

## A Entropy inequality

In the following, the required equations and derivatives are provided, which are finally inserted into the basic form of the entropy inequality (26). After further transformations the final form of the entropy inequality is obtained, from which the energy conserving and the dissipative restrictions evolve.

- Material time derivative of mass specific Helmholtz energy:

$$\rho^\beta (\psi^\beta)'_\beta = \rho^\beta \left( \frac{\partial \psi^{L\beta}}{\partial \rho^{L\beta}} \right) = \frac{\rho^\beta}{\rho^{L\beta}} (\psi^{L\beta})'_\beta - \frac{\rho^\beta}{(\rho^{L\beta})^2} \psi^{L\beta} (\rho^{L\beta})'_\beta \quad (51)$$

- Mass balance fluid in terms of components:

$$\begin{aligned} \sum_\beta [((\rho^\beta)'_\beta) + \rho^\beta \operatorname{div} \mathbf{x}'_\beta] &= \sum_\beta \hat{\rho}^\beta \\ \Rightarrow \sum_\beta (n^L)'_\beta \rho^{L\beta} + n^L (\rho^{L\beta})'_\beta + n^L \rho^{L\beta} \operatorname{div} \mathbf{x}'_\beta &= \hat{\rho}^L \\ \Rightarrow \sum_\beta w^{L\beta} (n^L)'_\beta + \frac{n^L}{\rho^{LR}} \sum_\beta (\rho^{L\beta})'_\beta + n^L \sum_\beta w^{L\beta} \operatorname{div} \mathbf{x}'_\beta &= \frac{\hat{\rho}^L}{\rho^{LR}} \end{aligned} \quad (52)$$

with

$$\sum_\beta w^{L\beta} (n^L)'_\beta = \underbrace{\sum_\beta w^{L\beta} (n^L)'_I}_{=1} + \operatorname{grad} n^L \sum_\beta w^{L\beta} \cdot \mathbf{w}_{\beta I} \quad (53)$$

(52) yields

$$(n^L)'_I + \operatorname{grad} n^L \sum_\beta w^{L\beta} \cdot \mathbf{w}_{\beta I} + \frac{n^L}{\rho^{LR}} \sum_\beta (\rho^{L\beta})'_\beta + n^L \sum_\beta w^{L\beta} \operatorname{div} \mathbf{x}'_\beta - \frac{\hat{\rho}^L}{\rho^{LR}} = 0. \quad (54)$$

- Material time derivative of saturation condition multiplied with Lagrange multiplier and implementing mass balances of ice and liquid:

$$\begin{aligned} \lambda (1 - n^\alpha)'_I &= 0 \\ \Rightarrow \lambda \left\{ \frac{n^I}{\rho^{IR}} (\rho^{IR})'_I \mathbf{D}_I \cdot \mathbf{I} - \frac{\hat{\rho}^I}{\rho^{IR}} + \operatorname{grad} n^I \sum_\beta w^{L\beta} \cdot \mathbf{w}_{\beta I} + \right. \\ \left. \frac{n^L}{\rho^{LR}} \sum_\beta (\rho^{L\beta})'_\beta + n^L \sum_\beta w^{L\beta} \mathbf{D}_\beta \cdot \mathbf{I} - \frac{\hat{\rho}^L}{\rho^{LR}} \right\} \end{aligned} \quad (55)$$

- Material time derivatives of Helmholtz energy functions:

$$\begin{aligned} \psi^I = \psi^I(\mathbf{C}_I, \theta) : \quad \rho^I (\psi^I)'_I &= 2 \rho^I \mathbf{F}_I \frac{\partial \psi^I}{\partial \mathbf{C}_I} (\mathbf{F}_I)^T \cdot \mathbf{D}_I + \rho^I \frac{\partial \psi^I}{\partial \theta} (\theta)'_I \\ \psi^{L\beta} = \psi^{L\beta}(\rho^{L\beta}, \theta) : \quad (\psi^{L\beta})'_\beta &= \frac{\partial \psi^{L\beta}}{\partial \rho^{L\beta}} (\rho^{L\beta})'_\beta + \frac{\partial \psi^{L\beta}}{\partial \rho^{L\beta}} (\theta)'_\beta \end{aligned} \quad (56)$$

- Implementation and rearranging yields the final representation of the entropy inequality:

$$\begin{aligned} \mathbf{D}_I \cdot \left\{ \mathbf{T}^I + \frac{n^I}{\rho^{IR}} (\rho^{IR})'_I \lambda \mathbf{I} - 2 \rho^I \mathbf{F}_I \frac{\partial \psi^I}{\partial \mathbf{C}_I} (\mathbf{F}_I)^T \right\} - (\theta)'_I \left\{ \rho^I \eta^I + \rho^I \frac{\partial \psi^I}{\partial \theta} \right\} \\ + \sum_\beta \mathbf{D}_\beta \cdot \left\{ \mathbf{T}^\beta + n^L w^{L\beta} \lambda \mathbf{I} \right\} - (\theta)'_\beta \left\{ \rho^\beta \eta^\beta + \rho^\beta \frac{\partial \psi^{L\beta}}{\partial \theta} \right\} \\ - \sum_\beta (\rho^{L\beta})'_\beta \left\{ \frac{\rho^\beta}{\rho^{L\beta}} \frac{\partial \psi^{L\beta}}{\partial \rho^{L\beta}} + \lambda \frac{n^L}{\rho^{LR}} - \frac{\rho^\beta}{(\rho^{L\beta})^2} \psi^{L\beta} \right\} \\ - \hat{\rho}^I \left\{ \psi^I - \psi^L + \lambda \left( \frac{1}{\rho^{IR}} - \frac{1}{\rho^{LR}} \right) \right\} - \sum_\beta \mathbf{w}_{\beta I} \cdot \left\{ w^{L\beta} \lambda \operatorname{grad} n^L + \hat{\mathbf{p}}^\beta \right\} - \frac{1}{\theta} \operatorname{grad} \theta \cdot \mathbf{q} \geq 0. \end{aligned} \quad (57)$$