$$|\mathsf{w}_0 - \mathsf{w}^\star|_l^2 < b^2,$$

there exists a unique local solution w *with initial condition* $\mathsf{w}(0, \boldsymbol{x}) = \mathsf{w}_0(\boldsymbol{x})$, *such that* $\mathsf{w}(t, \boldsymbol{x}) \in \mathcal{O}_1$ *for* $(t, \boldsymbol{x}) \in [0, \bar{\tau}] \times \mathbb{R}^d$, $\mathsf{w}_{\mathrm{I}''} = \nabla \mathsf{w}_{\mathrm{I}'}$, *and*

$$\mathsf{w}_{\mathrm{I}} - \mathsf{w}_{\mathrm{I}}^\star \in C^0\big([0, \bar{\tau}], H^l\big) \cap C^1\big([0, \bar{\tau}], H^{l-2}\big)$$

$$\mathsf{w}_{\mathrm{II}} - \mathsf{w}_{\mathrm{II}}^\star \in C^0\big([0, \bar{\tau}], H^l\big) \cap C^1\big([0, \bar{\tau}], H^{l-2}\big) \cap L^2\big((0, \bar{\tau}), H^{l+1}\big)$$

Moreover, there exists $\mathsf{c}_{\mathrm{loc}}(\mathcal{O}_1, b) \geq 1$ *such that*

$$\sup_{0 \leq \tau \leq \bar{\tau}} |\mathsf{w}(\tau) - \mathsf{w}^\star|_l^2 + \int_0^{\bar{\tau}} |\mathsf{w}_{\mathrm{II}}(\tau) - \mathsf{w}_{\mathrm{II}}^\star|_{l+1}^2 d\tau \leq \mathsf{c}_{\mathrm{loc}}^2 |\mathsf{w}_0 - \mathsf{w}^\star|_l^2.$$

Existence of Strong Solutions (3)

- **Application to multicomponent diffuse interface fluids**

Theorem 2. *Let* $d \geq 1$*,* $l \geq l_0 + 2$*, and* $b > 0$*. There exists* $\bar{\tau}(\mathcal{O}_1, b) > 0$ *such that for any* w_0 *with* $\mathsf{w}_0 \in \overline{\mathcal{O}}_0$*,* $\mathsf{w}_0 - \mathsf{w}^\star \in H^l$*,* $\boldsymbol{w}_0 = \boldsymbol{\nabla}\rho_0$ *and* $|\mathsf{w}_0 - \mathsf{w}^\star|_l^2 < b^2$ *there exists a unique local solution* w *with* $\mathsf{w}(0, \boldsymbol{x}) = \mathsf{w}_0(\boldsymbol{x})$*,* $\mathsf{w}(t, \boldsymbol{x}) \in \mathcal{O}_1$*,* $\boldsymbol{w} = \boldsymbol{\nabla}\rho$*, and*

$$\rho - \rho^\star \in C^0\big([0,\bar{\tau}], H^{l+1}\big), \qquad \rho_i - \rho_i^\star \in C^0\big([0,\bar{\tau}], H^{l}\big),$$

$$\boldsymbol{v} - \boldsymbol{v}^\star, T - T^\star \in C^0\big([0,\bar{\tau}], H^{l}\big) \cap L^2\big((0,\bar{\tau}), H^{l+1}\big)$$

$$g_2 - g_1, \dots, g_{\mathsf{n}_s} - g_1 \in C^0\big([0,\bar{\tau}], H^{l}\big) \cap L^2\big((0,\bar{\tau}), H^{l+1}\big).$$

Moreover, there exists $\mathsf{c}_{\text{loc}}(\mathcal{O}_1, b) \geq 1$ *such that*

$$\sup_{0\leq\tau\leq\bar{\tau}} \Big(\big|\rho(\tau) - \rho^\star\big|_{l+1}^2 + \big|\rho_i(\tau) - \rho_i^\star, \boldsymbol{v}(\tau) - \boldsymbol{v}^\star, T(\tau) - T^\star\big|_l^2 \Big) + \int_0^{\bar{\tau}} |\mathsf{w}_{\text{II}}(\tau) - \mathsf{w}_{\text{II}}^\star|_{l+1}^2 d\tau$$

$$\leq \mathsf{c}_{\text{loc}}^2 \Big(\big|\rho_0(\tau) - \rho^\star\big|_{l+1}^2 + \big|\rho_{i0} - \rho_i^\star, \boldsymbol{v}_0(\tau) - \boldsymbol{v}^\star, T_0(\tau) - T^\star\big|_l^2 \Big)$$

3 Construction of the Thermodynamics

Thermodynamics from equations of state (1)

- **Specific variables**

$$\nu = 1/\rho \qquad \mathsf{y}_i = \rho_i/\rho \quad i \in \mathfrak{S} \qquad \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_\mathrm{s}} \text{ independent}$$

$$e = \mathcal{E}/\rho \qquad s = \mathcal{S}/\rho \qquad g = \mathcal{G}/\rho, \qquad f = \mathcal{F}/\rho$$

- **Thermodynamic variables**

$$\zeta = (\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_\mathrm{s}}, T)^t \qquad \xi = (\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_\mathrm{s}}, e)^t$$

- **Expanded variables for $\boldsymbol{\lambda > 0}$**

$$\zeta_\lambda = (\lambda\nu, \lambda\mathsf{y}_1, \dots, \lambda\mathsf{y}_{\mathsf{n}_\mathrm{s}}, T)^t \qquad \xi_\lambda = \lambda\xi = (\lambda\nu, \lambda\mathsf{y}_1, \dots, \lambda\mathsf{y}_{\mathsf{n}_\mathrm{s}}, \lambda e)^t$$

Thermodynamics from equations of state (2)

- **Thermodynamics : e, p, s functions of $\zeta = (\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_s}, T)^t$ with**

  (T_0) e, p, s are $C^\gamma(\mathcal{O}_\zeta)$, $\mathcal{O}_\zeta \subset (0,\infty)^{2+\mathsf{n}_s}$ open, nonempty and connected, $\forall\lambda > 0,\ \forall\zeta \in \mathcal{O}_\zeta \quad \zeta_\lambda \in \mathcal{O}_\zeta, \quad e(\zeta_\lambda) = \lambda e(\zeta) \quad p(\zeta_\lambda) = p(\zeta) \quad s(\zeta_\lambda) = \lambda s(\zeta)$.

  (T_1) For any $\zeta \in \mathcal{O}_\zeta$ letting $g_k = \partial_{\mathsf{y}_k} e - T\partial_{\mathsf{y}_k} s$ we have Gibbs' relation

$$T\,ds = de + p\,d\nu - \sum_{k\in\mathfrak{S}} g_k\,d\mathsf{y}_k.$$

  (T_2) For any $(\mathsf{y}_1 \dots, \mathsf{y}_{\mathsf{n}_s}, T)^t \in (0,\infty)^{1+\mathsf{n}_s}$ there exists ν_{m} such that $\nu > \nu_{\mathrm{m}}$ implies $(\nu, \mathsf{y}_1 \dots, \mathsf{y}_{\mathsf{n}_s}, T)^t \in \mathcal{O}_\zeta$ and

$$\lim_{\nu\to\infty}(e - e^{\mathrm{id}}) = 0, \qquad \lim_{\nu\to\infty}\nu(p - p^{\mathrm{id}}) = 0, \qquad \lim_{\nu\to\infty}(s - s^{\mathrm{id}}) = 0.$$

  (T_3) $\mathcal{O}_\zeta$ is increasing with temperature and $\partial_T e(\zeta) > 0$.
  $\big($If $(\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_s}, T)^t \in \mathcal{O}_\zeta$ then $(\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_s}, T')^t \in \mathcal{O}_\zeta$ for any $T < T'$ $\big)$

Thermodynamics from equations of state (3)

- **Ideal mixtures in terms of the variable $\zeta = (\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n_s}}, T)^t$**

$$p^{\mathrm{id}} = \frac{RT}{\nu} \sum_{k\in\mathfrak{S}} \frac{\mathsf{y}_k}{m_k} \qquad \mathcal{O}_\zeta^{\mathrm{id}} = (0,\infty)^{\mathsf{n_s}+2}$$

$$e^{\mathrm{id}} = \sum_{k\in\mathfrak{S}} \mathsf{y}_k e_k^{\mathrm{id}} \qquad e_k^{\mathrm{id}} = e_k^{\mathrm{st}} + \int_{T^{\mathrm{st}}}^{T} c_{\mathrm{v}k}^{\mathrm{id}}(\theta)\, d\theta$$

$$s^{\mathrm{id}} = \sum_{k\in\mathfrak{S}} \mathsf{y}_k s_k^{\mathrm{id}} \qquad s_k^{\mathrm{id}} = s_k^{\mathrm{st}} + \int_{T^{\mathrm{st}}}^{T} \frac{c_{\mathrm{v}k}^{\mathrm{id}}(\theta)}{\theta}\, d\theta - \frac{R}{m_k} \log \frac{\mathsf{y}_k}{\nu m_k \gamma^{\mathrm{st}}}$$

- **First consequences**

$\zeta \mapsto \xi$ is a C^γ diffeomorphism from $\mathcal{O}_\zeta$ onto $\mathcal{O}_\xi$ open nonempty connected

The Gibbs function is given by $g = \sum_{i\in\mathfrak{S}} \mathsf{y}_i g_i$

Thermodynamics from equations of state (4)

- **Matrix $\boldsymbol{\Lambda = (\Lambda_{kl})_{k,l\in\mathfrak{S}}}$**

$$\Lambda_{kl} = \frac{\partial_{\mathsf{y}_k} g_l}{T} = \frac{\partial_{\mathsf{y}_l} g_k}{T}, \qquad k,l \in \mathfrak{S}, \qquad \widehat{\Lambda} = \Lambda - \frac{\Lambda \mathsf{y} \otimes \Lambda \mathsf{y}}{\langle \Lambda \mathsf{y}, \mathsf{y} \rangle}.$$

$$\Lambda \mathsf{y} = \frac{\nu}{T}(\partial_{\mathsf{y}_1} p, \ldots, \partial_{\mathsf{y}_{n_s}} p)^t, \qquad \langle \Lambda \mathsf{y}, \mathsf{y} \rangle = -\frac{\nu^2}{T}\partial_\nu p.$$

- **Thermodynamic stability**

(i) $\partial^2_{\xi\xi} s$ is negative semi-definite with null space $N(\partial^2_{\xi\xi} s) = \mathbb{R}\xi$.

(ii) $\partial_T e > 0$ and Λ is positive definite.

(iii) $\partial_T e > 0$, $\partial_\nu p < 0$, and $\widehat{\Lambda}$ is positive semi-definite with null space $\mathbb{R}\mathsf{y}$.

Thermodynamics from equations of state (5)

- **From** $(\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n_s}}, T)^t$ **to** $(\rho_1, \dots, \rho_{\mathsf{n_s}}, T)^t$

$$\mathcal{A}(\rho_1, \dots, \rho_{\mathsf{n_s}}, T) = \Big(\sum_{i \in \mathfrak{S}} \rho_i\Big)\, a\Big(\frac{1}{\sum_{i\in\mathfrak{S}} \rho_i}, \frac{\rho_1}{\sum_{i\in\mathfrak{S}} \rho_i}, \dots, \frac{\rho_{\mathsf{n_s}}}{\sum_{i\in\mathfrak{S}} \rho_i}, T\Big)$$

$$\mathcal{O}_{\mathcal{Z}} = \Big\{\, (\rho_1, \dots, \rho_{\mathsf{n_s}}, T) \in (0,\infty)^{1+\mathsf{n_s}};\ \Big(\frac{1}{\sum_{i\in\mathfrak{S}} \rho_i}, \frac{\rho_1}{\sum_{i\in\mathfrak{S}} \rho_i}, \dots, \frac{\rho_{\mathsf{n_s}}}{\sum_{i\in\mathfrak{S}} \rho_i}, T\Big) \in \mathcal{O}_\zeta \,\Big\}$$

- **From** $(\rho_1, \dots, \rho_{\mathsf{n_s}}, T)^t$ **to** $(\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n_s}}, T)^t$

$$a(\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n_s}}, T) = \nu \mathcal{A}\Big(\frac{\mathsf{y}_1}{\nu}, \dots, \frac{\mathsf{y}_{\mathsf{n_s}}}{\nu}, T\Big)$$

$$\mathcal{O}_\zeta = \Big\{\, (\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n_s}}, T) \in (0,\infty)^{2+\mathsf{n_s}};\ \Big(\frac{\mathsf{y}_1}{\nu}, \dots, \frac{\mathsf{y}_{\mathsf{n_s}}}{\nu}, T\Big) \in \mathcal{O}_{\mathcal{Z}} \,\Big\}$$

- **Equivalence**

$$(\mathcal{T}_0) - (\mathcal{T}_3) \qquad \Longleftrightarrow \qquad (\mathsf{T}_0) - (\mathsf{T}_3)$$

Thermodynamics from equations of state (6)

- **Construction from an equation of state**

$$p = p^{\mathrm{id}} + \phi \qquad \phi \in C^{\varkappa+1}(\mathcal{O}_\zeta) \qquad \forall \zeta \in \mathcal{O}_\zeta \quad \forall \lambda > 0 \quad \zeta_\lambda \in \mathcal{O}_\zeta$$

$\mathcal{O}_\zeta$ increasing with temperature and volume : If $\zeta = (\nu, \mathsf{y}_1, \ldots, \mathsf{y}_{\mathsf{n_s}}, T)^t \in \mathcal{O}_\zeta$ then $(\nu', \mathsf{y}_1, \ldots, \mathsf{y}_{\mathsf{n_s}}, T')^t \in \mathcal{O}_\zeta$ for $\nu' > \nu$ and $T' > T$

- **Assumption of the state law**

p is 0-homogeneous $\quad \forall \lambda > 0 \quad p(\zeta) = p(\zeta_\lambda)$

For any $\beta = (\beta_T, \beta_\nu, \beta_1, \ldots, \beta_{\mathsf{n_s}}) \in \mathbb{N}^{2+\mathsf{n_s}}$

$$|\partial_\zeta^\beta \phi| = |\widetilde{\partial}_T^{\beta_T} \widetilde{\partial}_\nu^{\beta_\nu} \widetilde{\partial}_{\mathsf{y}_1}^{\beta_1} \cdots \widetilde{\partial}_{\mathsf{y}_{\mathsf{n_s}}}^{\beta_{\mathsf{n_s}}} \phi| \leqslant \frac{\mathsf{c}(\beta, T)}{\nu^{\beta_\nu + 2}}$$

$$T \int_\nu^\infty \partial_T^2 \phi \, d\nu' \quad < \quad \sum_{k \in \mathfrak{S}} \mathsf{y}_k c_{\mathrm{v}k}^{\mathrm{id}}$$

Thermodynamics from equations of state (7)

- **Necessary and sufficient conditions**

$$e = e^{\mathrm{id}} - \int_\nu^\infty T^2 \partial_T \left(\frac{\phi}{T}\right) d\nu' \qquad s = s^{\mathrm{id}} - \int_\nu^\infty \partial_T \phi \, d\nu'$$

$$g_k = g_k^{\mathrm{id}} - \int_\nu^\infty \partial_{\mathsf{y}_k} \phi \, d\nu', \qquad k \in \mathfrak{S}$$

Theorem 3. *e, s, p is a thermodynamics in the sense of* $(\mathsf{T}_0) - (\mathsf{T}_3)$

- **Thermodynamic relations**

$$\partial_\nu e = T^2 \partial_T \left(\frac{p}{T}\right) = T^2 \partial_T \left(\frac{\phi}{T}\right) \qquad g_k = \partial_{\mathsf{y}_k} e - T \partial_{\mathsf{y}_k} s$$

$$\partial_\nu (Ts - e) = p \qquad \partial_\nu (Ts - e) - \partial_\nu (Ts^{\mathrm{id}} - e^{\mathrm{id}}) = \phi$$

The Soave-Redlich-Kwong equation of state (1)

- **Soave-Redlich-Kwong equation of state (SRK)**

$$p = \sum_{i \in \mathfrak{S}} \frac{\rho_i}{m_i} \frac{RT}{1 - \rho b} - \frac{\rho^2 a}{1 + \rho b}$$

$$a = \sum_{i,j \in \mathfrak{S}} \mathsf{y}_i \mathsf{y}_j \alpha_i \alpha_j \qquad b = \sum_{i \in \mathfrak{S}} \mathsf{y}_i b_i$$

- **Coefficients $\alpha_i(T)$ and β_i**

$$\alpha_i \geqslant 0 \qquad \alpha_i(0) > 0 \qquad \lim_{+\infty} \alpha_i = 0$$

$$\partial_T \alpha_i \leqslant 0 \qquad \partial^2_{TT} \alpha_i \geqslant 0 \qquad \alpha_i \ \ \text{smooth}$$

$$b_i = \mathrm{Cte} \qquad b_i > 0.$$

The Soave-Redlich-Kwong equation of state (2)

- **Coefficients $\alpha_i = \sqrt{a_i}$ and β_i for SRK**

Stable species $a_i\big(T_{c,i}\big) = 0.42748\frac{R^2T_{c,i}^2}{m_i^2 p_{c,i}}$ $\quad b_i = 0.08664\frac{RT_{c,i}}{m_i p_{c,i}}$

Unstable species $a_i\big(T_{c,i}\big) = \big(5.55 \pm 0.12\big)\frac{\mathrm{N}^2\epsilon_i\sigma_i^3}{m_i^2}$ $\quad b_i = \big(0.855 \pm 0.018\big)\frac{\mathrm{N}\sigma_i^3}{m_i}$

Pseudo critical temperature $T_{c,i} = \frac{\epsilon_i}{k_\mathrm{B}}(1.316 \pm 0.006)$

$$\alpha_i(T) = \alpha_i(T_{c,i})\widetilde{\alpha}_i(T_i^*) \qquad T_i^* = T/T_{c,i}$$

$$\widetilde{\alpha}_i = 1 + \mathcal{A}\big(\mathrm{s}_i(1-\sqrt{T_i^*})\big) \qquad \begin{cases} \mathcal{A}(x) = x, & x \geq 0 \\ \mathcal{A}(x) = \tanh(x), & x \leq 0 \end{cases}$$

$\mathrm{s}_i = 0.48508 + 1.5517\varpi_i - 0.151613\varpi_i^2$ $\qquad$ Acentric factor ϖ_i

The Soave-Redlich-Kwong equation of state (3)

- **Specific energy and entropy for SRK EOS**

$$e = \sum_{i\in\mathfrak{S}} \mathsf{y}_i e_i^{\mathrm{id}} + \big(T\partial_T a - a\big)\frac{\ln\big(1+\rho b\big)}{b}$$

$$s = \sum_{i\in\mathfrak{S}} \mathsf{y}_i s_i^{\mathrm{id}\star} - \sum_{i\in\mathfrak{S}} \frac{\mathsf{y}_i R}{m_i}\ln\Big(\frac{\rho_i RT}{m_i(1-\rho b)p^{\mathrm{st}}}\Big) + \partial_T a\frac{\ln\big(1+\rho b\big)}{b}$$

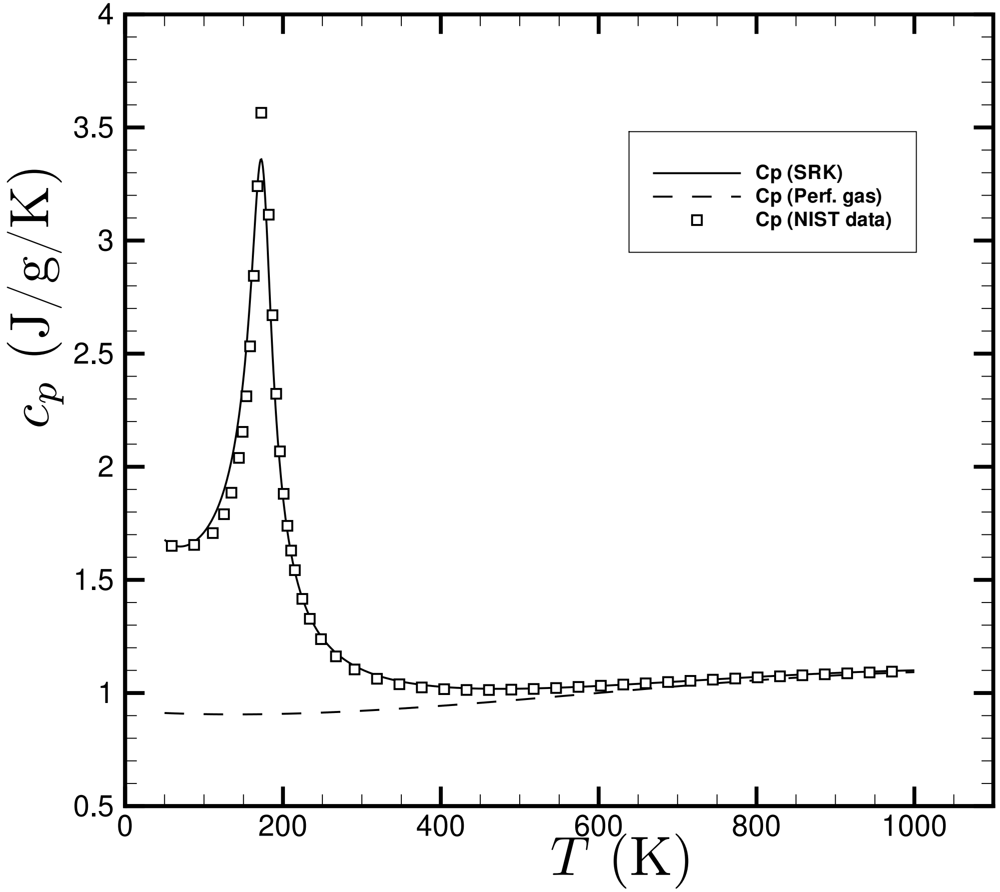
- **Matrix Λ**

$$\Lambda_{ij} = \frac{R\delta_{ij}}{m_i\mathsf{y}_i} + \frac{R}{\nu-b}\big(\frac{b_i}{m_j}+\frac{b_j}{m_i}\big) + \sum_{k\in\mathfrak{S}}\frac{\mathsf{y}_k}{m_k}\frac{R}{(\nu-b)^2}b_ib_j - \frac{2}{T}\frac{\alpha_i\alpha_j}{b}\log\big(1+\frac{b}{\nu}\big)$$
$$+\frac{2}{T}\sum_{k\in\mathfrak{S}}\mathsf{y}_k\big(\alpha_i\alpha_k b_j + \alpha_j\alpha_k b_i\big)\Big(\frac{1}{b^2}\log\big(1+\frac{b}{\nu}\big) - \frac{1}{b(\nu+b)}\Big)$$
$$+\frac{1}{T}ab_ib_j\Big(-\frac{2}{b^3}\log\big(1+\frac{b}{\nu}\big) + \frac{2}{b^2(\nu+b)} + \frac{1}{b(\nu+b)^2}\Big), \qquad i,j\in\mathfrak{S}.$$

4 Numerical experiments

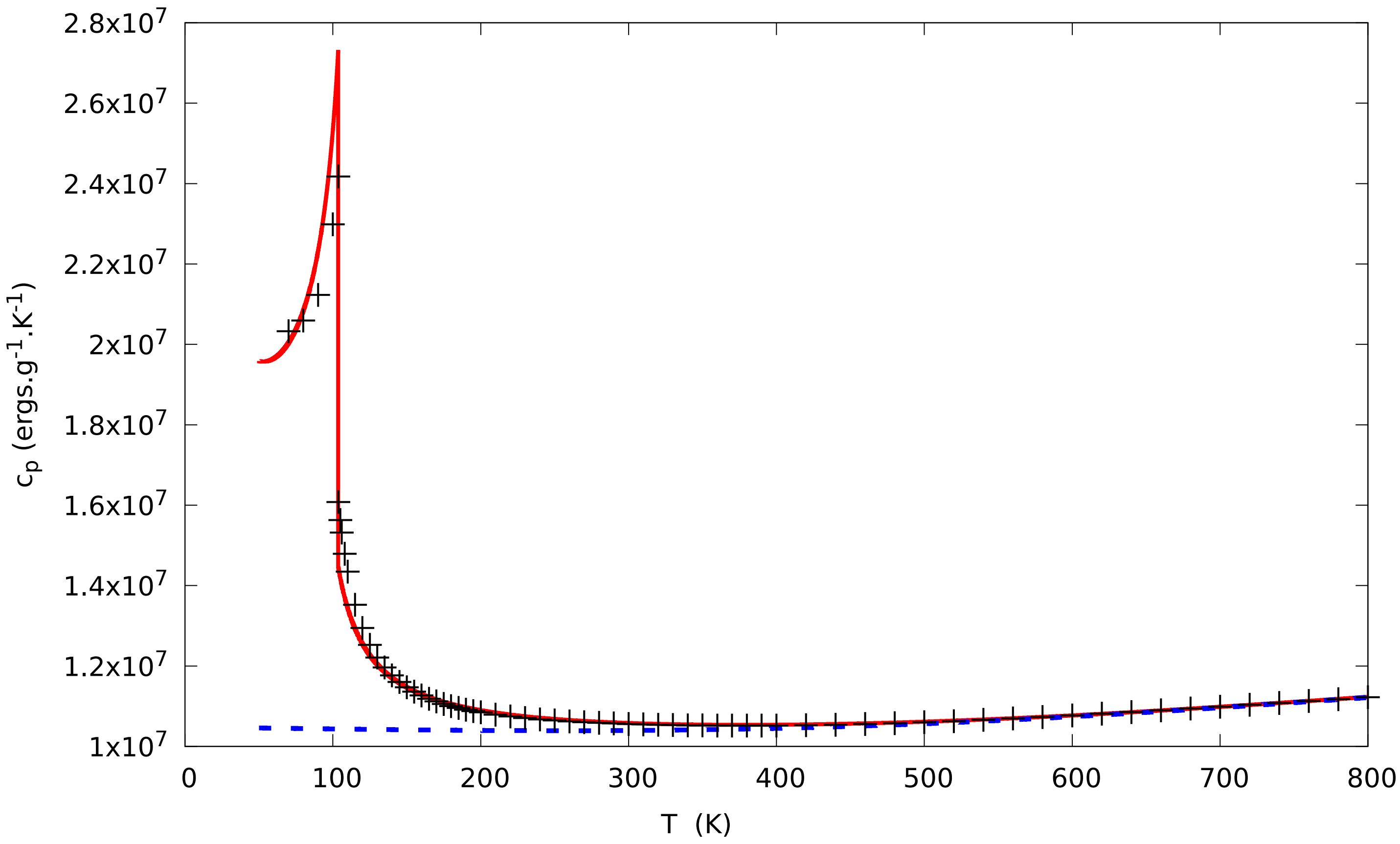
Numerical experiments with thermodynamics (1)

- **Specific heat at constant pressure c_p of O_2 at 100 atm**



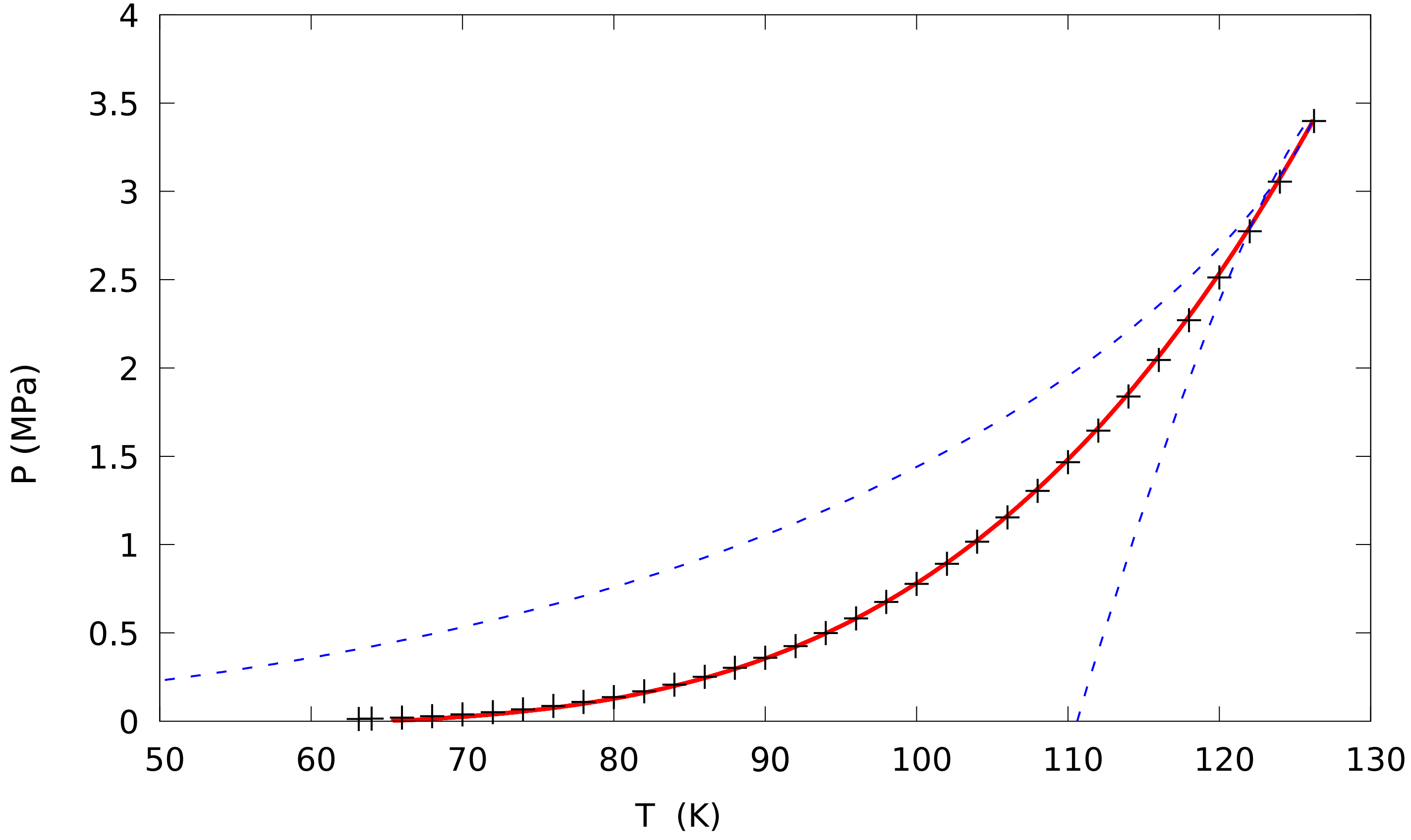
Numerical experiments with thermodynamics (2)

- **Specific heat at constant pressure c_p of N_2 at 100 atm**



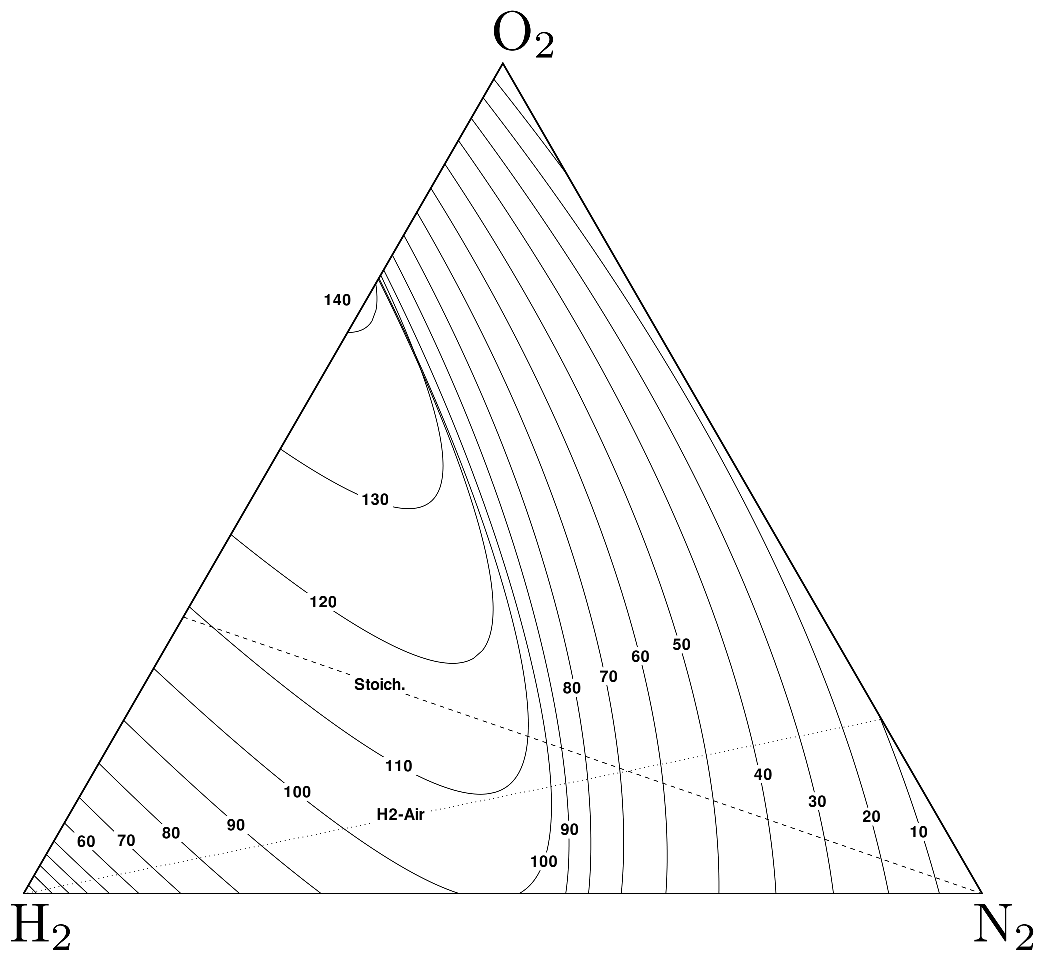
Numerical experiments with thermodynamics (3)

- **Saturation pressure of N_2**



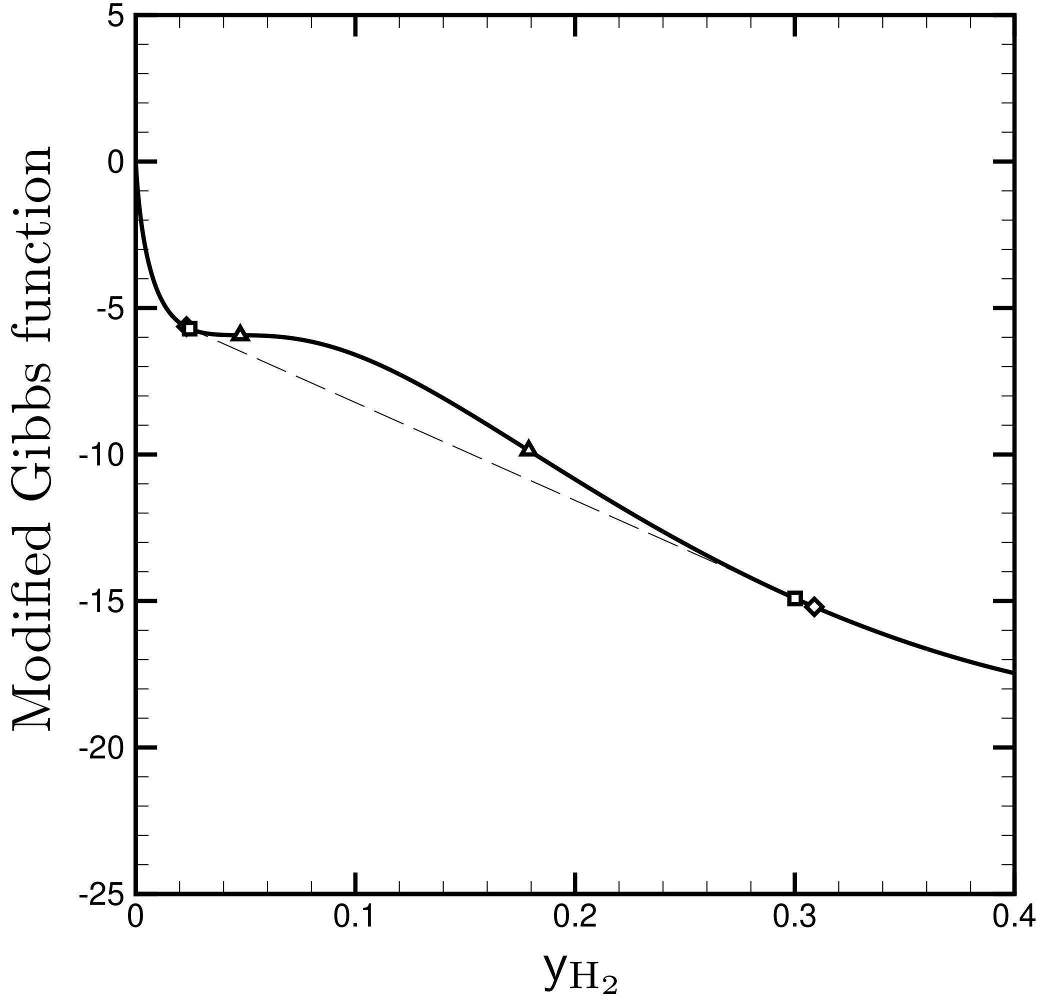
Numerical experiments with thermodynamics (4)

- **Stability of $H_2/O_2/N_2$ mixtures at $p = 100$ atm**



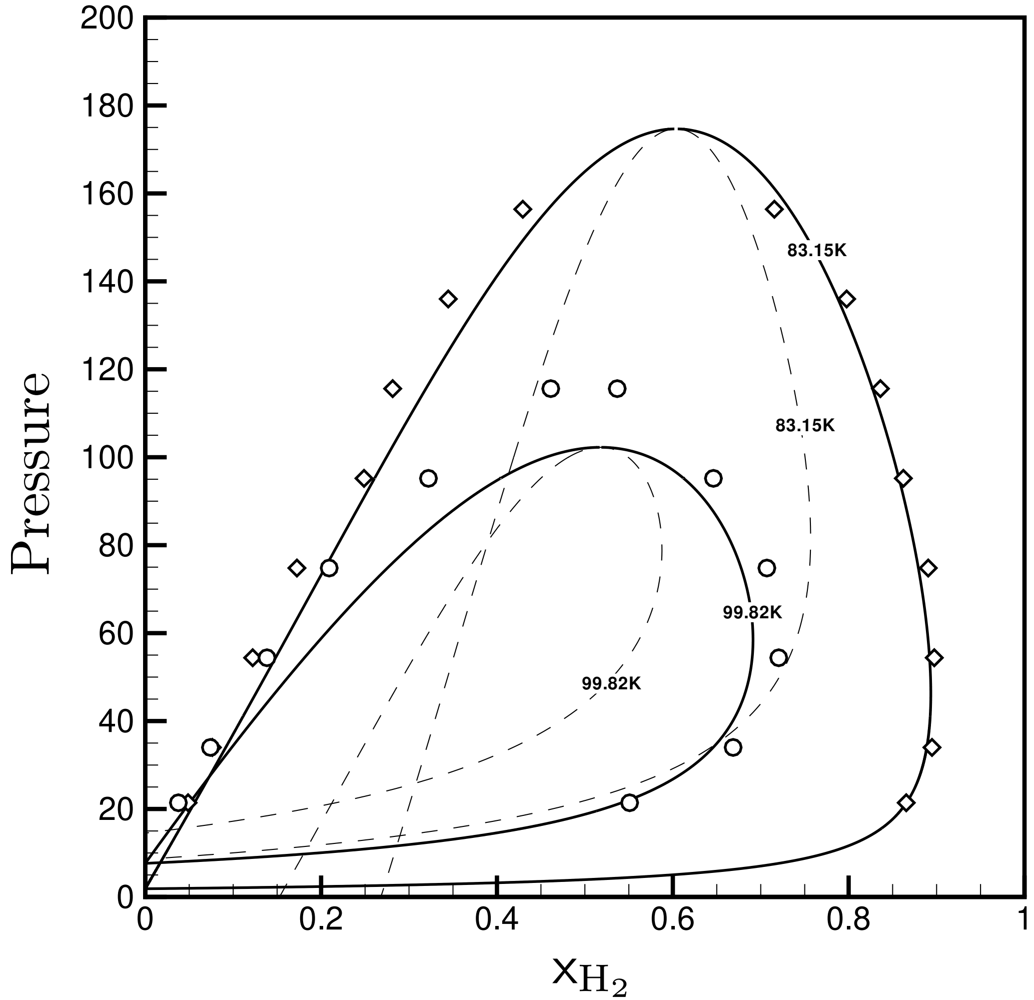
Numerical experiments with thermodynamics (5)

- **Stability ofs H_2/N_2 mixtures at $T = 83.15$ K and $p = 95.2$ atm**



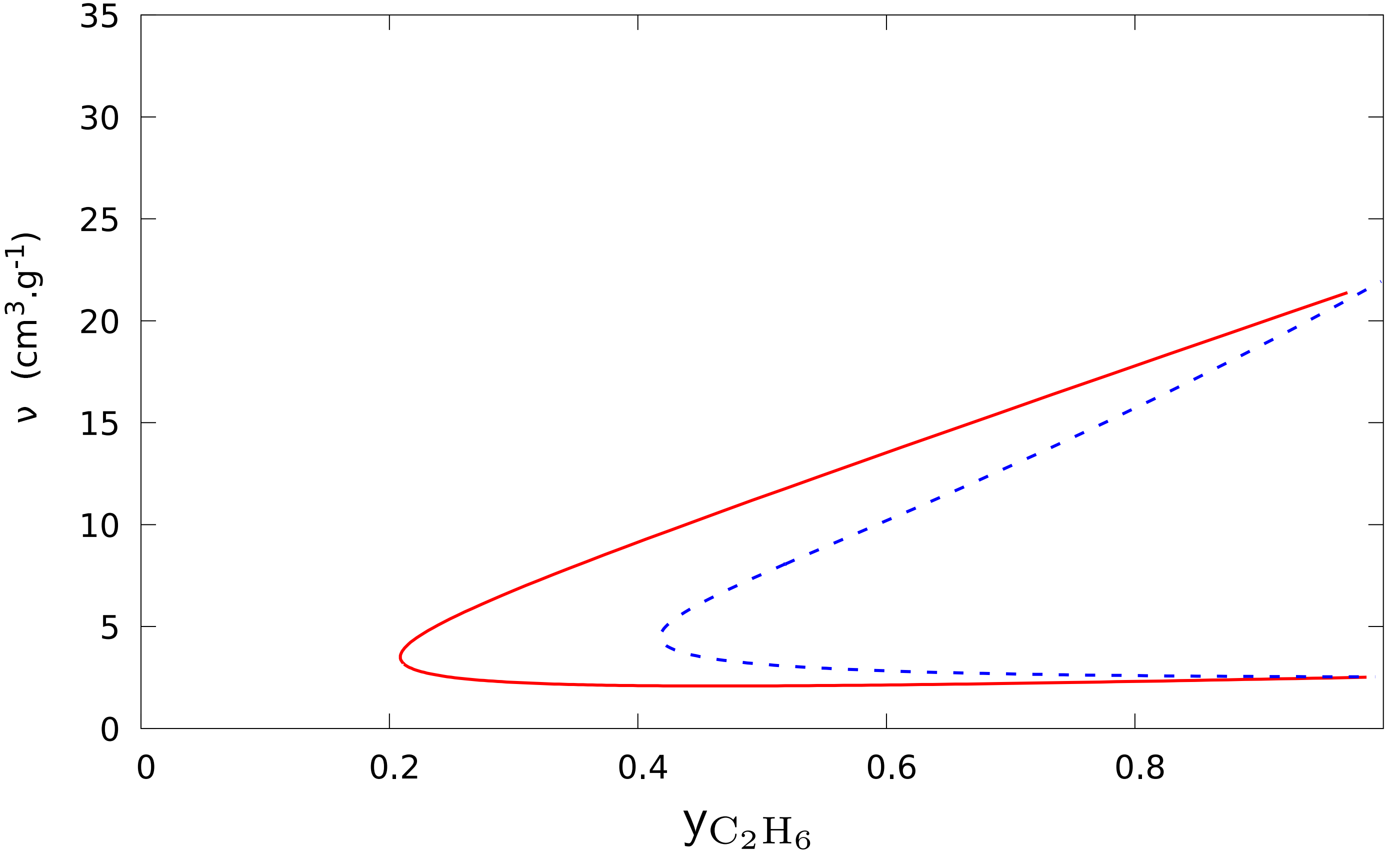
Numerical experiments with thermodynamics (6)

- **Stability of H_2/N_2 mixtures, Comparaison with Eubank experiment**



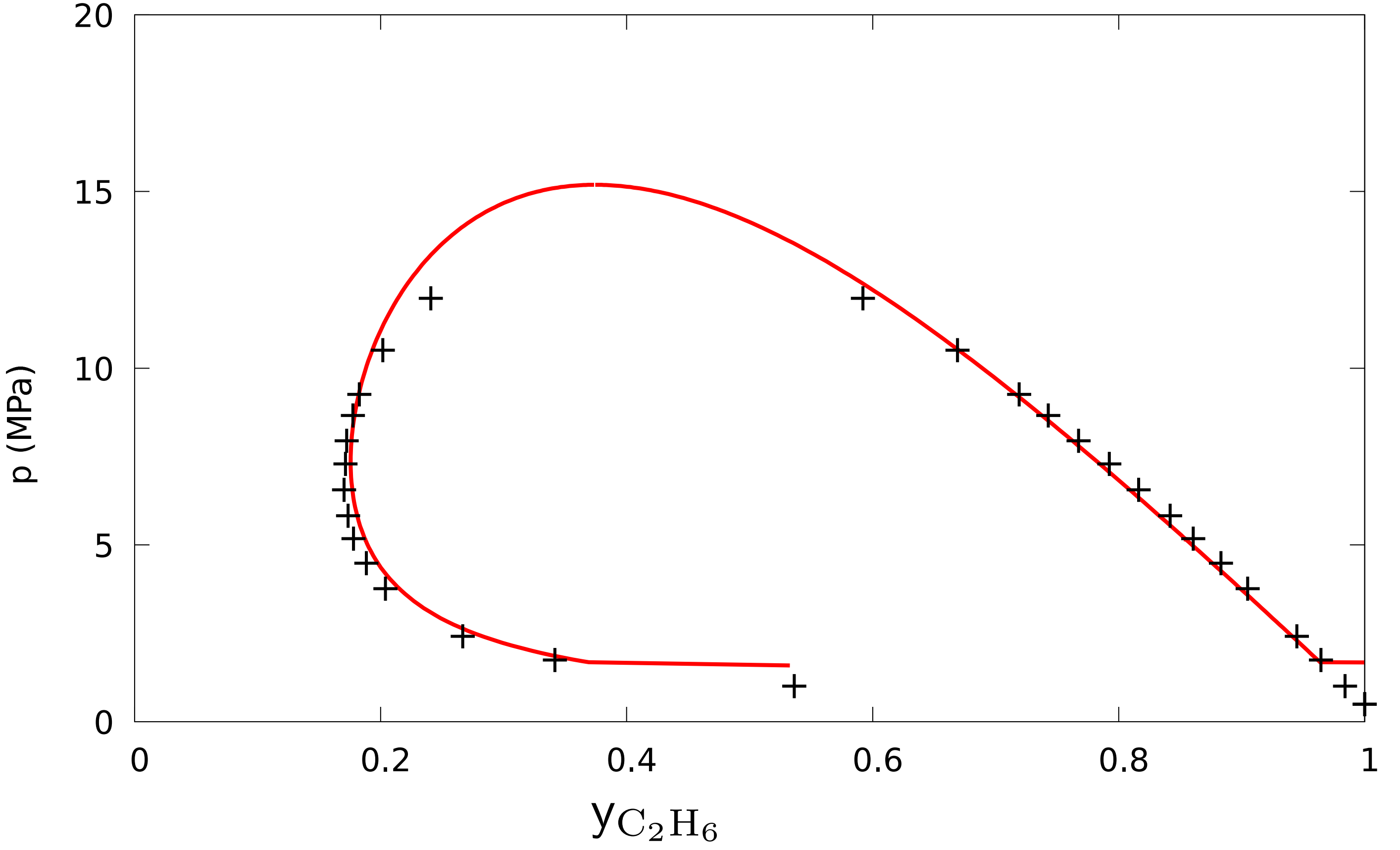
Numerical experiments with thermodynamics (7)

- **Stability domain for N_2 and C_2H_6 at $T = 220$ K**



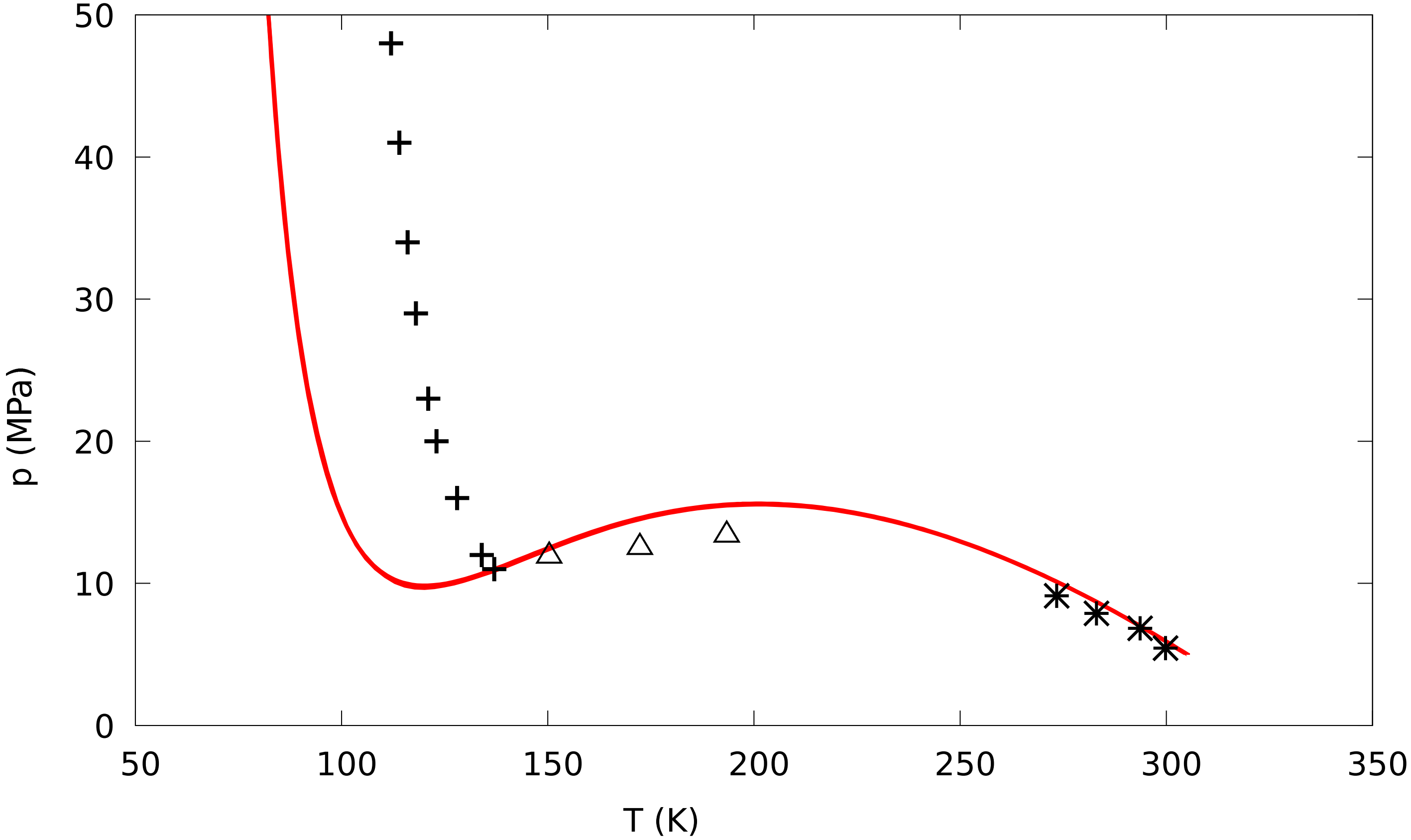
Numerical experiments with thermodynamics (8)

- **Equilibrium between N_2 and C_2H_6 at $T = 220$ K**



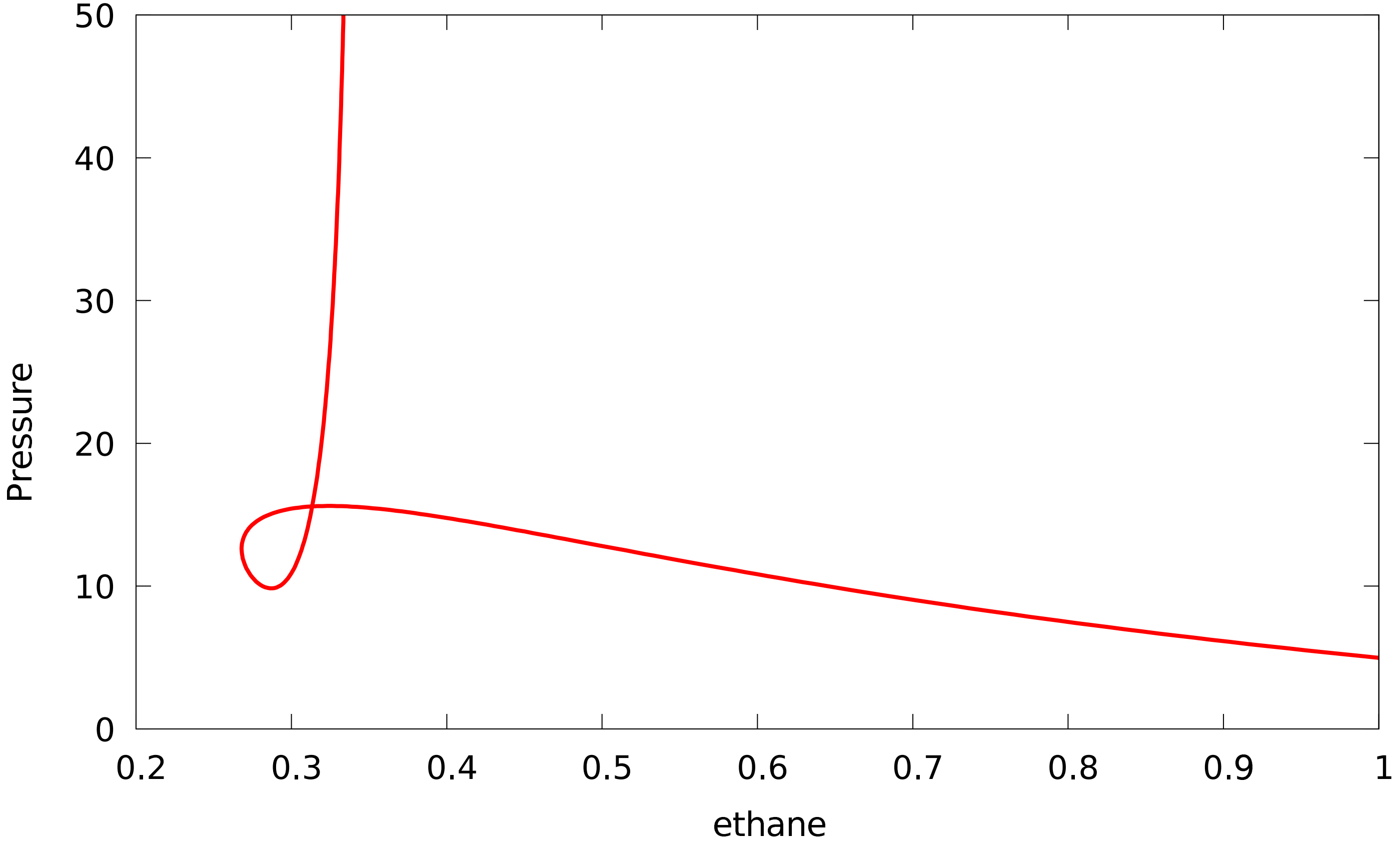
Numerical experiments with thermodynamics (9)

- **Critical points of N_2 and C_2H_6**



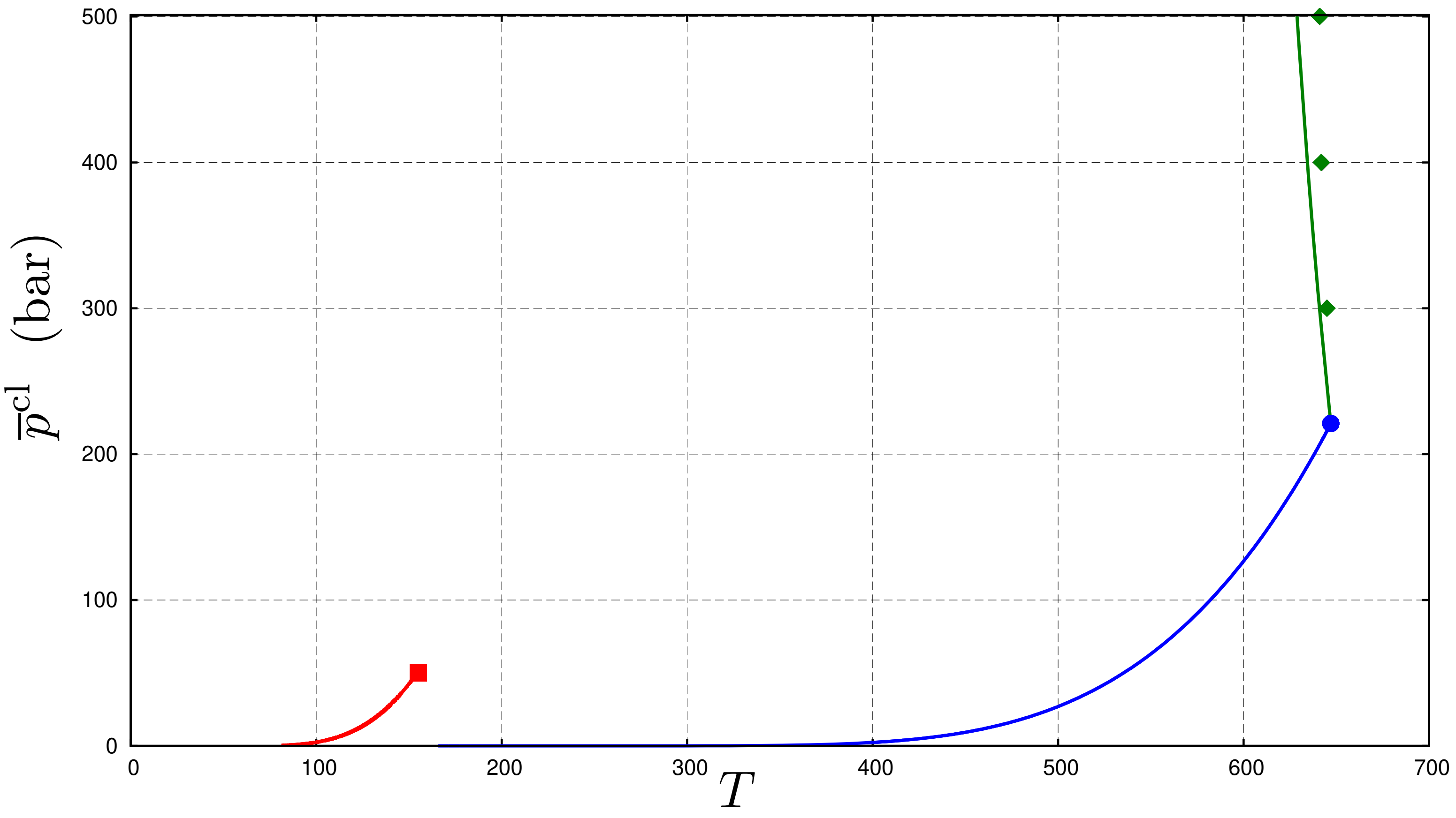
Numerical experiments with thermodynamics (10)

- **Critical points of N_2 and C_2H_6**



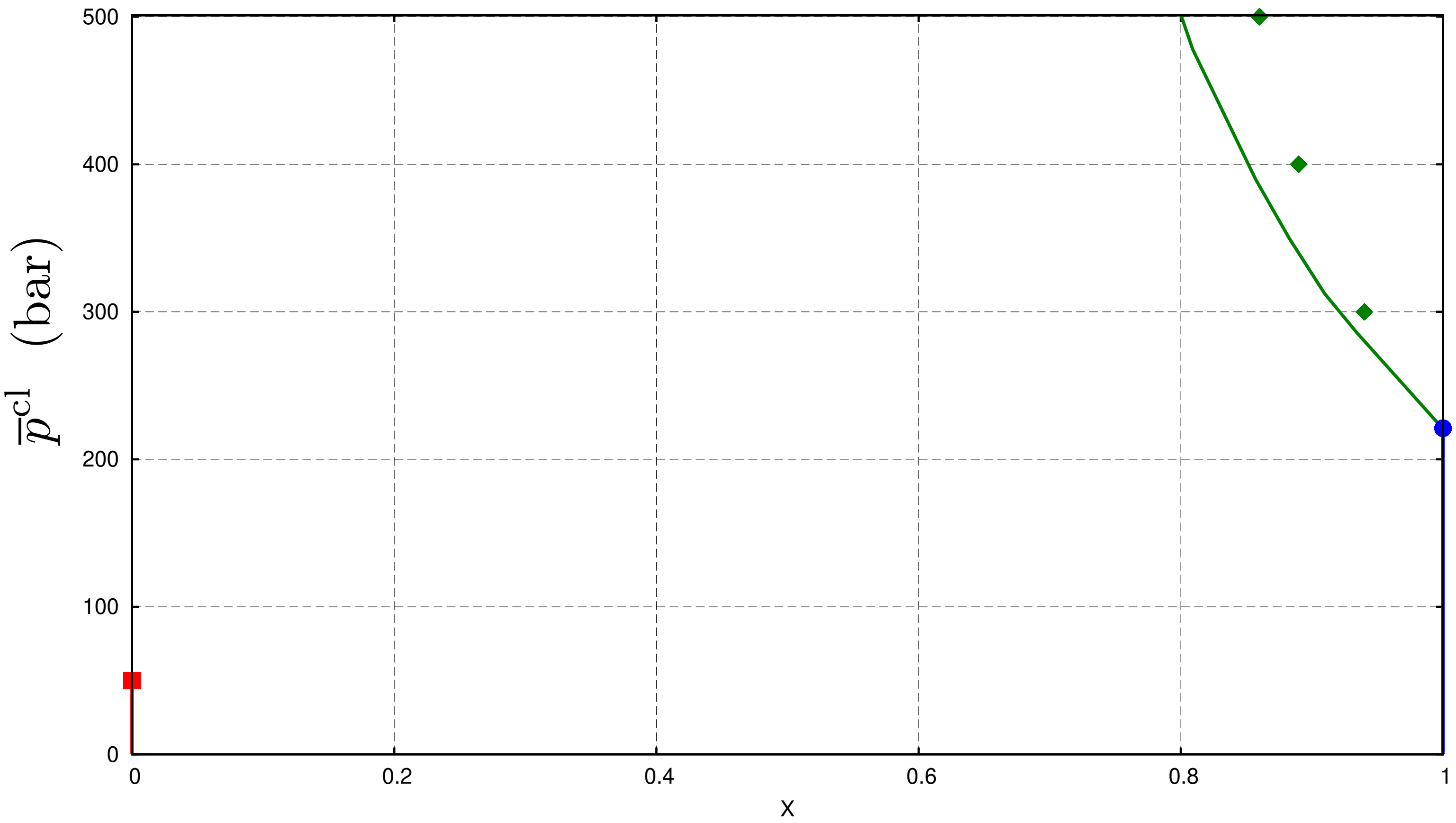
Numerical experiments with thermodynamics (11)

- **Type III phase diagram for O_2/H_2O**



Numerical experiments with thermodynamics (12)

- **Type III phase diagram for O_2/H_2O**



6 Strained Diffuse Interface Fluids

Strained Diffuse Interface Fluids (1)

- **Small Mach expansion of $\boldsymbol{p}^{\mathrm{cl}}$**

$$p^{\mathrm{cl}} = \overline{p}^{\mathrm{cl}} + \epsilon^2 \widetilde{p}^{\mathrm{cl}}$$

$$\boldsymbol{\nabla}\boldsymbol{\cdot}\overline{\boldsymbol{\mathcal{P}}} = \boldsymbol{\nabla}\boldsymbol{\cdot}\Big(\overline{p}^{\mathrm{cl}}\boldsymbol{I} - \tfrac{1}{2}\kappa|\boldsymbol{\nabla}\rho|^2 + \kappa\boldsymbol{\nabla}\rho\otimes\boldsymbol{\nabla}\rho - \kappa\rho\Delta\rho\boldsymbol{I}\Big) = 0$$

- **Flat interface**

$$T = T(t,\zeta) \qquad \overline{p}^{\mathrm{cl}} = \overline{p}^{\mathrm{cl}}(t,\zeta) \qquad \rho = \rho(t,\zeta)$$

$$\rho' = \partial_\zeta \rho \qquad \rho'' = \partial_\zeta^2 \rho \qquad \overline{p}^{\mathrm{cl}} + \kappa\tfrac{1}{2}\rho'^{\,2} - \kappa\rho\rho'' = p^\infty$$

- **Surface tension tangential forces**

$$\overline{\boldsymbol{\mathcal{P}}} = \big(p^\infty - \kappa\rho'^{\,2}\big)(\boldsymbol{I} - \mathbf{e}_\zeta\otimes\mathbf{e}_\zeta) + p^\infty\mathbf{e}_\zeta\otimes\mathbf{e}_\zeta$$

Strained Diffuse Interface Fluids (2)

- **Small Mach expansions**

  Bulk thermodynamic quantities evaluated at $\overline{p}^{\mathrm{cl}}$

  Other quantities than pressure denoted as their zeroth order expansion

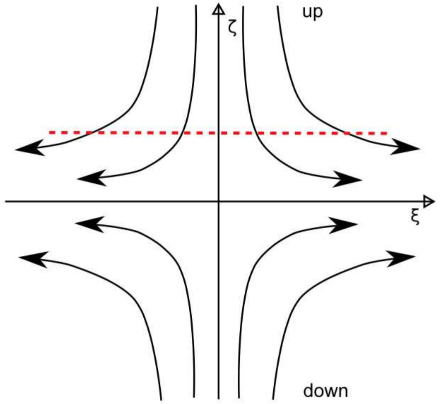
  Only the pressure $\widetilde{p}^{\mathrm{cl}}$ in the second order tangential momentum equation

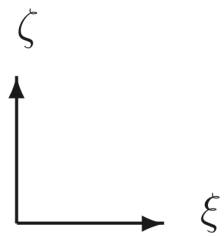
- **Small Mach energy equation**

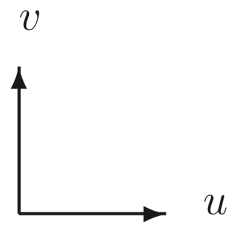
$$\rho \partial_t h^{\mathrm{cl}} + \rho \boldsymbol{v} \boldsymbol{\nabla} \cdot h^{\mathrm{cl}} + \boldsymbol{\nabla} \cdot \boldsymbol{q} = \partial_t \overline{p}^{\mathrm{cl}} + \boldsymbol{v} \cdot \boldsymbol{\nabla} \overline{p}^{\mathrm{cl}},$$

Strained Diffuse Interface Fluids (3)

- **Schematic of the flow**







- **Self similar structure**

$$T = T(t,\zeta) \qquad \rho = \rho(t,\zeta) \qquad \overline{p}^{\mathrm{cl}} = \overline{p}^{\mathrm{cl}}(t,\zeta)$$

$$u = \xi\, \widetilde{u}(t,\zeta) \qquad v = v(t,\zeta) \qquad \mathsf{y}_i = \mathsf{y}_i(t,\zeta)$$

$$\widetilde{p}^{\mathrm{cl}} = -\tfrac{1}{2} J \xi^2 + \widehat{p}(t,\zeta) \qquad \mathcal{J}_i = \mathcal{J}_i(t,\zeta) \qquad q = q(t,\zeta)$$

Strained Diffuse Interface Fluids (4)

- **Diffuse strained flame equations**

$$\partial_t \rho + \rho \widetilde{u} + \partial_\zeta(\rho v) = 0$$

$$\rho \partial_t \mathsf{y}_i + \rho v \partial_\zeta \mathsf{y}_i + \partial_\zeta \mathcal{J}_i = m_i \omega_i$$

$$\rho \partial_t \widetilde{u} + \rho \widetilde{u}^2 + \rho v \partial_\zeta \widetilde{u} - J + \partial_\zeta(\eta \partial_\zeta \widetilde{u}) = 0$$

$$\rho \partial_t h^{\mathrm{cl}} + \rho v \partial_\zeta h^{\mathrm{cl}} - v \partial_\zeta \overline{p}^{\mathrm{cl}} + \partial_\zeta q = 0$$

- **Boundary conditions**

$$T(-\infty) = T_{\mathrm{lo}} \qquad T(+\infty) = T_{\mathrm{up}} \qquad v(0) = 0$$

$$\mathsf{y}_k(-\infty) = \mathsf{y}_{k\mathrm{lo}} \qquad \mathsf{y}_k(+\infty) = \mathsf{y}_{k\mathrm{up}} \qquad \alpha = \left(\frac{J}{\rho_{\mathrm{up}}}\right)^{\frac{1}{2}}$$

$$\widetilde{u}(-\infty) = \alpha \left(\frac{\rho_{\mathrm{up}}}{\rho_{\mathrm{lo}}}\right)^{\frac{1}{2}} \qquad \widetilde{u}(+\infty) = \alpha,$$

Strained Diffuse Interface Fluids (5)

- **Pressure**

$$\overline{p}^{\rm cl} = p^{\infty} - \kappa \tfrac{1}{2}(\partial_\zeta \rho)^2 + \kappa \rho \partial_\zeta^2 \rho$$

- **Dissipative transport fluxes**

$$\mathcal{J}_i = -\sum_{j \in \mathfrak{S}} L_{ij} \partial_\zeta \Big(\frac{g_j^{\rm cl}}{T} \Big) - L_{ie} \partial_\zeta \Big(\frac{-1}{T} \Big)$$

$$q = -\sum_{i \in \mathfrak{S}} L_{ei} \partial_\zeta \Big(\frac{g_i^{\rm cl}}{T} \Big) - L_{ee} \partial_\zeta \Big(\frac{-1}{T} \Big)$$

Dense and/or supercritical fluids

- **Kinetic theory or statistical mechanics**

  Enskog (1922), Thorne (Unpublished), Hirschfelder, Curtiss and Bird (1954), Bearman and Kirkwood (1958), Mori (1958), Chapman and Cowling (1970), Ferziger and Kaper (1972), Bajaras, Garcia-Colin and Piña (1973), Van Beijeren and Ernst (1973), Kurochkin, Makarenko and Tirskii (1984)

- **Thermodynamics of irreversible processes/statistical thermodynamics**

  Marcelin (1910), Meixner (1943), Prigogine (1947), Keizer (1987)

- **Supercritical combustion and/or coefficients**

  Ely and Hanley (1981,1983), Chung et al. (1988)
  Belan and Harstad (2004), Oefelein (2005), Palle and Miller (2007), Giovangigli, Manuszewski and Dupoirieux (2011)

Thermochemistry (1)

- **Soave-Redlich-Kwong state law (SRK)**

$$\overline{p}^{\mathrm{cl}} = \sum_{i\in\mathfrak{S}} \frac{\rho_i}{m_i}\frac{RT}{1-\rho b} - \frac{\rho^2 a}{1+\rho b}$$

$$a = \sum_{i,j\in\mathfrak{S}} \mathsf{y}_i\mathsf{y}_j\alpha_i\alpha_j \qquad b = \sum_{i\in\mathfrak{S}} \mathsf{y}_i b_i$$

- **Coefficients $\boldsymbol{\alpha_i(T)}$ and $\boldsymbol{\beta_i}$**

$$\alpha_i \geqslant 0 \qquad \alpha_i(0) > 0 \qquad \lim_{+\infty} \alpha_i = 0$$

$$\partial_T\alpha_i \leqslant 0 \qquad \partial^2_{TT}\alpha_i \geqslant 0$$

$$b_i = \mathrm{Cte} \qquad b_i > 0$$

Thermochemistry (2)

- **Coefficients $\alpha_i = \sqrt{a_i}$ for SRK**

$$a_i\big(T_{c,i}\big) = 0.42748\frac{R^2T_{c,i}^2}{m_i^2 p_{c,i}} \qquad b_i = 0.08664\frac{RT_{c,i}}{m_i p_{c,i}}$$

$$a_i\big(T_{c,i}\big) = \big(5.55 \pm 0.12\big)\frac{\mathrm{N}^2\epsilon_i\sigma_i^3}{m_i^2} \qquad b_i = \big(0.855 \pm 0.018\big)\frac{\mathrm{N}\sigma_i^3}{m_i}$$

$$\alpha_i(T) = \alpha_i(T_{c,i})\widetilde{\alpha}_i(T_i^*) \qquad T_i^* = T/T_{c,i}$$

$$\widetilde{\alpha}_i = 1 + \mathcal{A}\big(\mathrm{s}_i(1-\sqrt{T_i^*})\big) \qquad \begin{cases} \mathcal{A}(x) = x, & x \geq 0 \\ \mathcal{A}(x) = \tanh(x), & x \leq 0 \end{cases}$$

$$\mathrm{s}_i = 0.48508 + 1.5517\varpi_i - 0.151613\varpi_i^2$$

Thermochemistry (3)

- **Construction of the thermodynamics**

  There exists a unique thermodynamics whose state law is SRK and

$$e^{\mathrm{cl}} = \sum_{i\in\mathfrak{S}} \mathsf{y}_i e_i^{\mathrm{id}} + \big(T\partial_T a - a\big)\frac{\ln\big(1+\rho b\big)}{b}$$

$$s^{\mathrm{cl}} = \sum_{i\in\mathfrak{S}} \mathsf{y}_i s_i^{\mathrm{id}\star} - \sum_{i\in\mathfrak{S}} \frac{\mathsf{y}_i R}{m_i} \ln\Big(\frac{\rho_i RT}{m_i(1-\rho b)p^{\mathrm{st}}}\Big) + \partial_T a \frac{\ln\big(1+\rho b\big)}{b}$$

- **Thermodynamic stability**

$$\partial_T e^{\mathrm{cl}} > 0 \qquad \Lambda \text{ is positive definite} \qquad \Lambda_{kl} = \partial_{\mathsf{y}_k} g_l^{\mathrm{cl}}/T = \partial_{\mathsf{y}_l} g_k^{\mathrm{cl}}/T$$

Transport matrix L (1)

- **Evaluation of L**

$$L = \begin{pmatrix} \mathcal{D} & \mathcal{D}\hbar \\ (\mathcal{D}\hbar)^t & \lambda + \langle \mathcal{D}\hbar, \hbar \rangle \end{pmatrix}$$

$$\mathcal{D}_{ij} = \rho \mathsf{y}_i \mathsf{y}_j \frac{m}{R} D_{ij}, \qquad \hbar_i = h_i + RT \frac{\widetilde{\chi}_i}{m_i}$$

- **Transport coefficients**

Multicomponent diffusion coefficients D_{ij}

Thermal diffusion ratios $\chi_i = \mathsf{x}_i \widetilde{\chi}_i$

Soret coefficient $\theta_i = \sum_{j \in \mathfrak{S}} D_{ij} \chi_j$

Thermal conductivity λ

Transport matrix L (2)

- **Viscosity η and thermal conductivity λ**

  Ely and Hanley (1983) Chung et al. (1988)

- **Reduced thermal diffusion ratios $\widetilde{\chi}$**

  Thermal diffusion ratios $\widetilde{\chi}$ evaluated as for perfect gases

- **Multicomponent diffusion coefficients $D = (\Delta + \mathsf{y}\otimes\mathsf{y})^{-1} - \mathbb{1}\otimes\mathbb{1}$**

$$\Delta_{kk} = \sum_{l\neq k} \frac{\mathsf{x}_k \mathsf{x}_l}{\mathcal{D}_{kl}} \qquad \Delta_{kl} = -\frac{\mathsf{x}_k \mathsf{x}_l}{\mathcal{D}_{kl}} \quad k \neq l$$

$$\mathcal{D}_{kl} = \mathcal{D}_{kl}^{\mathrm{id}} / \Upsilon_{kl} \qquad \mathsf{y} = (\mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_\mathrm{s}})^t \qquad \mathbb{1} = (1, \dots, 1)^t$$

$$\Upsilon_{ij} = 1 + \sum_{k\in\mathfrak{S}} \frac{\pi \mathfrak{n}_k}{12} \Big(8(\sigma_{ik}^3 + \sigma_{jk}^3) - 6(\sigma_{ik}^2 + \sigma_{jk}^2)\sigma_{ij} - 3(\sigma_{ik}^2 - \sigma_{jk}^2)^2 \sigma_{ij}^{-1} + \sigma_{ij}^3 \Big)$$

Transport matrix L (3)

- **Diffusion driving forces and reduced chemical potential**

$$\widehat{d}_k = \mathsf{x}_k\ (\partial_\zeta \mu_k)_T = \frac{\mathsf{x}_k m_k \nu_k}{RT} \partial_\zeta \overline{p}^{\mathrm{cl}} + \sum_{l\in\mathfrak{S}} \Gamma_{jl} \partial_\zeta \mathsf{x}_l \qquad \mu_k = \frac{m_k g_k^{\mathrm{cl}}}{RT}$$

$$\mu_k = \mu_k(T, \overline{p}^{\mathrm{cl}}, \mathsf{x}_1, \ldots, \mathsf{x}_{\mathsf{n}_\mathrm{s}}) \qquad \Gamma_{jl} = \mathsf{x}_j (\partial_{\mathsf{x}_l} \mu_j)_{\overline{p}^{\mathrm{cl}}, \mathsf{x}} \qquad d_k = \widehat{d}_k - \mathsf{y}_k \sum_{l\in\mathfrak{S}} \widehat{d}_l$$

- **Traditional formulation**

$$\frac{\mathcal{J}_k}{\rho_k} = \mathcal{V}_k = -\sum_{l\in[1,\mathsf{n}_\mathrm{s}]} D_{kl} d_l - \theta_k \partial_\zeta \log T$$

$$q - \sum_{k\in[1,\mathsf{n}_\mathrm{s}]} h_k \rho_k \mathcal{V}_k = -\frac{\rho RT}{m} \sum_{k\in[1,\mathsf{n}_\mathrm{s}]} \theta_k d_k - \widehat{\lambda} \partial_\zeta T$$

Transport matrix L (4)

- **Mechanical thermodynamic unstable points**

  Mechanical stability limit $\partial_\rho \overline{p}^{\mathrm{cl}} = 0$

  Expression of fixed pressure quantities

$$(\partial_{\mathsf{y}_k} \phi)_{T,\overline{p}^{\mathrm{cl}},\mathsf{y}_l} = (\partial_{\mathsf{y}_k} \phi)_{T,\rho,\mathsf{y}_l} + (\partial_\rho \phi)_{T,\mathsf{y}_l} (\partial_{\mathsf{y}_k} \rho)_{T,\overline{p}^{\mathrm{cl}},\mathsf{y}_l}$$

$$(\partial_{\mathsf{y}_k} \rho)_{T,\overline{p}^{\mathrm{cl}},\mathsf{y}_l} = -\frac{(\partial_{\mathsf{y}_k} \overline{p}^{\mathrm{cl}})_{T,\rho,\mathsf{y}_l}}{(\partial_\rho \overline{p}^{\mathrm{cl}})_{T,\mathsf{y}_l}}$$

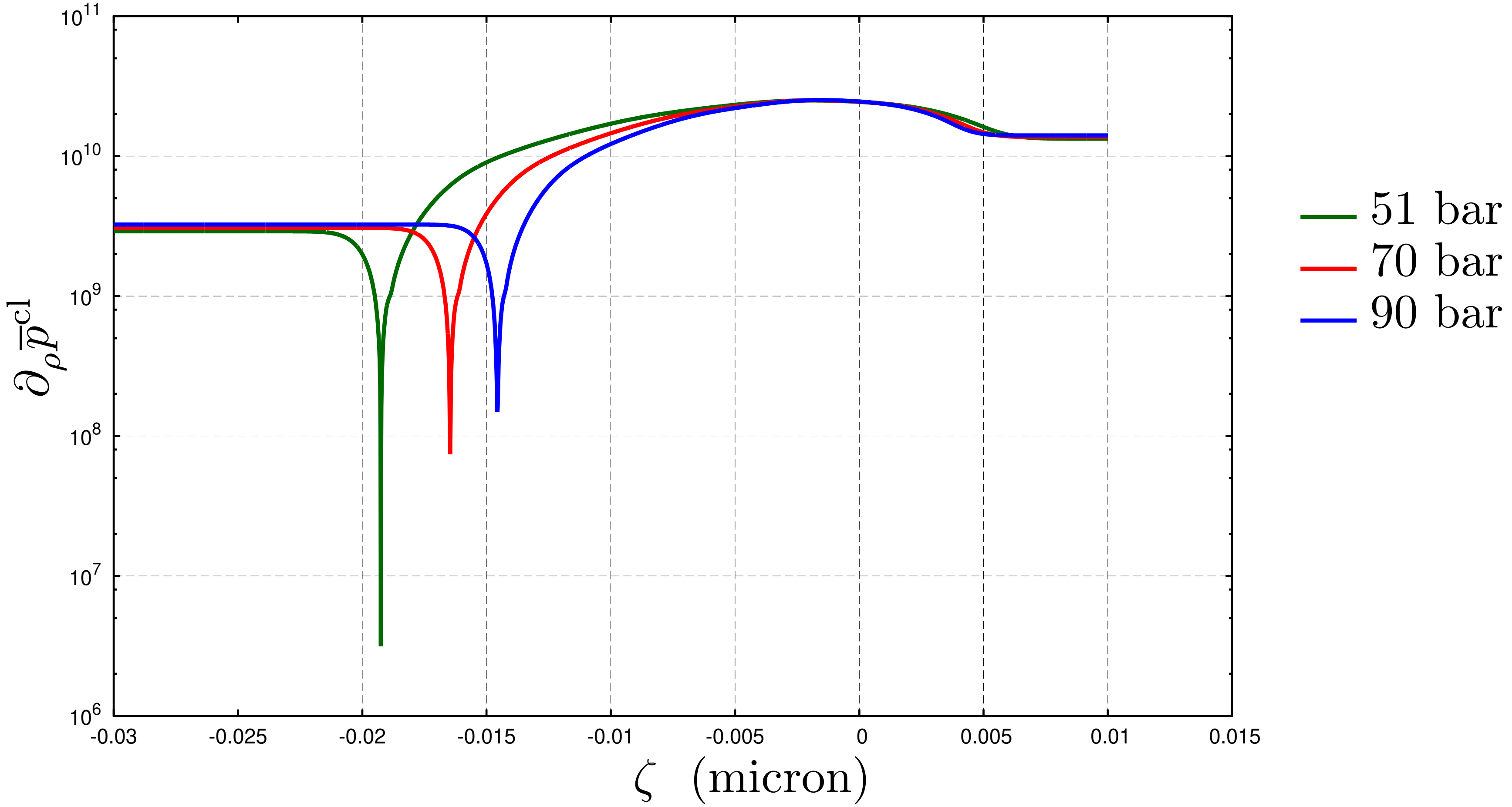
- **Explosion at mechanical thermodynamical unstable points**

  Explosion of h_i ν_i Γ_{ij}

  Thermodynamic form of transport fluxes

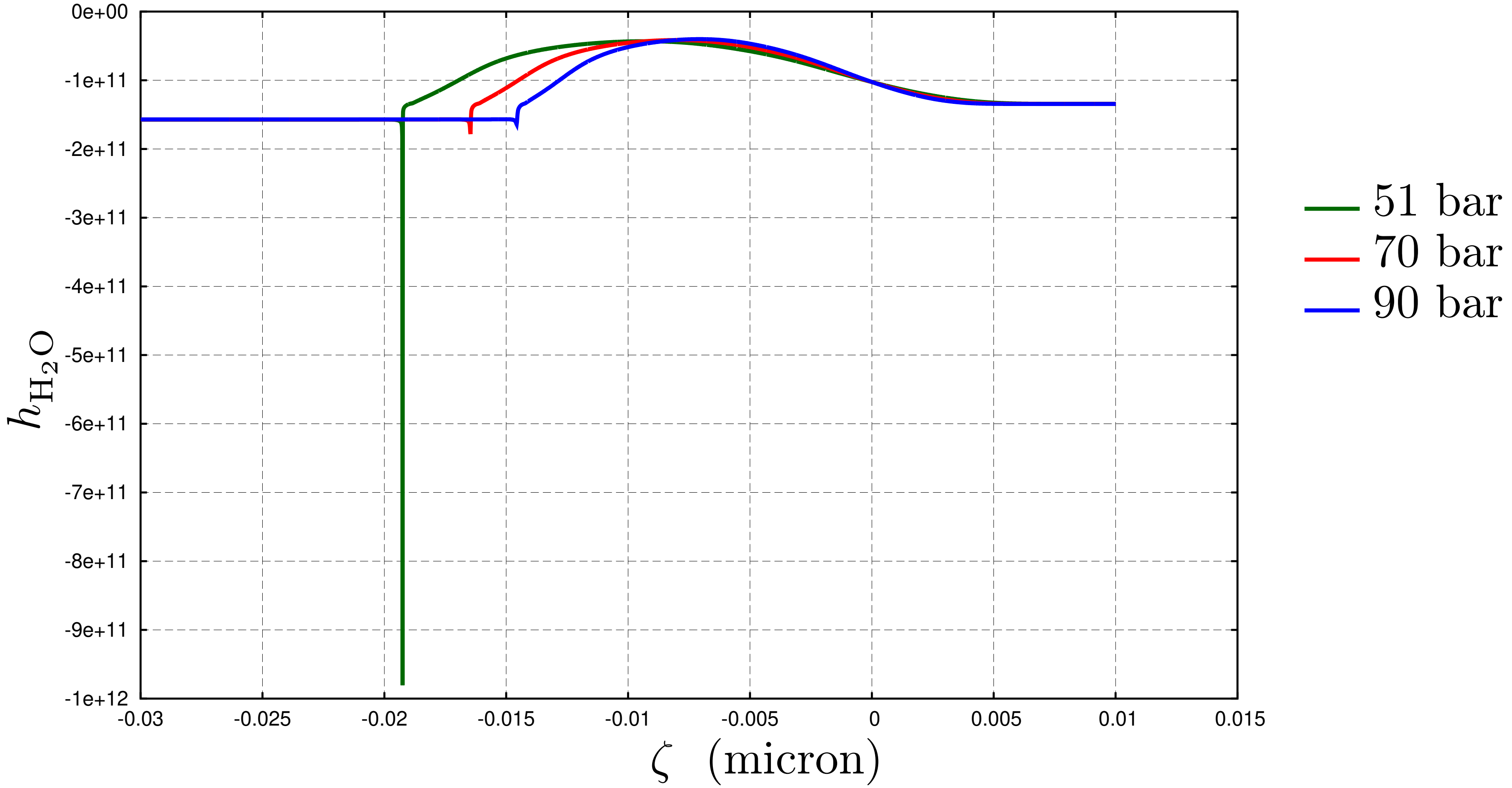
Transport matrix *L* (5)

- **Mechanical stability criterium in high pressure flames**



Transport matrix *L* (6)

- **Explosion of water mass specific enthalpy**



Transport matrix L (7)

- **New expression of specific enthalpies**

$$(\partial_{\mathsf{y}_k}\overline{p}^{\mathrm{cl}})_{T,\rho,\mathsf{y}_l} = \frac{m}{\rho m_i}(\partial_\rho \overline{p}^{\mathrm{cl}})_{T,\mathsf{y}_l} + \mathcal{R}_k$$

$$\mathcal{R}_k = \Big(\frac{RT}{(\nu-b)^2} + \frac{ma}{\nu(\nu+b)^2}\Big)\sum_{j\in\mathfrak{S}} \mathsf{y}_j\Big(\frac{b_k}{m_j} - \frac{b_j}{m_k}\Big)$$

$$+\frac{2m}{\nu(\nu+b)}\sum_{j,l\in\mathfrak{S}} \mathsf{y}_j\mathsf{y}_l\alpha_l\Big(\frac{\alpha_j}{m_k} - \frac{\alpha_k}{m_j}\Big)$$

- **Stabilization of the matrix L**

$$\widetilde{h}_k = (\partial_{\mathsf{y}_k}h)_{T,\rho,\mathsf{y}_l} - \frac{m}{\rho m_k}(\partial_\rho h)_{T,\mathsf{y}_l} - (\partial_\rho h)_{T,\mathsf{y}_l}\,\mathcal{R}_k \,/\, \mathcal{A}\big((\partial_\rho \overline{p}^{\mathrm{cl}})_{T,\mathsf{y}_l}\big)$$

$\mathcal{A}(x)$ smoothed version of $\mathcal{A}(x) = \max(\delta, x)$

Supercritical fluids

- **Experiments**

  Schilling and Franck (1988), Chehroudi, Talley, and Coy (2002),

  Candel, Juniper, Singla, Scouflaire, Rolon (2006), Habiballah et al. (2006)

- **Numerical simulations of supercritical laminar flames**

  El Gamal, Gutheil and Warnatz (2000), Okongo and Bellan (2002),

  Saur, Behrendt, and Franck (1993),

  Ribert, Zong, Yang, Pons, Darabiha, and Candel, (2008), Pons et al. (2009),

- **Numerical simulations of supercritical turbulent flames**

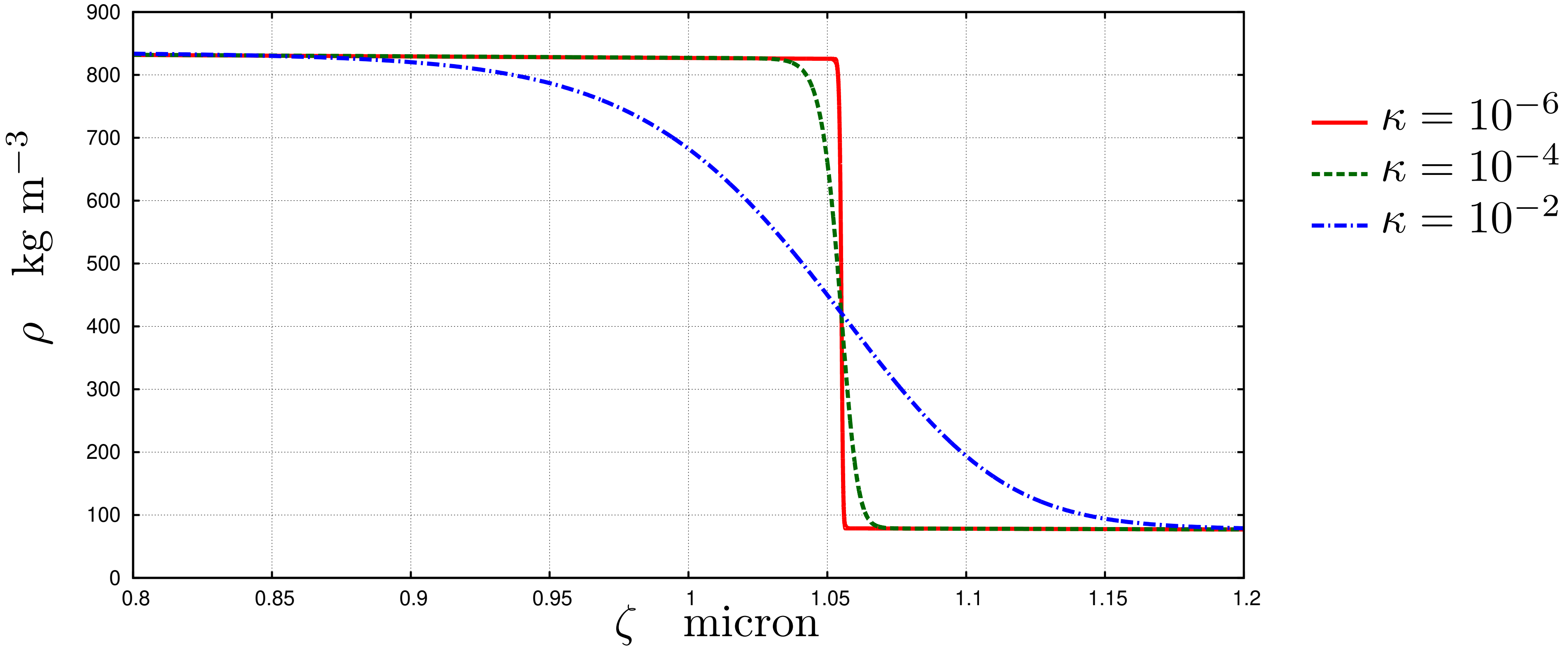
  Zong and Yang (2006), Bellan (2006), Oefelein (2005),

  Zong and Yang (2007), Schmitt, Selle, Cuenot, and Poinsot (2008)

  Schmitt, Méry, Boileau, and Candel (2011), Dahms and Oefelein (2013)

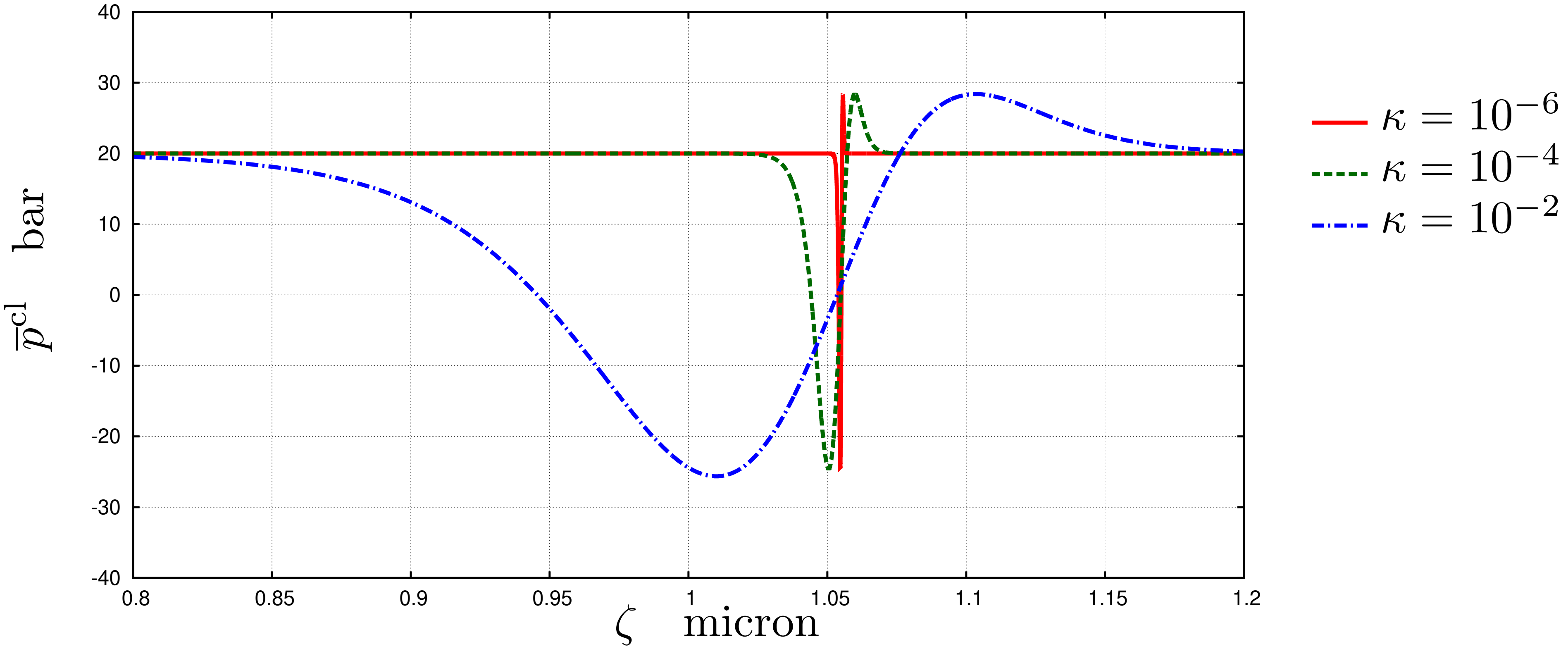
Oxygen vaporizing diffuse interfaces (1)

- $p^\infty = 20$ bars $T_{\rm lo} = 100$ K $T_{\rm up} = 300$ K $\alpha = 1000\ {\rm s}^{-1}$



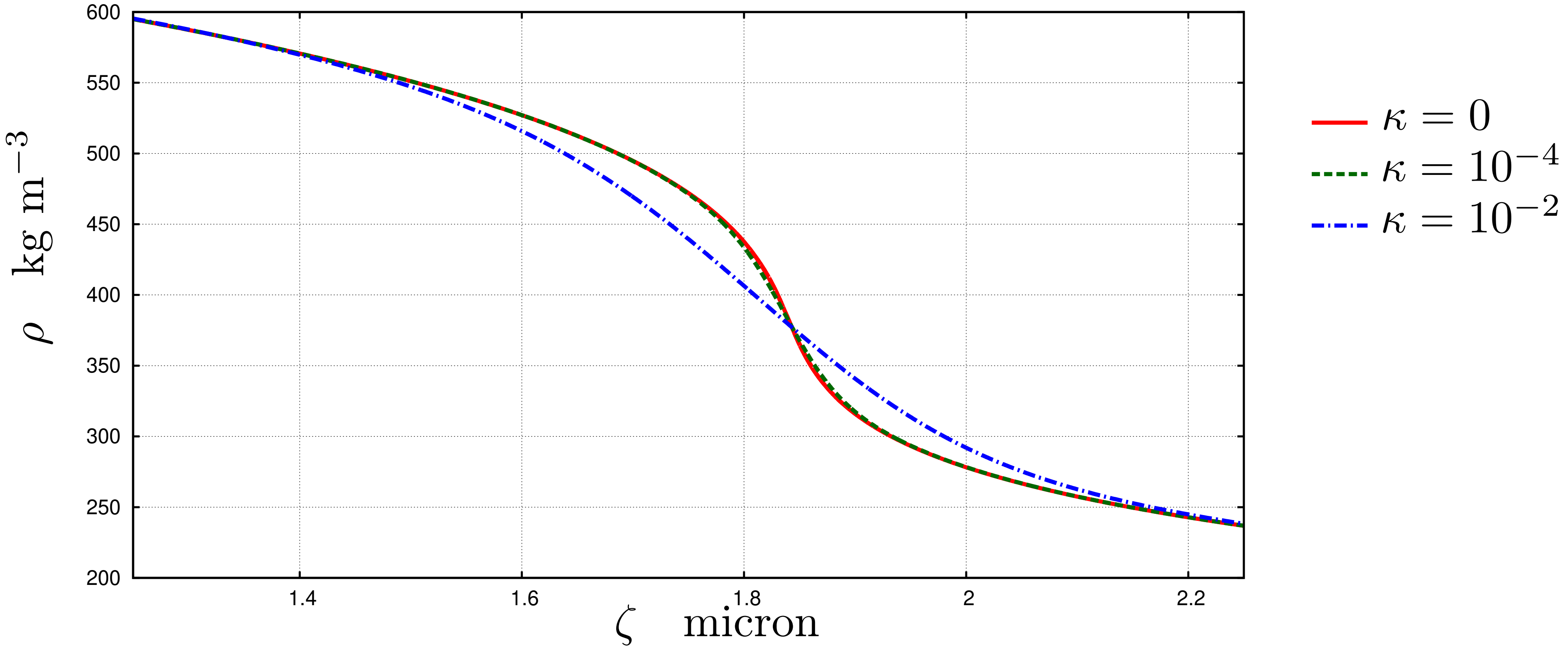
Oxygen vaporizing diffuse interfaces (2)

- $p^\infty = 20$ bars $\quad T_{\text{lo}} = 100$ K $\quad T_{\text{up}} = 300$ K $\quad \alpha = 1000\ \text{s}^{-1}$



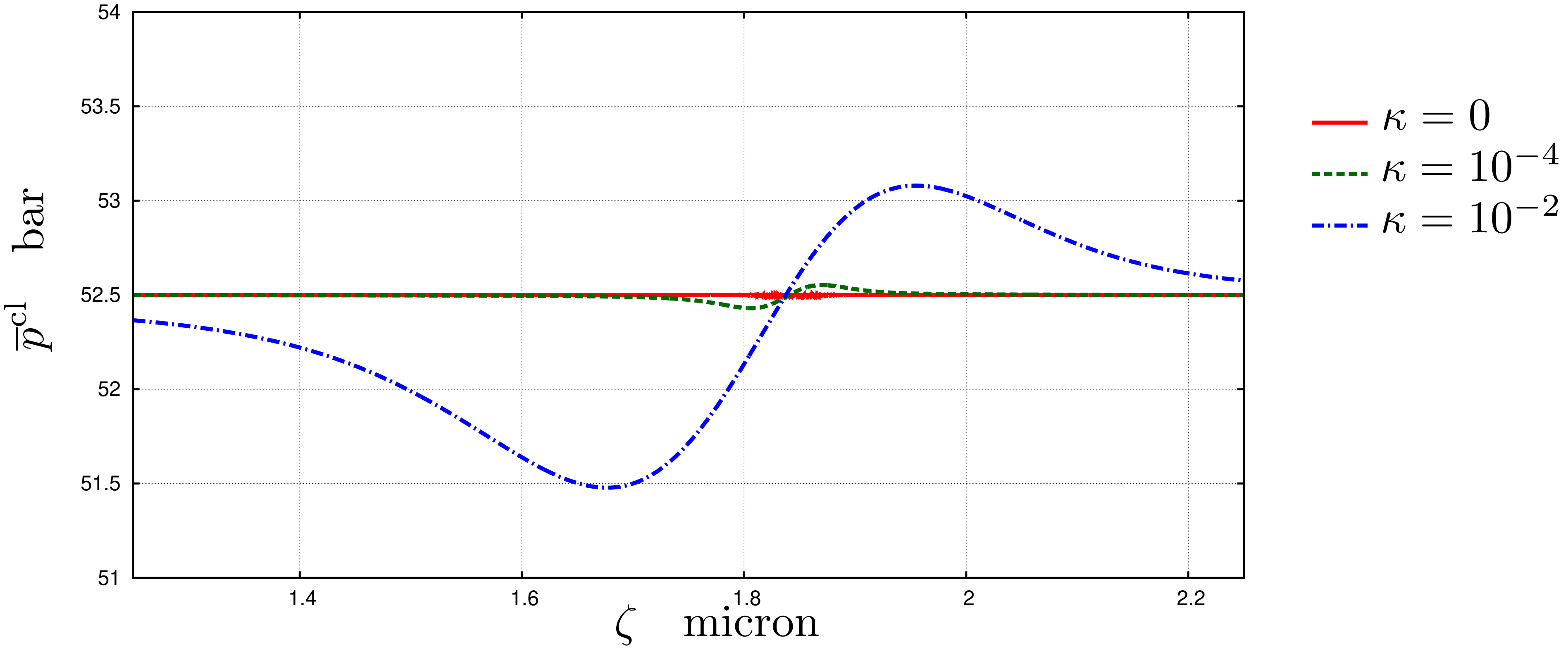
Oxygen vaporizing diffuse interfaces (3)

- $p^\infty = 52.5$ bars $T_{\rm lo} = 100$ K $T_{\rm up} = 300$ K $\alpha = 1000\ {\rm s}^{-1}$



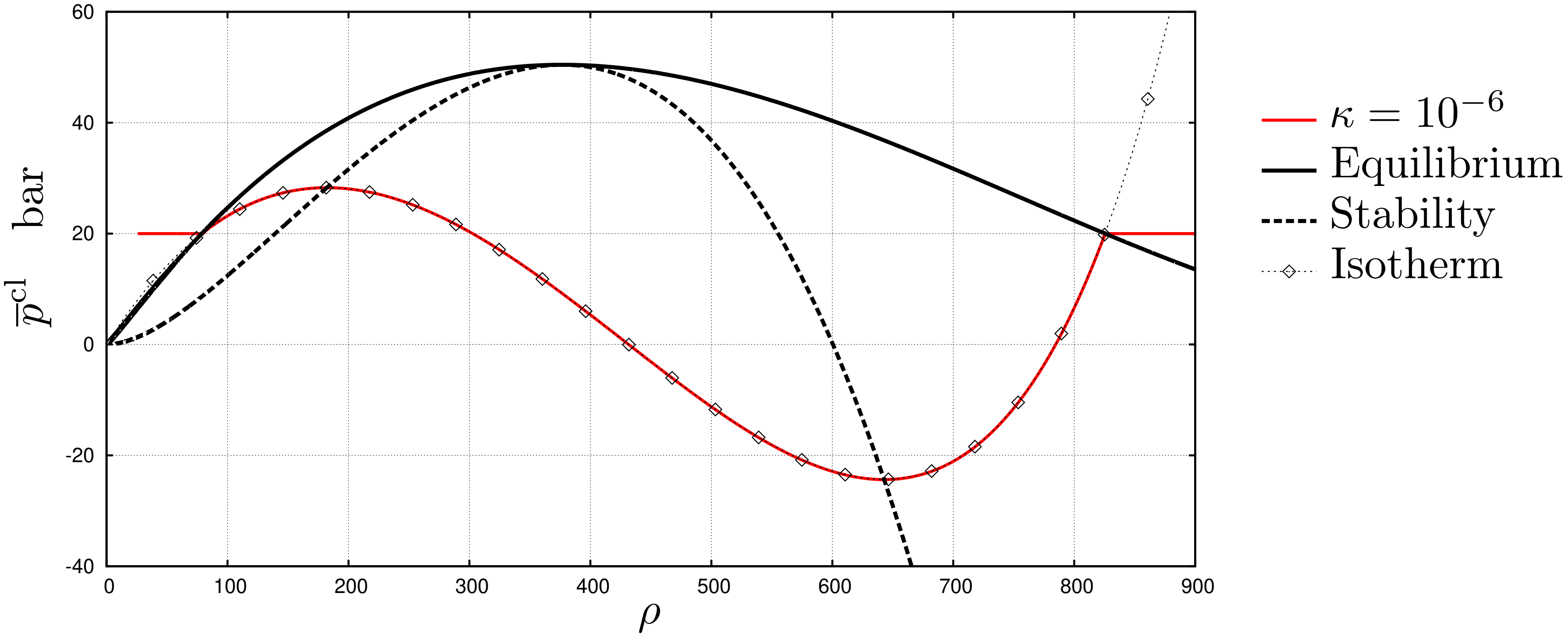
Oxygen vaporizing diffuse interfaces (4)

- $p^{\infty} = 52.5$ **bars** $\quad T_{\rm lo} = 100$ **K** $\quad T_{\rm up} = 300$ **K** $\quad \alpha = 1000\ \mathrm{s}^{-1}$



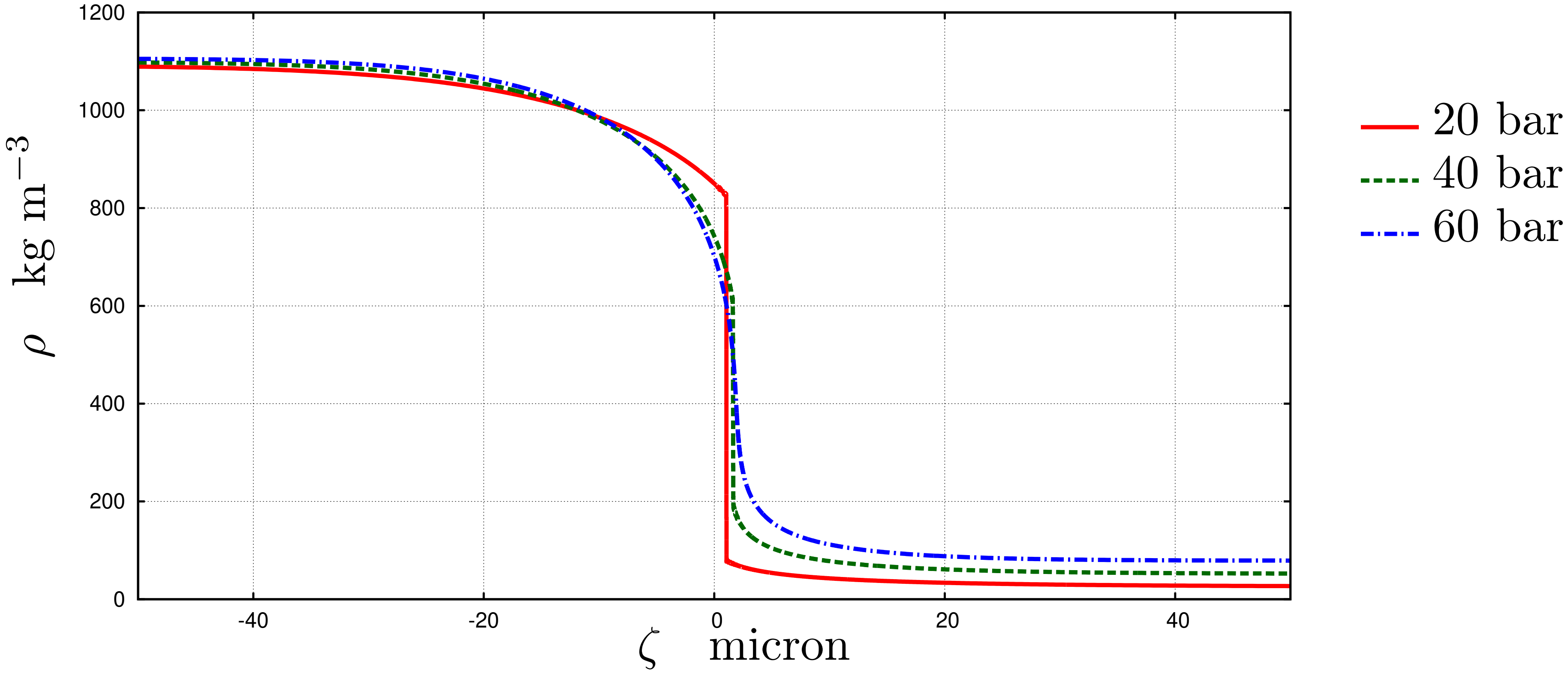
Oxygen vaporizing diffuse interfaces (5)

- $p^{\infty} = 20$ bars $\quad T_{lo} = 100$ K $\quad T_{up} = 300$ K $\quad \alpha = 1000\ s^{-1}$



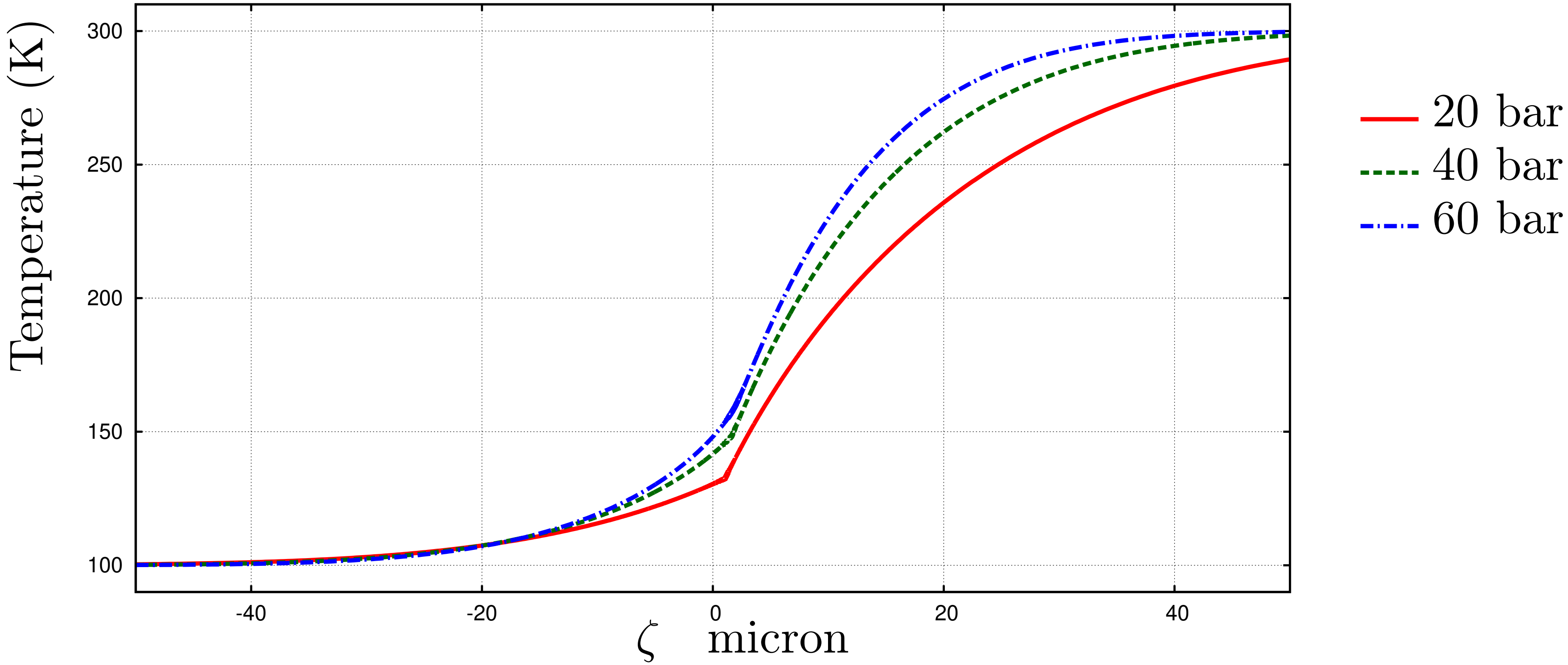
Oxygen vaporizing diffuse interfaces (6)

- $T_{lo} = 100$ K $\quad T_{up} = 300$ K $\quad \alpha = 1000$ s^{-1}



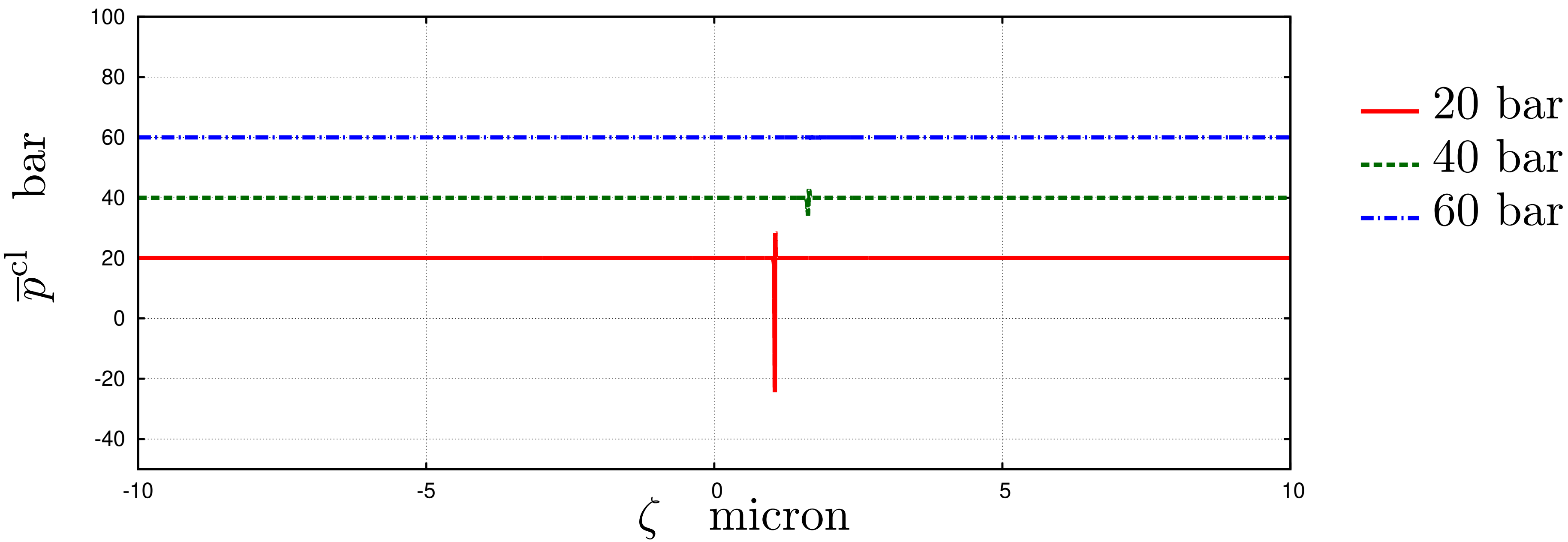
Oxygen vaporizing diffuse interfaces (7)

- $T_{\mathrm{lo}} = 100$ K $\quad T_{\mathrm{up}} = 300$ K $\quad \alpha = 1000\ \mathrm{s}^{-1}$



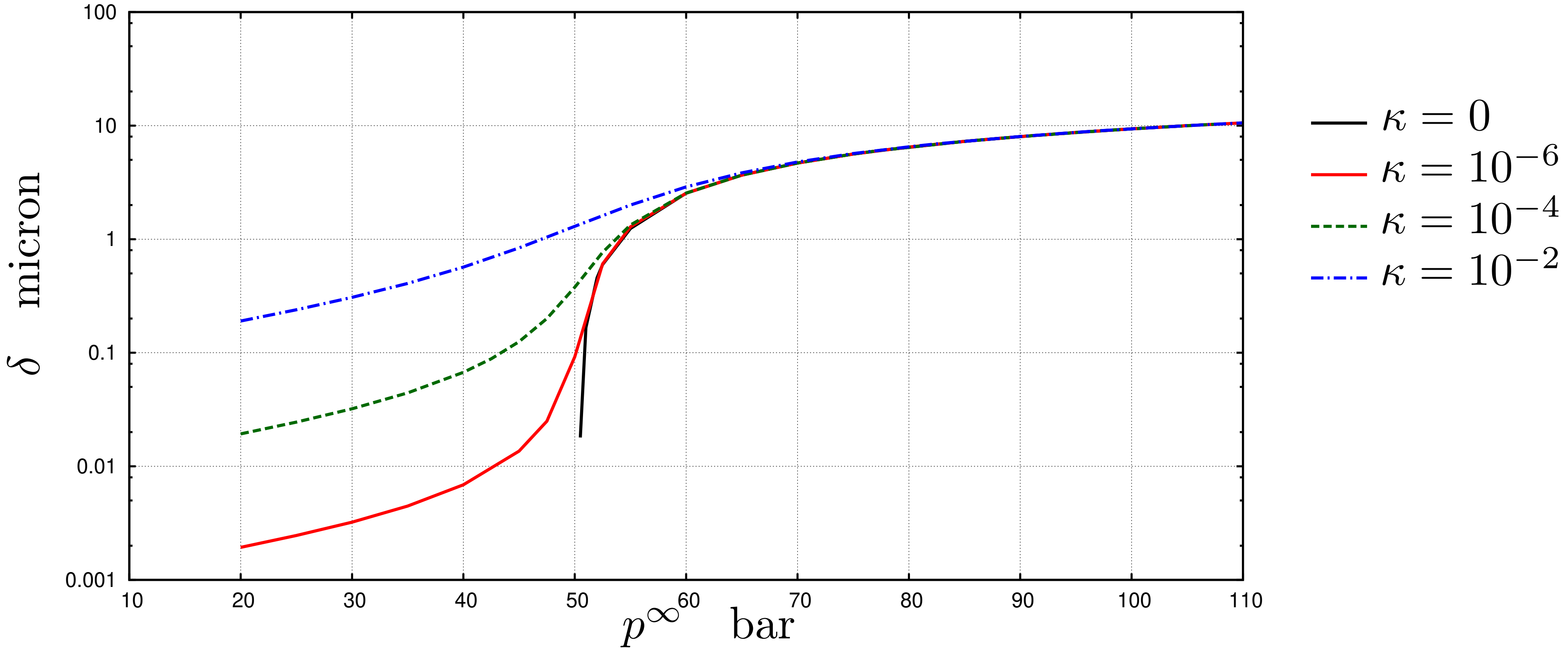
Oxygen vaporizing diffuse interfaces (8)

- $T_{\mathrm{lo}} = 100$ K $\quad T_{\mathrm{up}} = 300$ K $\quad \alpha = 1000\ \mathrm{s}^{-1}$



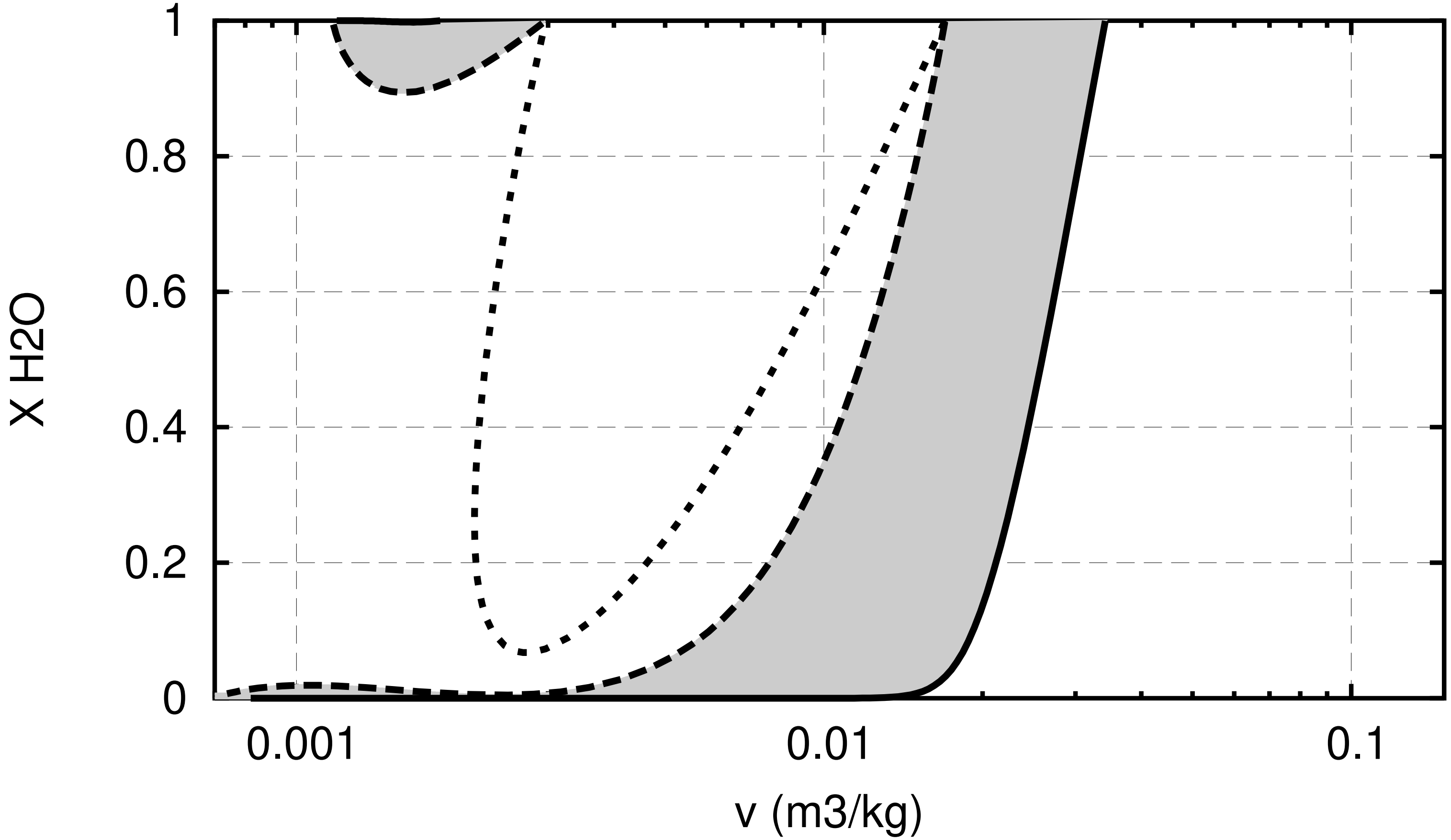
Oxygen vaporizing diffuse interfaces (9)

- $T_{\rm lo} = 100$ K $\quad T_{\rm up} = 300$ K $\quad \alpha = 1000\ \mathrm{s}^{-1}$



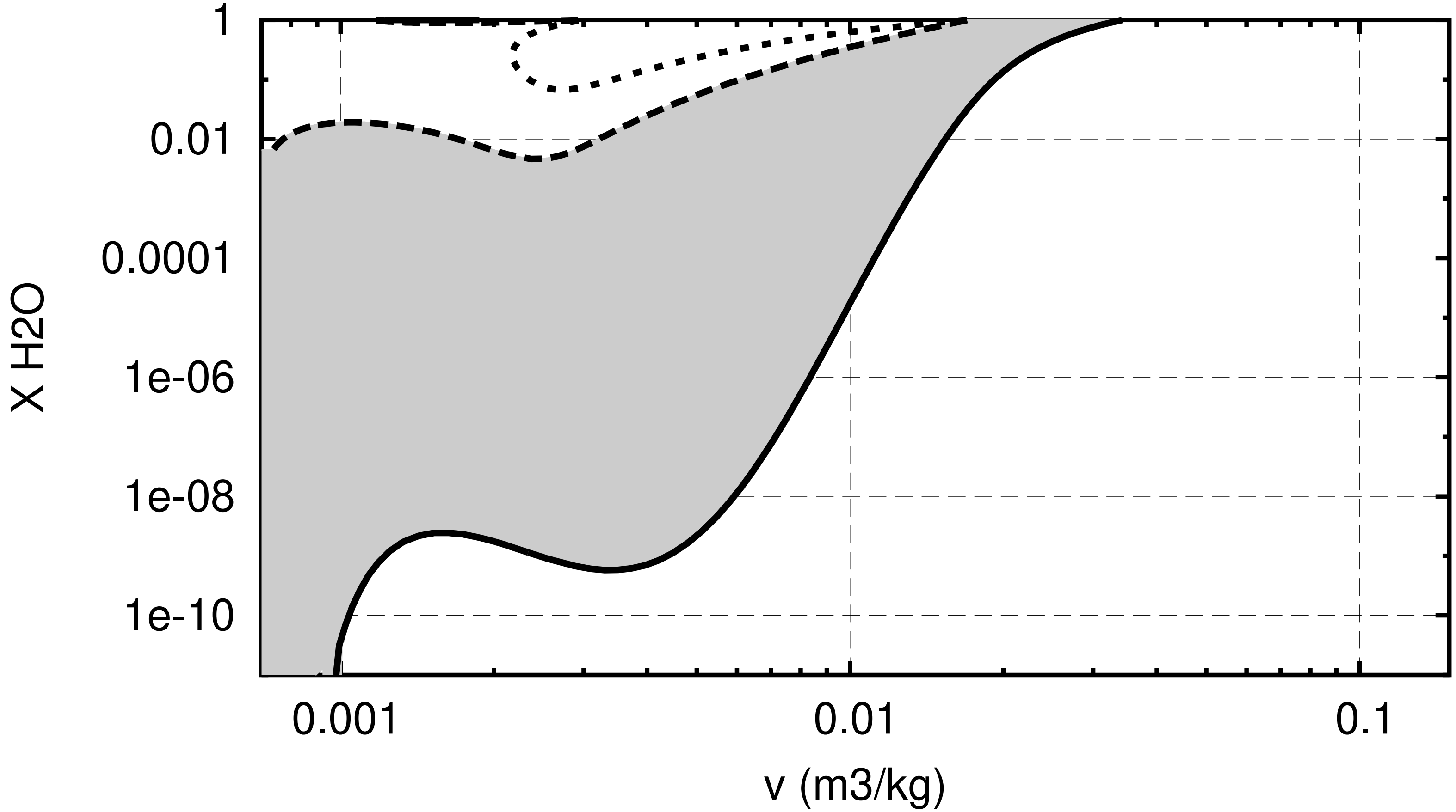
Stability (1)

- **Stability of O_2/H_2O mixtures at $p^\infty = 60$ atm**



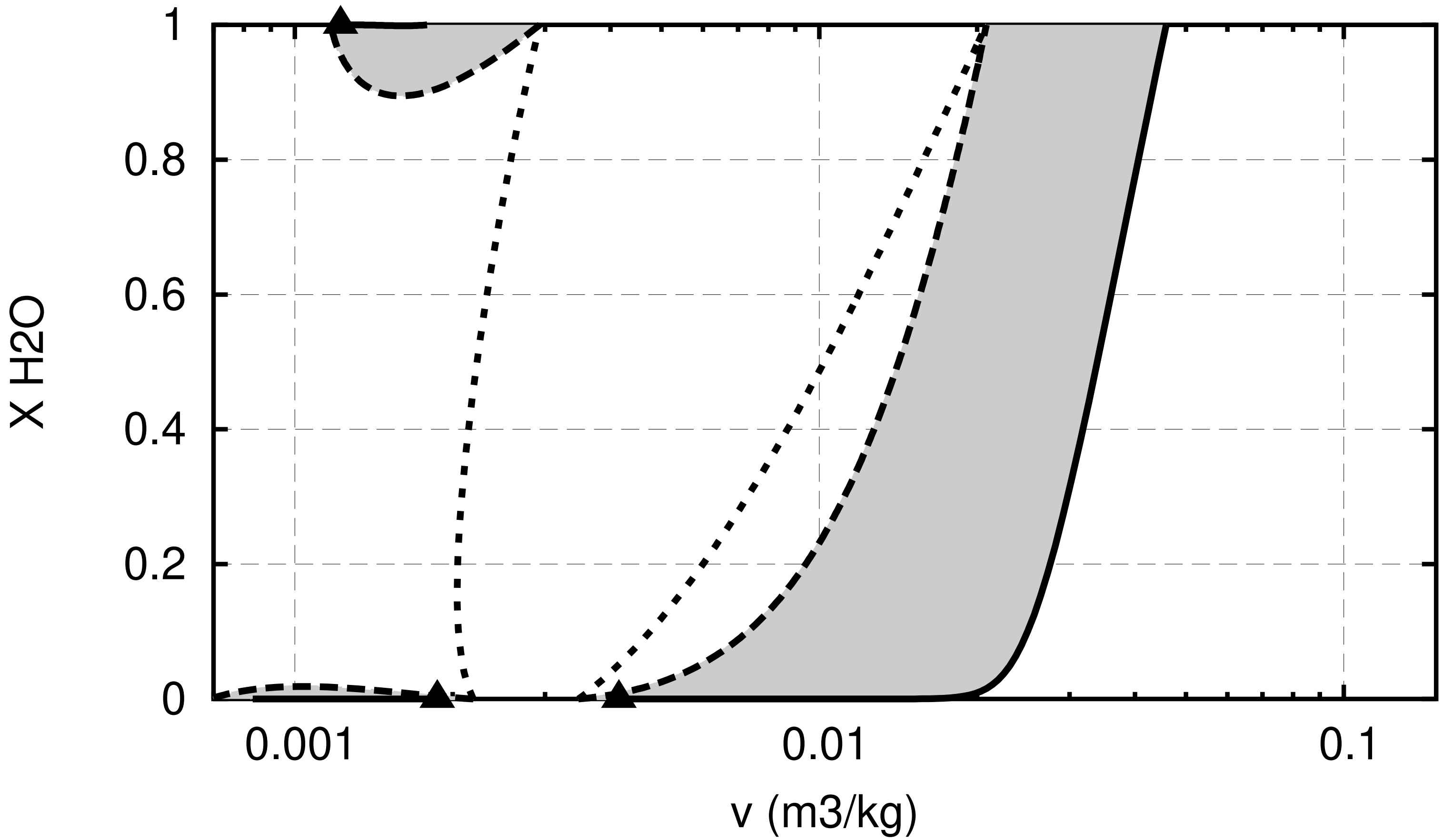
Stability (2)

- **Stability of O_2/H_2O mixtures at $p^\infty = 60$ atm**



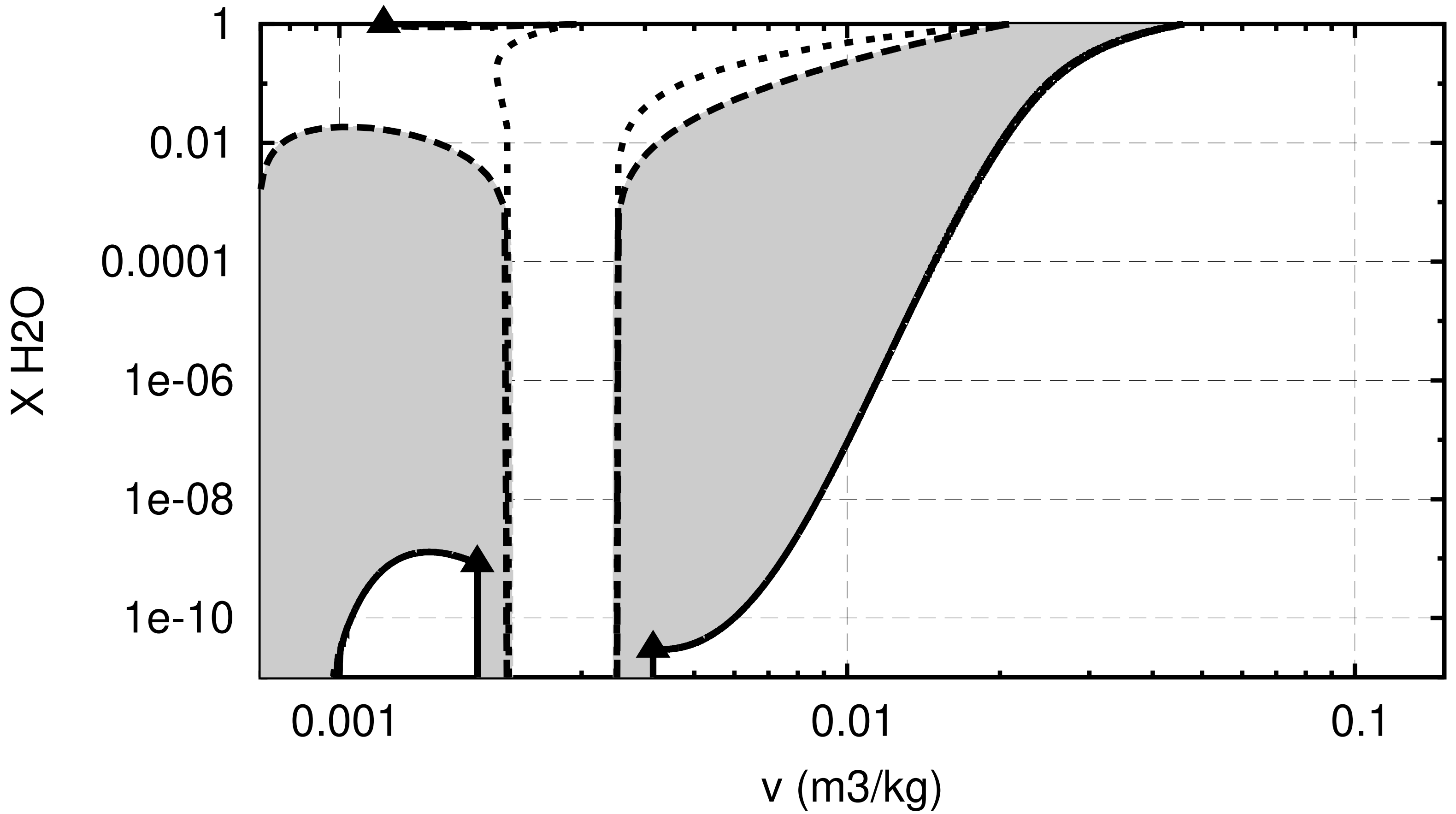
Stability (3)

- **Stability of O_2/H_2O mixtures at $p^\infty = 45$ atm**



Stability (4)

- **Stability of O_2/H_2O mixtures at $p^{\infty} = 45$ atm**



Stability (5)

- **Types of binary phase diagrams from Van Konynenburg and Scott**

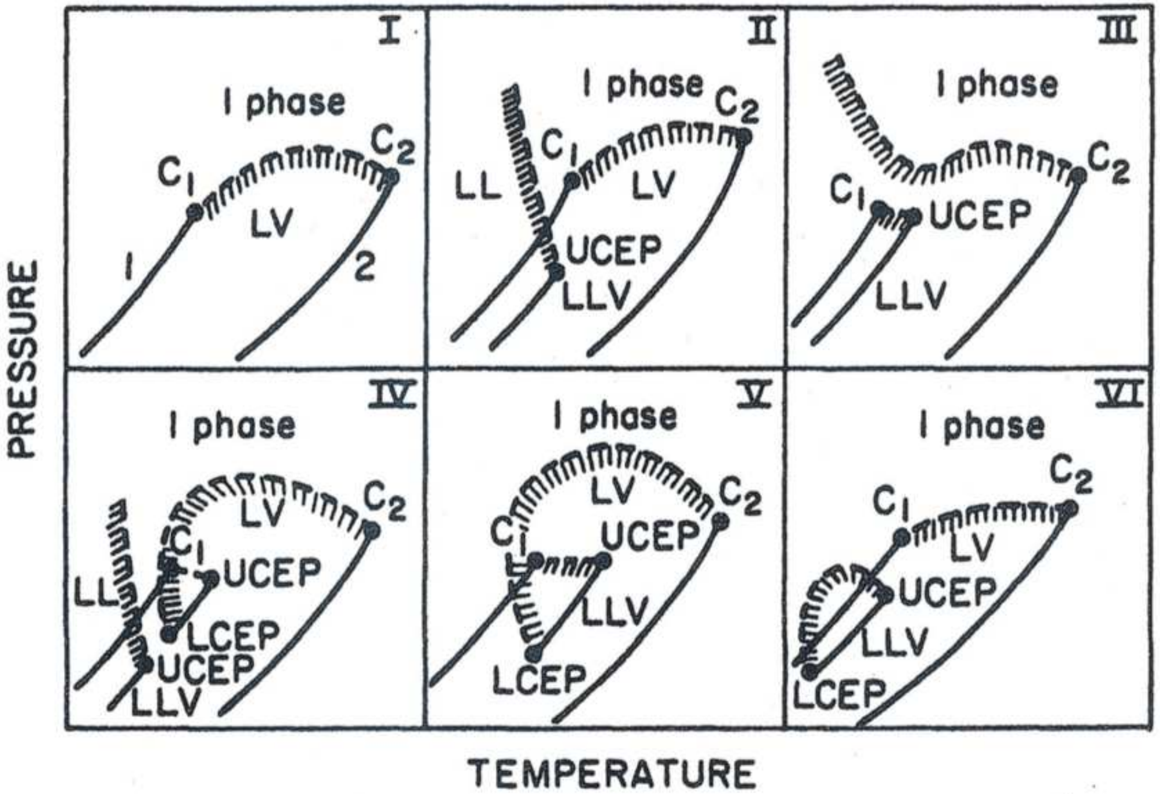


Figure 12-6 Six types of phase behavior in binary fluid systems. C = critical point; L = liquid; V = vapor; UCEP = upper critical end point; LCEP = lower critical end point. Dashed curves are critical lines and hatching marks heterogeneous regions.

Liquid water governing equations

- **Condensation of water (below 220.6 bar)**

$$H_2O \rightleftarrows H_2O(l) \qquad (\star)$$

- **Governing equation**

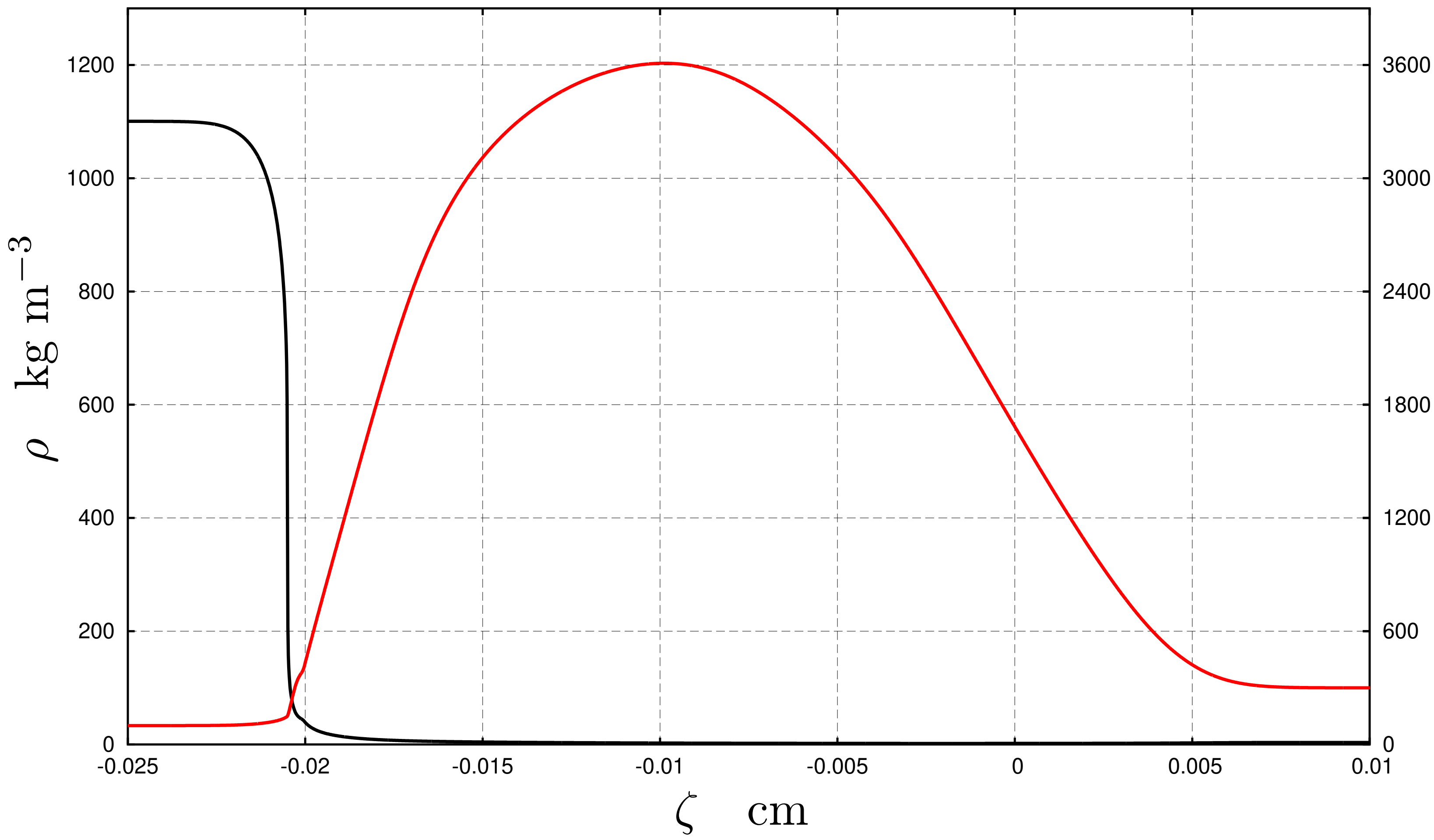
$$\partial_t \rho_{\mathsf{n}_s+1} + \boldsymbol{\nabla}\cdot(\rho_{\mathsf{n}_s+1}\boldsymbol{v}) = m_{\mathsf{n}_s+1}\omega_{\mathsf{n}_s+1}$$

- **Condensation source term**

$$\omega_{\mathsf{n}_s+1} = \mathcal{K}_\star\big(\exp(\mu_{\mathrm{H_2O}}) - \exp(\mu_{\mathrm{H_2O}(l)})\big) \simeq \mathcal{K}'_\star\big(g^{\mathrm{cl}}_{\mathrm{H_2O}} - g^{\mathrm{cl}}_{\mathrm{H_2O}(l)}\big)$$

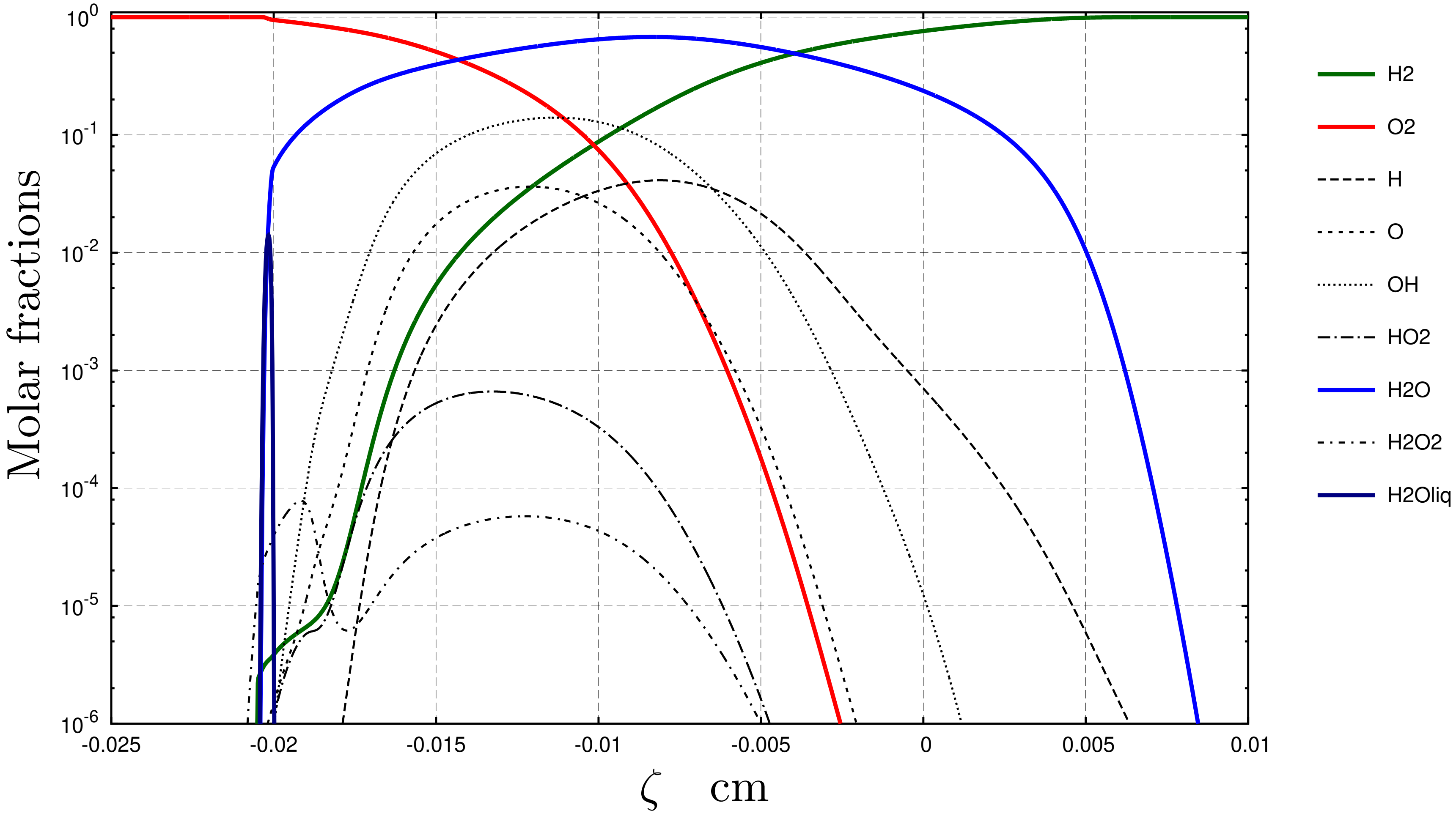
Oxygen/Hydrogen diffusion flame (1)

- $p^{\infty} = 45$ bars $\quad T_{\text{lo}} = 100$ K $\quad T_{\text{up}} = 300$ K $\quad \alpha = 10000\ \text{s}^{-1}$



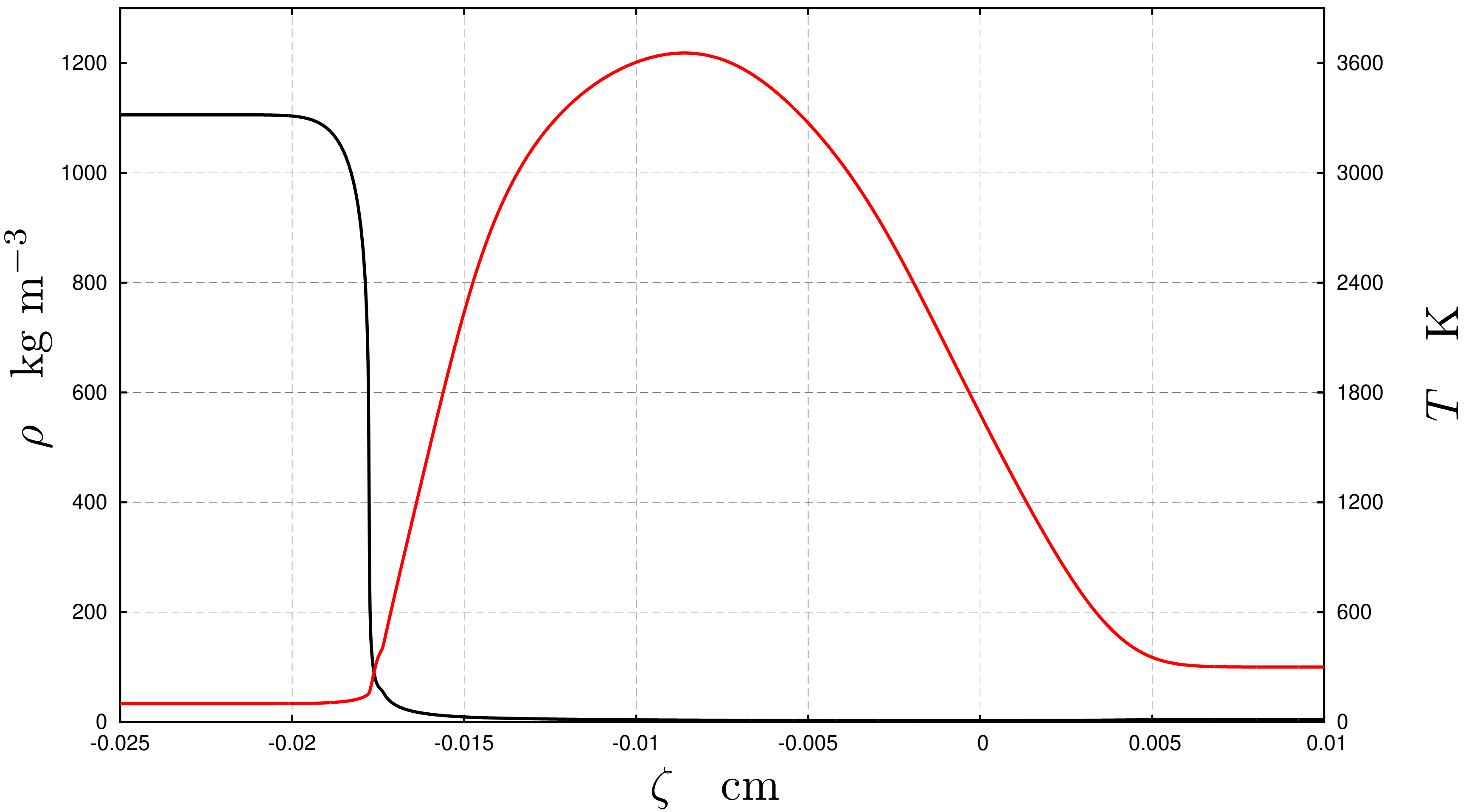
Oxygen/Hydrogen diffusion flame (2)

- $p^{\infty} = 45$ **bars** $\quad T_{\mathrm{lo}} = 100$ **K** $\quad T_{\mathrm{up}} = 300$ **K** $\quad \alpha = 10000$ $\mathbf{s^{-1}}$



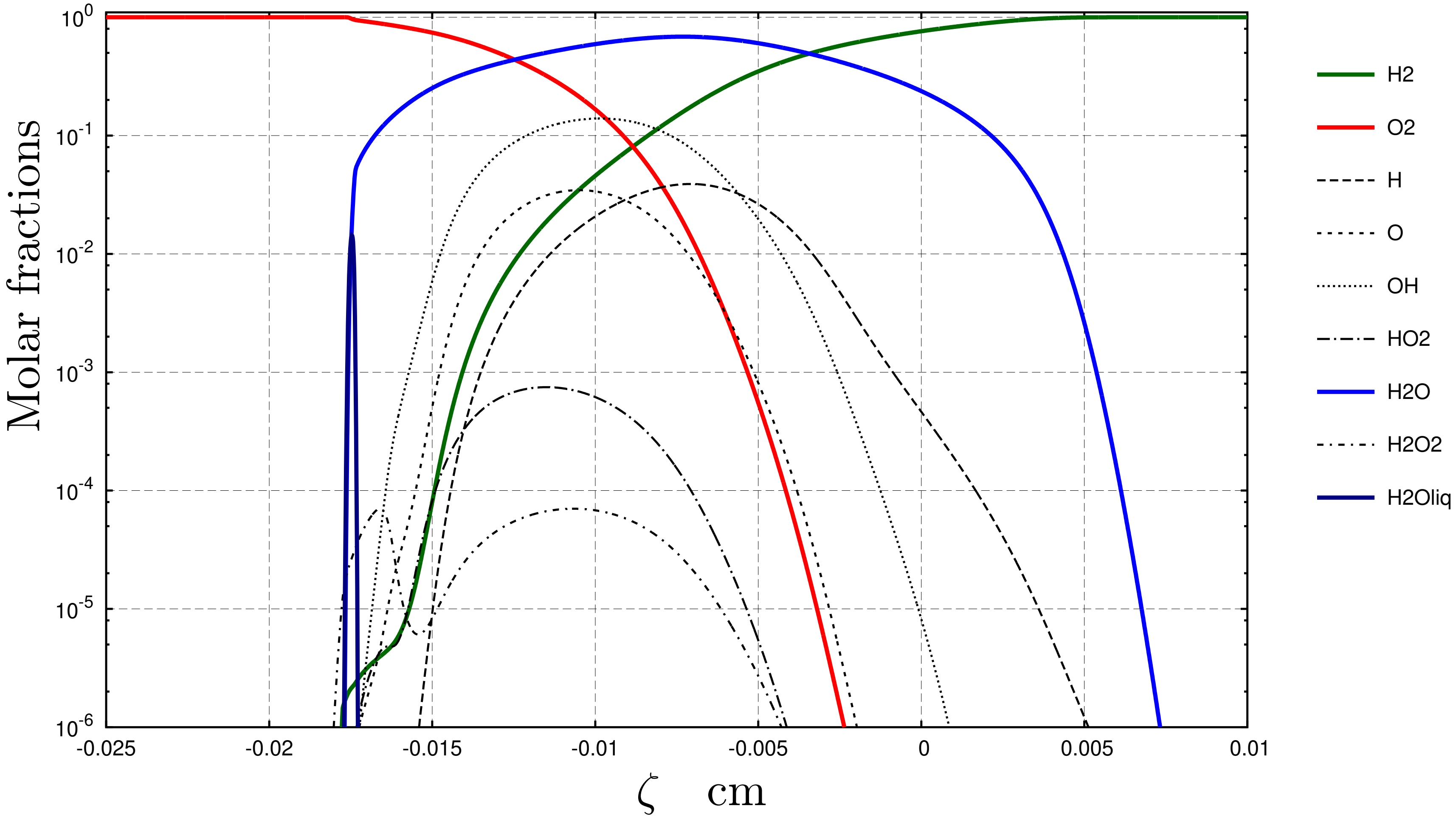
Oxygen/Hydrogen diffusion flame (3)

- $p^{\infty} = 60$ bars $\quad T_{\mathrm{lo}} = 100$ K $\quad T_{\mathrm{up}} = 300$ K $\quad \alpha = 10000\ \mathrm{s}^{-1}$



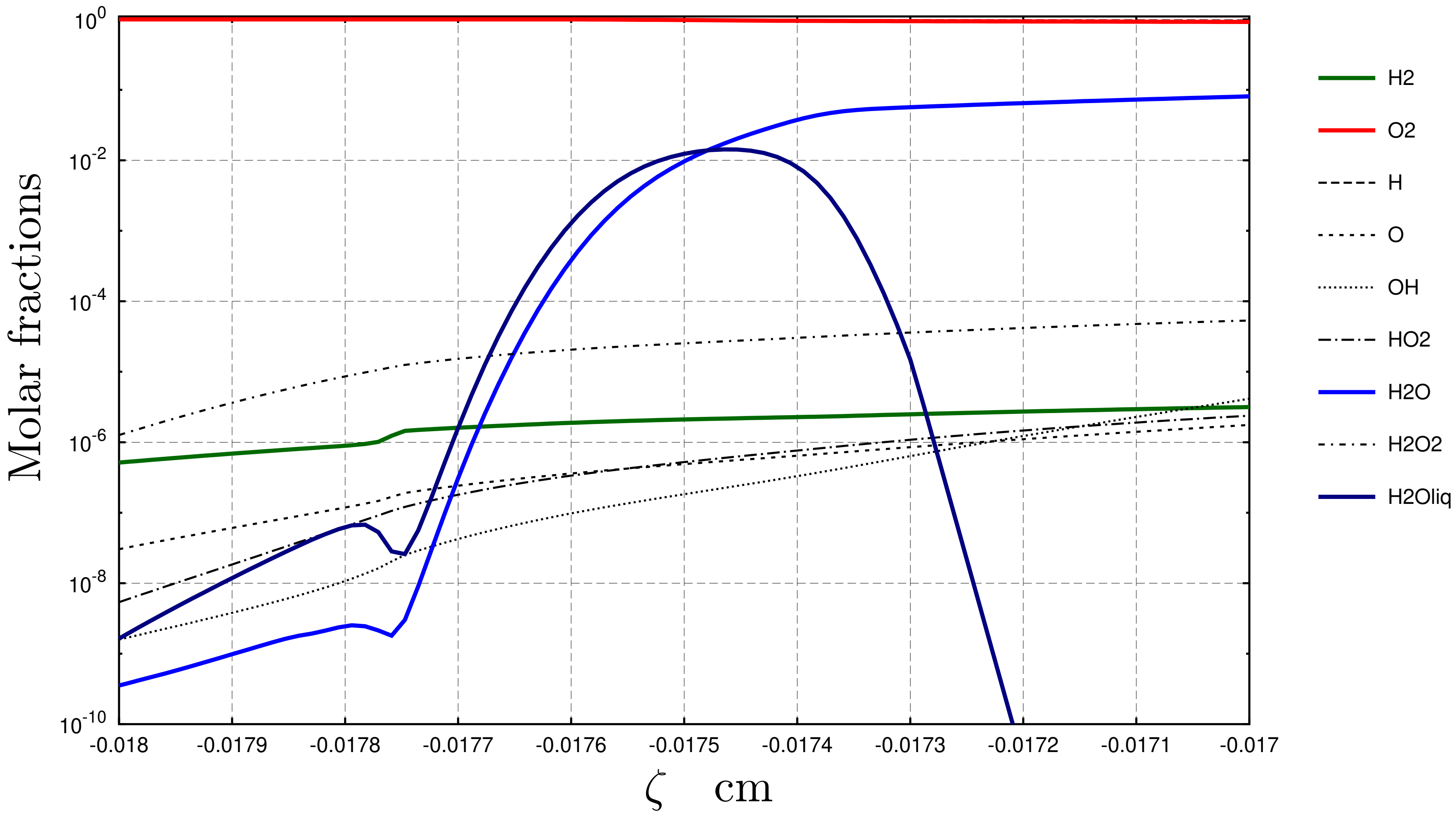
Oxygen/Hydrogen diffusion flame (4)

- $p^\infty = 60$ bars $\quad T_{\mathrm{lo}} = 100$ K $\quad T_{\mathrm{up}} = 300$ K $\quad \alpha = 10000\ \mathrm{s}^{-1}$



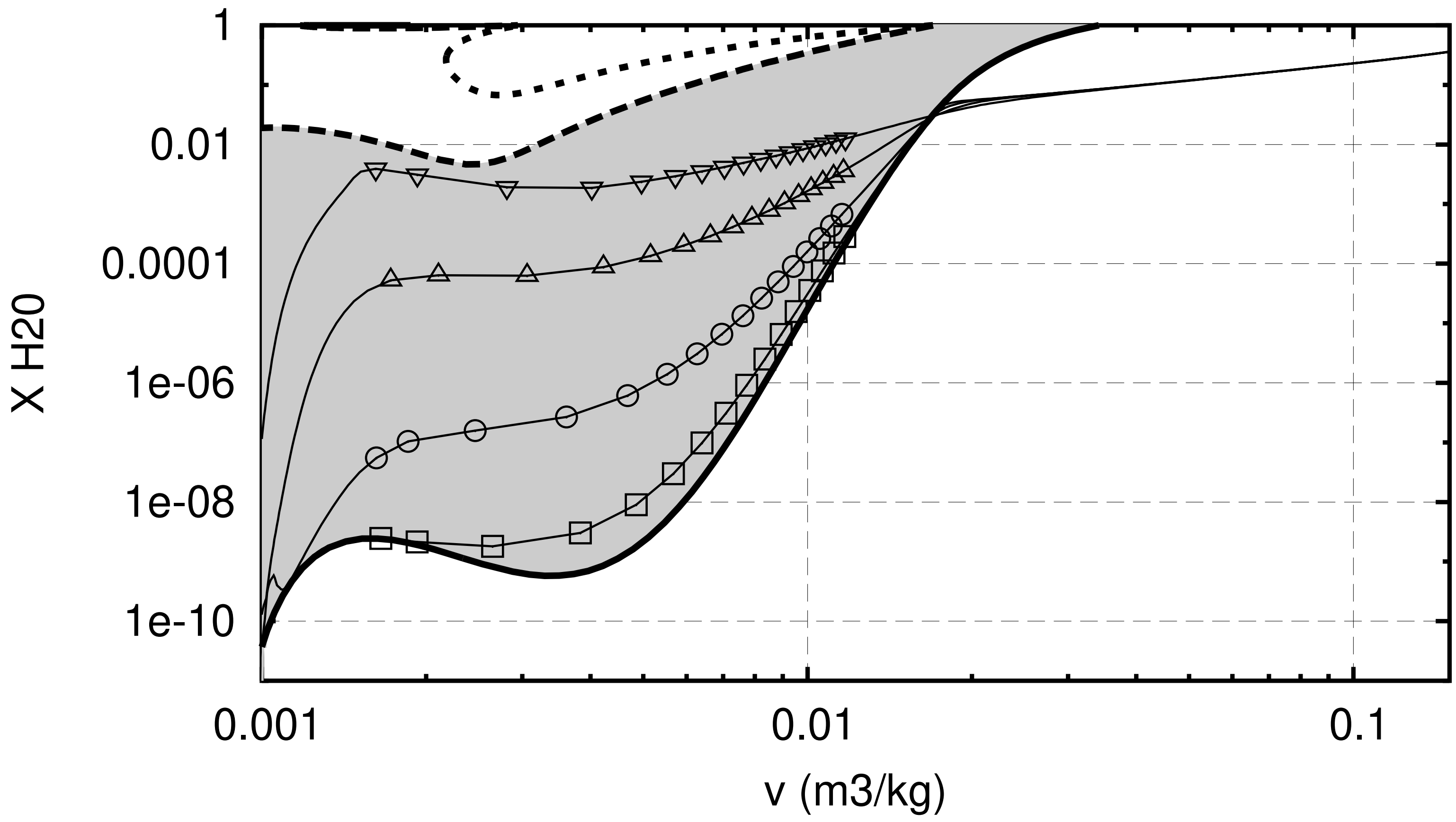
Oxygen/Hydrogen diffusion flame (5)

- $p^{\infty} = 60$ bars $\quad T_{\mathrm{lo}} = 100$ K $\quad T_{\mathrm{up}} = 300$ K $\quad \alpha = 10000\ \mathrm{s}^{-1}$



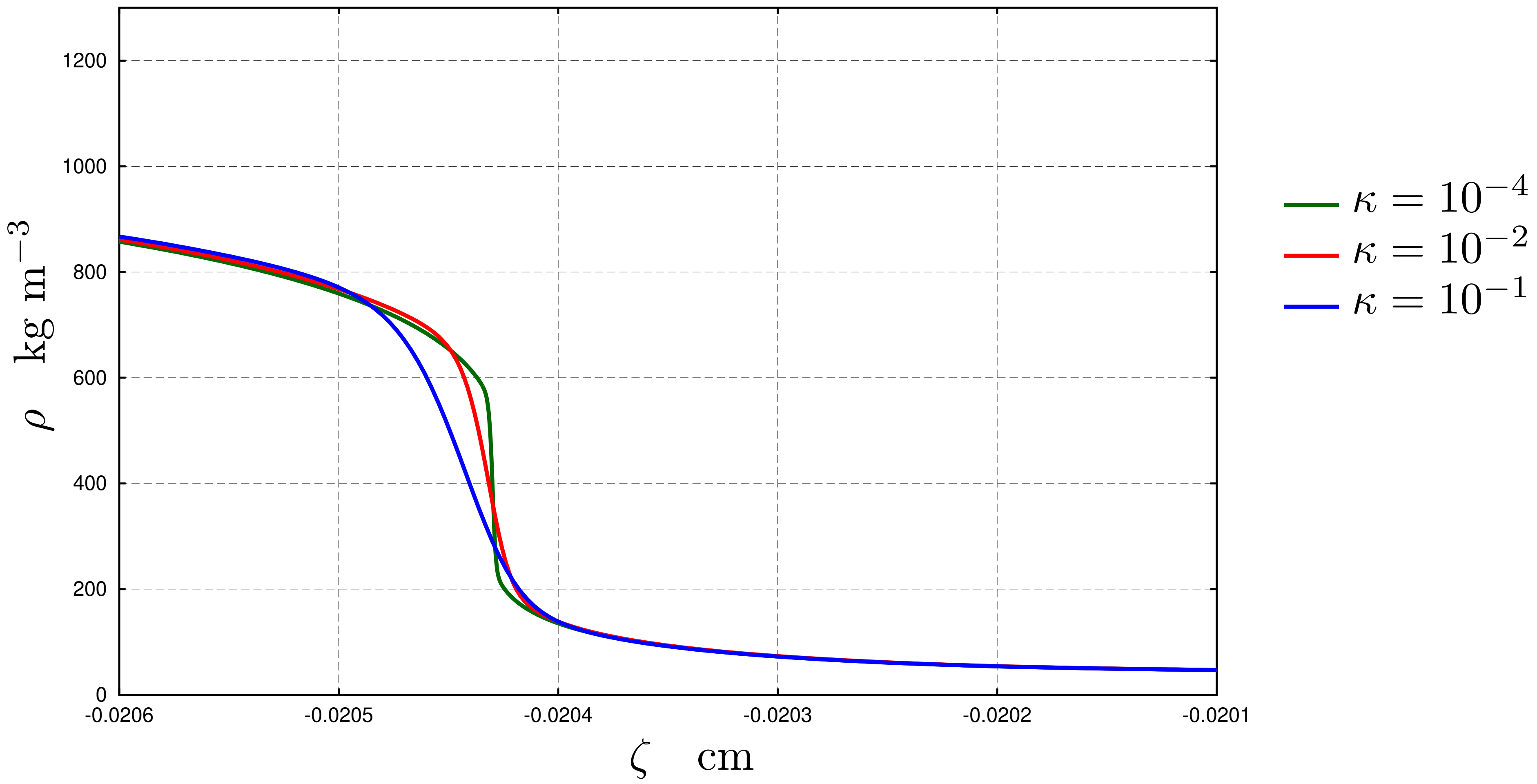
Oxygen/Hydrogen diffusion flame (6)

- $p^{\infty} = 60$ bars $\quad T_{\mathrm{lo}} = 100$ K $\quad T_{\mathrm{up}} = 300$ K $\quad \alpha = 10000\ \mathrm{s}^{-1}$



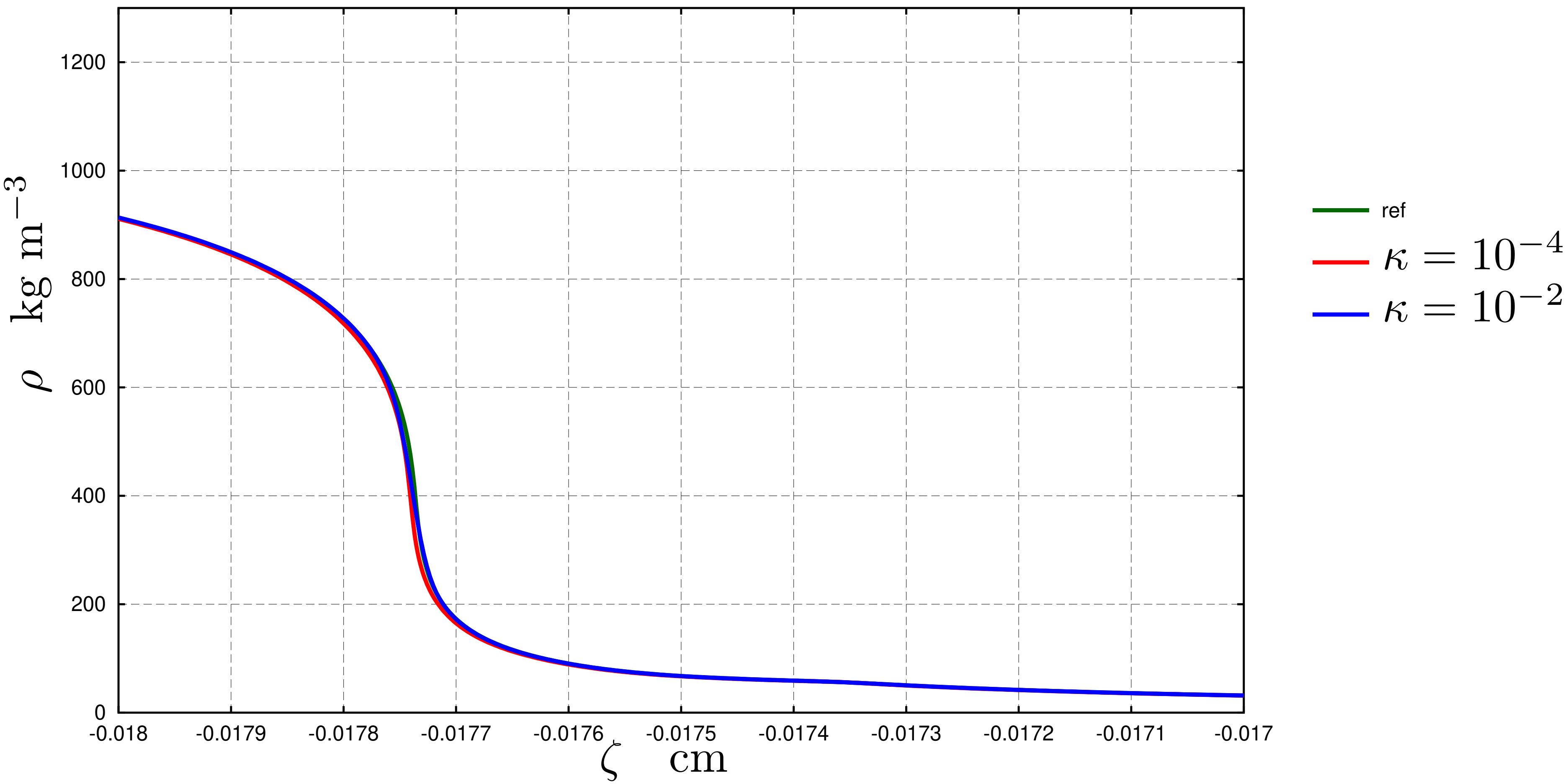
Oxygen/Hydrogen diffusion flame (7)

- $p^\infty = 45$ bars $\quad T_{\text{lo}} = 100$ K $\quad T_{\text{up}} = 300$ K $\quad \alpha = 10000\ \text{s}^{-1}$



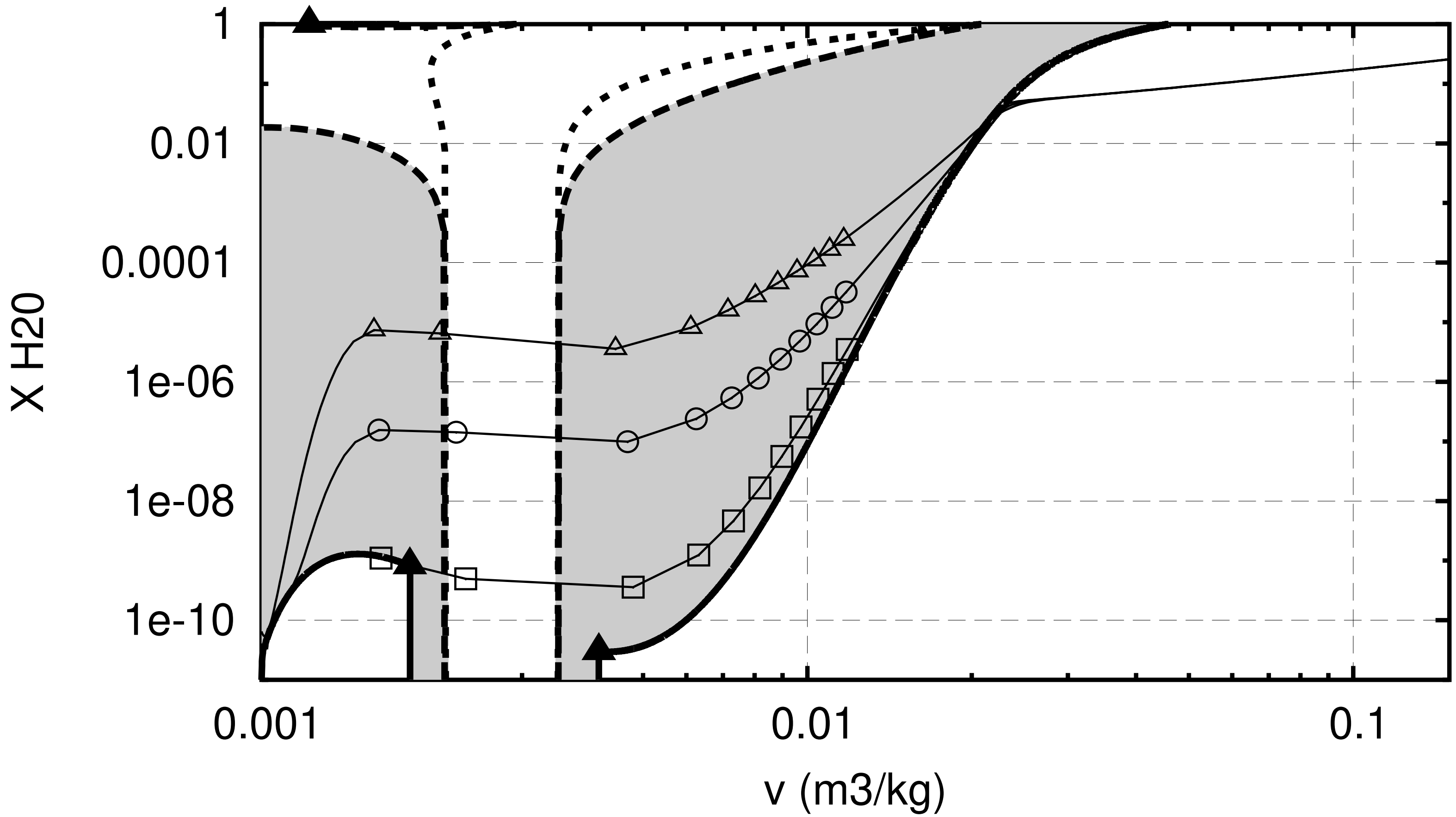
Oxygen/Hydrogen diffusion flame (8)

- $p^{\infty} = 60$ **bars** $\quad T_{\text{lo}} = 100$ **K** $\quad T_{\text{up}} = 300$ **K** $\quad \alpha = 10000\ \text{s}^{-1}$



Oxygen/Hydrogen diffusion flame (9)

- **Stability of O_2/H_2O mixtures at $p^\infty = 45$ atm**



7 Conclusion

Conclusion/Future work

- **Physical/Modeling aspects**

  High pressure transport coefficients

  Numerical simulations at the Molecular/Boltzmann/Fluid levels

  Boundary equations at solid walls

- **Mathematical and numerical aspects aspects**

  Numerical simulations of subcritical to supercritical mixtures of fluids

  Global existence results around stationary nonconstant equilibrium states

  Decay estimates for multicomponent reactive flows and augmented systems

  Multicomponent mixtures and Cahn-Hilliard equations

High pressure multicomponent fluid models (1)

- **Cahn-Hilliard fluid mixtures from the kinetic theory**

$$\partial_t \rho_i + \boldsymbol{\nabla}\cdot(\rho_i \boldsymbol{v}) + \boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{F}}_i = m_i \omega_i \qquad i \in \mathfrak{S} = \{1, \dots, \mathsf{n}_{\mathrm{s}}\}$$

$$\partial_t(\rho \boldsymbol{v}) + \boldsymbol{\nabla}\cdot(\rho \boldsymbol{v} \otimes \boldsymbol{v}) + \boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{P}} = 0$$

$$\partial_t\big(\mathcal{E} + \tfrac{1}{2}\rho|\boldsymbol{v}|^2\big) + \boldsymbol{\nabla}\cdot\big(\boldsymbol{v}(\mathcal{E} + \tfrac{1}{2}\rho|\boldsymbol{v}|^2)\big) + \boldsymbol{\nabla}\cdot(\boldsymbol{\mathcal{Q}} + \boldsymbol{\mathcal{P}}\cdot\boldsymbol{v}) = 0$$

- **Pressure tensor and heat flux**

$$\boldsymbol{\mathcal{P}} = p\boldsymbol{I} + \sum_{i,j\in\mathfrak{S}} \varkappa_{ij} \boldsymbol{\nabla}\rho_i \otimes \boldsymbol{\nabla}\rho_j - \sum_{i,j\in\mathfrak{S}} \rho_i \boldsymbol{\nabla}\cdot(\varkappa_{ij} \boldsymbol{\nabla}\rho_j) + \boldsymbol{\mathcal{P}}^{\mathrm{d}}$$

$$\boldsymbol{\mathcal{Q}} = \sum_{i,j\in\mathfrak{S}} \varkappa_{ij} \boldsymbol{\nabla}\rho_j \big(\rho_i \boldsymbol{\nabla}\cdot\boldsymbol{v} + \boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{F}}_i - m_i\omega_i\big) - \sum_{i,j\in\mathfrak{S}} \boldsymbol{\nabla}\cdot(\varkappa_{ij} \boldsymbol{\nabla}\rho_j)\boldsymbol{\mathcal{F}}_i + \boldsymbol{\mathcal{Q}}^{\mathrm{d}}$$

High pressure multicomponent fluid models (2)

- **Thermodynamic form for multicomponent fluxes**

$$\boldsymbol{\mathcal{P}}^{\mathrm{d}} = -\,\mathfrak{v}\boldsymbol{\nabla\cdot v}\,\boldsymbol{I} - \eta\big(\boldsymbol{\nabla v} + \boldsymbol{\nabla v}^t - \tfrac{2}{d}\boldsymbol{\nabla\cdot v}\,\boldsymbol{I}\big)$$

$$\boldsymbol{\mathcal{F}}_i = -\sum_{j\in\mathfrak{S}} L_{ij}\Big(\boldsymbol{\nabla}\Big(\frac{g_j}{T}\Big) - \frac{\boldsymbol{\nabla}\,\boldsymbol{\nabla\cdot}(\sum_{l\in\mathfrak{S}} \varkappa_{jl}\boldsymbol{\nabla}\rho_l)}{T}\Big) - L_{i\mathrm{e}}\boldsymbol{\nabla}\Big(\frac{-1}{T}\Big)$$

$$\boldsymbol{\mathcal{Q}}^{\mathrm{d}} = -\sum_{i\in\mathfrak{S}} L_{\mathrm{e}i}\Big(\boldsymbol{\nabla}\Big(\frac{g_i}{T}\Big) - \frac{\boldsymbol{\nabla}\,\boldsymbol{\nabla\cdot}(\sum_{l\in\mathfrak{S}} \varkappa_{il}\boldsymbol{\nabla}\rho_l)}{T}\Big) - L_{\mathrm{ee}}\boldsymbol{\nabla}\Big(\frac{-1}{T}\Big)$$

- **Structure of the matrix L**

$L = (L_{ij})_{i,j\in\mathfrak{S}\cup\{\mathrm{e}\}}$ symmetric positive semi-definite

$N(L) = \mathrm{Span}(1,\ldots,1,0)^t$ Mass conservation constraint $\sum_{i\in\mathfrak{S}} \boldsymbol{\mathcal{F}}_i = 0$

- **Compatibility with thermodynamics**

High pressure multicomponent fluid models (3)

- **Van der Waals type free energy** $\boldsymbol{\mathcal{F} = \mathcal{F}^{\mathrm{cl}} + \frac{1}{2}\sum_{i,j\in\mathfrak{S}} \varkappa_{ij} \nabla\rho_i \cdot \nabla\rho_j}$

$$p = p^{\mathrm{cl}} - \frac{1}{2}\sum_{i,j\in\mathfrak{S}} \varkappa_{ij} \boldsymbol{\nabla}\rho_i \cdot \boldsymbol{\nabla}\rho_j \qquad \mathcal{E} = \mathcal{E}^{\mathrm{cl}} + \frac{1}{2}\sum_{i,j\in\mathfrak{S}} (\varkappa_{ij} - T\partial_T \varkappa_{ij}) \boldsymbol{\nabla}\rho_i \cdot \boldsymbol{\nabla}\rho_j$$

$$\mathcal{S} = \mathcal{S}^{\mathrm{cl}} - \frac{1}{2}\sum_{i,j\in\mathfrak{S}} \partial_T \varkappa_{ij} \boldsymbol{\nabla}\rho_i \cdot \boldsymbol{\nabla}\rho_j \qquad g_i = g_i^{\mathrm{cl}}(\rho_1, \ldots, \rho_{\mathsf{n}_{\mathrm{s}}}, T) \qquad \varkappa_{ij} = \varkappa_{ij}(T)$$

- **Gibbs relation**

$$T\, d\mathcal{S} = d\mathcal{E} - \sum_{i\in\mathfrak{S}} g_i\, d\rho_i - \sum_{i,j\in\mathfrak{S}} \varkappa_{ij} \boldsymbol{\nabla}\rho_i \cdot d\boldsymbol{\nabla}\rho_j$$

- **Simplifying assumptions on capillarities**

$$\varkappa_{ij} = \varkappa(T) \qquad i, j \in \mathfrak{S}$$

High pressure multicomponent fluid models (4)

- **Simplifications using $\sum_{i\in\mathfrak{S}} \boldsymbol{\mathcal{F}}_i = \mathbf{0}$ and $\sum_{i\in\mathfrak{S}} \boldsymbol{m_i\omega_i} = \mathbf{0}$**

$$\sum_{i,j\in\mathfrak{S}} \varkappa_{ij}\boldsymbol{\nabla}\rho_i\otimes\boldsymbol{\nabla}\rho_j = \varkappa\boldsymbol{\nabla}\rho\otimes\boldsymbol{\nabla}\rho \qquad \sum_{i,j\in\mathfrak{S}} \rho_i\boldsymbol{\nabla\cdot}(\varkappa_{ij}\boldsymbol{\nabla}\rho_j) = \rho\boldsymbol{\nabla\cdot}(\varkappa\boldsymbol{\nabla}\rho)$$

$$\sum_{i,j\in\mathfrak{S}} \varkappa_{ij}\rho_i\boldsymbol{\nabla}\rho_j\boldsymbol{\nabla\cdot v} = \rho\boldsymbol{\nabla}\rho\boldsymbol{\nabla\cdot v} \qquad \sum_{i,j\in\mathfrak{S}} \varkappa_{ij}\boldsymbol{\nabla}\rho_j\,\boldsymbol{\nabla\cdot\mathcal{F}}_i = \sum_{i,j\in\mathfrak{S}} \boldsymbol{\nabla\cdot}(\varkappa_{ij}\boldsymbol{\nabla}\rho_j)\boldsymbol{\mathcal{F}}_i = 0$$

$$\sum_{l\in\mathfrak{S}} \varkappa_{il}\boldsymbol{\nabla}\rho_l = \varkappa\boldsymbol{\nabla}\rho \qquad \sum_{i,j\in\mathfrak{S}} \varkappa_{ij}\boldsymbol{\nabla}\rho_i\boldsymbol{\cdot}\boldsymbol{\nabla}\rho_j = \varkappa|\boldsymbol{\nabla}\rho|^2 \qquad \sum_{i,j\in\mathfrak{S}} \varkappa_{ij}\boldsymbol{\nabla}\rho_j m_i\omega_i = 0$$

- **Simplified fluxes**

$$\boldsymbol{\mathcal{P}} = p\boldsymbol{I} + \varkappa\boldsymbol{\nabla}\rho\otimes\boldsymbol{\nabla}\rho - \rho\boldsymbol{\nabla\cdot}(\varkappa\boldsymbol{\nabla}\rho)\boldsymbol{I} + \boldsymbol{\mathcal{P}}^{\mathrm{d}} \qquad \boldsymbol{\mathcal{Q}} = \varkappa\rho\boldsymbol{\nabla}\rho\boldsymbol{\nabla\cdot v} + \boldsymbol{\mathcal{Q}}^{\mathrm{d}}$$

$$\boldsymbol{\mathcal{F}}_i = -\sum_{j\in\mathfrak{S}} L_{ij}\boldsymbol{\nabla}\Big(\frac{g_j}{T}\Big) - L_{ie}\boldsymbol{\nabla}\Big(\frac{-1}{T}\Big) \qquad \boldsymbol{\mathcal{Q}}^{\mathrm{d}} = -\sum_{i\in\mathfrak{S}} L_{\mathrm{e}i}\boldsymbol{\nabla}\Big(\frac{g_i}{T}\Big) - L_{\mathrm{ee}}\boldsymbol{\nabla}\Big(\frac{-1}{T}\Big)$$

High pressure multicomponent fluid models (5)

- **Multicomponent diffuse interface fluid model**

$$\partial_t \rho_i + \boldsymbol{\nabla}\cdot(\rho_i \boldsymbol{v}) + \boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{F}}_i = m_i \omega_i \qquad i \in \mathfrak{S} = \{1,\ldots,\mathsf{n}_{\mathrm{s}}\}$$

$$\partial_t(\rho \boldsymbol{v}) + \boldsymbol{\nabla}\cdot(\rho \boldsymbol{v}\otimes\boldsymbol{v}) + \boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{P}} = 0$$

$$\partial_t\big(\mathcal{E} + \tfrac{1}{2}\rho|\boldsymbol{v}|^2\big) + \boldsymbol{\nabla}\cdot\Big(\boldsymbol{v}\big(\mathcal{E} + \tfrac{1}{2}\rho|\boldsymbol{v}|^2\big)\Big) + \boldsymbol{\nabla}\cdot(\boldsymbol{\mathcal{Q}} + \boldsymbol{\mathcal{P}}\cdot\boldsymbol{v}) = 0$$

- **Multicomponent fluxes**

$$\boldsymbol{\mathcal{P}} = p\boldsymbol{I} + \varkappa\boldsymbol{\nabla}\rho\otimes\boldsymbol{\nabla}\rho - \rho\boldsymbol{\nabla}\cdot(\varkappa\boldsymbol{\nabla}\rho)\boldsymbol{I} - \mathfrak{v}\boldsymbol{\nabla}\cdot\boldsymbol{v}\,\boldsymbol{I} - \eta\big(\boldsymbol{\nabla}\boldsymbol{v} + \boldsymbol{\nabla}\boldsymbol{v}^t - \tfrac{2}{d}\boldsymbol{\nabla}\cdot\boldsymbol{v}\,\boldsymbol{I}\big)$$

$$\boldsymbol{\mathcal{F}}_i = -\sum_{j\in\mathfrak{S}} L_{ij}\boldsymbol{\nabla}\Big(\frac{g_j}{T}\Big) - L_{i\mathrm{e}}\boldsymbol{\nabla}\Big(\frac{-1}{T}\Big) \qquad i \in \mathfrak{S}$$

$$\boldsymbol{\mathcal{Q}} = \varkappa\rho\boldsymbol{\nabla}\rho\boldsymbol{\nabla}\cdot\boldsymbol{v} - \sum_{i\in\mathfrak{S}} L_{\mathrm{e}i}\boldsymbol{\nabla}\Big(\frac{g_i}{T}\Big) - L_{\mathrm{ee}}\boldsymbol{\nabla}\Big(\frac{-1}{T}\Big)$$

The Matrix $\boldsymbol{L}$ (1)

- **Change of variables**

$$(\rho_1,\ldots,\rho_{\mathsf{n_s}},T) \mapsto (\nu,\mathsf{y}_1,\ldots,\mathsf{y}_{\mathsf{n_s}},T) \quad \text{with} \quad \nu = 1/\rho \quad \text{and} \quad \mathsf{y}_i = \rho_i/\rho$$

$$\phi(\rho_1,\ldots,\rho_{\mathsf{n_s}},T) = \phi\big(\tfrac{\mathsf{y}_1}{\nu},\ldots,\tfrac{\mathsf{y}_{\mathsf{n_s}}}{\nu},T\big) \qquad \text{independent mass fractions } \mathsf{y}_i$$

$$(\nu,\mathsf{y}_1,\ldots,\mathsf{y}_{\mathsf{n_s}},T) \mapsto (\nu,\mathsf{x}_1,\ldots,\mathsf{x}_{\mathsf{n_s}},T) \qquad \mathsf{x}_i = \mathsf{y}_i\tfrac{m}{m_i} \qquad \textstyle\sum_{i\in\mathfrak{S}} \tfrac{\mathsf{y}_i}{m_i} = \tfrac{\sum_{i\in\mathfrak{S}} \mathsf{y}_i}{m}$$

- **Pressure based variable**

Mechanically stable states $\partial_\rho p > 0$ or $\partial_\nu p < 0$

$$(\nu,\mathsf{y}_1,\ldots,\mathsf{y}_{\mathsf{n_s}},T) \mapsto (p,\mathsf{y}_1,\ldots,\mathsf{y}_{\mathsf{n_s}},T) \qquad \text{classical variable for diffusion}$$

$$\mathcal{H} = \mathcal{E} + p \qquad h = \mathcal{H}/\rho \qquad h_i = \partial_{\mathsf{y}_i} h(p,\mathsf{y}_1,\ldots,\mathsf{y}_{\mathsf{n_s}},T)$$

- **Classical driving forces**

$$\boldsymbol{d}_i = \mathsf{x}_i \boldsymbol{\nabla}\big(\tfrac{m_i g_i}{RT}\big)_T = \mathsf{x}_i \boldsymbol{\nabla}\big(\tfrac{m_i g_i}{RT}\big) + \tfrac{\mathsf{x}_i m_i h_i}{RT^2}\boldsymbol{\nabla} T$$

The Matrix L (2)

- **Modified matrix $\widehat{L}$**

$$A = \begin{pmatrix} 1 & 0 & \dots & 0 & -h_1 \\ 0 & 1 & \ddots & \vdots & -h_2 \\ \vdots & \ddots & \ddots & 0 & \vdots \\ 0 & \dots & 0 & 1 & -h_{\mathsf{n_s}} \\ 0 & \dots & 0 & 0 & 1 \end{pmatrix} \qquad \widehat{L} = ALA^t$$

- **Identification from low pressure transport fluxes**

$$\widehat{L}_{ij} = \frac{\rho_i \rho_j D_{ij}}{\mathrm{N}R} \qquad \widehat{L}_{ie} = \widehat{L}_{ei} = \frac{\rho_i \theta_i T}{\mathrm{N}R} \qquad \widehat{L}_{ee} = \widehat{\lambda} T^2$$

$$\boldsymbol{\mathcal{F}}_i = -\sum_{j \in \mathfrak{S}} \rho_i D_{ij} \boldsymbol{d}_j - \rho_i \theta_i \boldsymbol{\nabla} \log T \qquad \boldsymbol{\mathcal{Q}} = -\mathrm{N}RT \sum_{i \in \mathfrak{S}} \theta_i \boldsymbol{d}_i - \widehat{\lambda} \boldsymbol{\nabla} T + \sum_{i \in \mathfrak{S}} h_i \boldsymbol{\mathcal{F}}_i$$

The Matrix L (1')

- **Homogeneous thermodynamics**

  New variables $(\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_s}, T)$ with $\nu = 1/\rho$ and $\mathsf{y}_i = \rho_i/\rho$

  $\phi(\rho_1, \dots, \rho_{\mathsf{n}_s}, T) = \phi\Big(\frac{\mathsf{y}_1}{\nu}, \dots, \frac{\mathsf{y}_{\mathsf{n}_s}}{\nu}, T\Big)$

  Energy and entropy densities $e = \mathcal{E}/\rho \quad s = \mathcal{S}/\rho$

  s and e are 1-homogeneous with respect to $(\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_s})$

  p is 0-homogeneous with respect to $(\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_s})$

- **Pressure based thermodynamic functions**

  Mechanically stable states $\partial_\rho p > 0$ or $\partial_\nu p < 0$

  Assume $(\nu, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_s}, T) \mapsto (p, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_s}, T)$ invertible

  Then if $\mathcal{H} = \mathcal{E} + p \quad h = \mathcal{H}/\rho \quad$ we have $\quad h_i = \partial_{\mathsf{y}_i} h(p, \mathsf{y}_1, \dots, \mathsf{y}_{\mathsf{n}_s}, T)$

The Matrix L (2')

- **Modified matrix $\widehat{L}$**

$$A = \begin{pmatrix} 1 & 0 & \dots & 0 & -h_1 \\ 0 & 1 & \ddots & \vdots & -h_2 \\ \vdots & \ddots & \ddots & 0 & \vdots \\ 0 & \dots & 0 & 1 & -h_{\mathsf{n_s}} \\ 0 & \dots & 0 & 0 & 1 \end{pmatrix} \qquad \widehat{L} = ALA^t$$

- **Identification from low pressure transport fluxes**

$$\widehat{L}_{ij} = \frac{\rho_i \rho_j D_{ij}}{\mathrm{N}R} \qquad \widehat{L}_{ie} = \widehat{L}_{ei} = \frac{\rho_i \theta_i T}{\mathrm{N}R} \qquad \widehat{L}_{ee} = \widehat{\lambda} T^2$$

Driving forces $\quad \mathsf{x}_i \boldsymbol{\nabla} \mu_i = \mathsf{x}_i \boldsymbol{\nabla} \frac{m_i g_i}{RT} = \boldsymbol{d}_i - \frac{\mathsf{x}_i m_i h_i}{RT^2} \boldsymbol{\nabla} T \qquad \boldsymbol{d}_i = \mathsf{x}_i (\boldsymbol{\nabla} \mu_i)_T$

$$\boldsymbol{\mathcal{F}}_i = -\sum_{j \in \mathfrak{S}} \rho_i D_{ij} \boldsymbol{d}_j - \rho_i \theta_i \boldsymbol{\nabla} \log T \qquad \boldsymbol{\mathcal{Q}} = -\mathrm{N}RT \sum_{i \in \mathfrak{S}} \theta_i \boldsymbol{d}_i - \widehat{\lambda} \boldsymbol{\nabla} T + \sum_{i \in \mathfrak{S}} h_i \boldsymbol{\mathcal{F}}_i$$

The Matrix L (3')

- **Alternative coefficients**

$$D\chi = \theta \quad < \chi, \mathbb{1} >= 0 \qquad \lambda = \hat{\lambda} - \mathrm{N}R < \theta, \chi > \qquad h'_i = h_i + \chi_i \mathrm{N}R/\rho_i$$

- **The matrix L for stable states**

$$L = \frac{1}{\mathrm{N}R} \begin{pmatrix} \rho_1^2 D_{1,1} & \dots & \rho_1 \rho_{\mathsf{n_s}} D_{1,\mathsf{n_s}} & \sum_{j\in\mathfrak{S}} \rho_1 \rho_j h'_j \\ \vdots & & \vdots & \vdots \\ \rho_{\mathsf{n_s}} \rho_1 D_{\rho_{\mathsf{n_s}},1} & \dots & \rho_{\mathsf{n_s}}^2 D_{\rho_{\mathsf{n_s}},\mathsf{n_s}} & \sum_{j\in\mathfrak{S}} \rho_{\mathsf{n_s}} \rho_j h'_j \\ \sum_{i\in\mathfrak{S}} \rho_1 \rho_i h'_i & \dots & \sum_{i\in\mathfrak{S}} \rho_{\mathsf{n_s}} \rho_i h'_i & \lambda + \sum_{i,j\in\mathfrak{S}} \rho_i \rho_j h'_i h'_j \end{pmatrix}$$