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Supporting information:

A non-adiabatic model for jacketed agitated batch reactors experiencing thermal losses

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This supporting information document presents a commented derivation of the non-adiabatic heat transfer model for jacketed agitated batch reactors introduced in the main article.

Model derivation

Reactor configuration and governing energy balances

The geometry considered for the purpose of this derivation, includes a jacketed batch reactor with a dished base, the thermal jacket surrounds the entirety of the *wetted* portion of the vessel (above the liquid line on the process side) including the dished base. The reactor has a lid which through which some heat may be lost. A total condenser is installed on the process side to condense vapour from the reactor ullage and maintain a constant volume in

Nomenclature

A heat transfer area E thermal effectiveness E_{mod} modified thermal effectiveness Q quantity of heat transferred T temperature Φ Phi-factor, or thermal inertia α heat transfer film coefficient α_0 free convection film coefficient c_p specific heat capacity t time C_A concentration of reagent A E_a activation energy M mass R gas constant U overall heat transfer coefficient V volume ΔH^{rxn} enthalpy of reaction \dot{M} mass flow \dot{Q} heat flow k_A reaction rate constant for reagent A	u measurement uncertainty x position along the length of the jacket Superscripts \cdot time derivative \sim dimensionless Subscripts 0 initial condition, $t = 0$ 1 inlet 2 outlet ∞ ambient C cooling c condenser H heating j jacket $jloss$ jacket losses p process $ploss$ process losses s source (generation/consumption) set target
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the process. Heat may also be lost from the outer wall of the thermal jacket. While the model presented is relevant to this specific geometry, the following derivation can be easily adapted to consider alternative reactor geometries. A schematic of the reactor geometry, presented in the main article, is repeated in Figure 1 for reference.

Figure 1 identifies six rates of heat transfer for this reactor configuration necessary to fully characterize the system. Identities for each of these terms are defined in eq. (1):

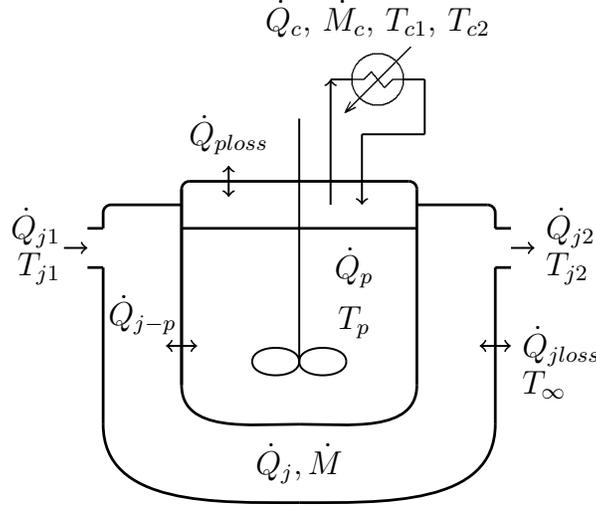


Figure S1: Schematic of heat transfer processes in a non-adiabatic jacketed batch reactor experiencing thermal losses to its surroundings and with a total condenser on the process-side.

$$\begin{aligned}
 \dot{Q}_p &= \sum_{n=1}^N (M c_p)_n \frac{dT_p}{dt} \\
 \dot{Q}_j &= \int_0^L (\dot{M} c_p)_j \frac{dT_j(x)}{dx} dx \\
 \dot{Q}_{j-p} &= \int_0^L (UA')_p (T_j(x) - T_p) dx \\
 \dot{Q}_{jloss} &= \int_0^L (UA')_{jloss} (T_j(x) - T_\infty) dx \\
 \dot{Q}_{ploss} &= (UA)_{ploss} (T_p - T_\infty) \\
 \dot{Q}_c &= (\dot{M} c_p)_c (T_{c1} - T_{c2})
 \end{aligned} \tag{1}$$

The (UA) terms represent products of overall heat transfer coefficients and heat transfer areas (OHTCA), while (UA') terms represent the OHTCAs expressed per unit length of the jacket. These (UA') terms are fundamental to the derivation but will be eliminated from the ultimate ordinary differential equation (ODE) model.

The signs of each rate of heat transfer are significant to the governing energy balance and require clarification. From the identities in eq. (1), in the case that $T_j(x) > T_p > T_\infty$ (i.e. heat is transferred from the jacket to the process and heat is lost from both the jacket

and the process), all of the rates of heat transfer will be positive except for the jacket duty which will be negative, since the jacket temperature will diminish along the length of the jacket ($T_{j1} > T_{j2}$).

Two governing heat balances can be defined in terms of these rates of heat transfer. First, all heat transferred from the jacket is either transferred across the jacket-process wall, or across the outer jacket wall and lost to the surroundings. This *jacket-side* energy balance is given in eq. (2):

$$-\dot{Q}_j = \dot{Q}_{j-p} + \dot{Q}_{jloss} \quad (2)$$

Second, the combined rates of heat transfer across the wall from the jacket to the process and heat generation within the process must balance the sum of (1) heat accumulation in the process, (2) thermal losses from the process to the surroundings, and (3) heat removal from the process by the condenser. This *process-side* energy balance is defined in eq. (3):

$$\dot{Q}_{j-p} + \dot{Q}_s = \dot{Q}_p + \dot{Q}_{ploss} + \dot{Q}_c \quad (3)$$

The rate of heat generation in the process, \dot{Q}_s , may include heats of crystallisation, reaction, mixing, and the mechanical heat introduced by the impeller and will be discussed further following the model derivation. For the heat transfer investigation in the main article, an inert fluid was used on the process side and the mechanical input from the impeller was considered negligible, thereby eliminating the heat generation term, $\dot{Q}_s \rightarrow 0$.

Expanding the jacket-side energy balance

Substituting the relevant identities from eq. (1) into the jacket-side energy balance, eq. (2), generates the integral expression in eq. (4):

$$-\int_0^L (\dot{M}c_p)_j \frac{dT_j}{dx} dx = \int_0^L [(UA')_p(T_j(x) - T_p) + (UA')_{jloss}(T_j(x) - T_\infty)] dx \quad (4)$$

Rearranging and factorizing the jacket temperature terms on the right hand side enables eq. (4) to be expressed as follows:

$$\int_0^L \frac{dT_j}{dx} dx = \int_0^L [-aT_j(x) + b] dx \quad (5)$$

where $a = \frac{(UA')_p + (UA')_{jloss}}{(\dot{M}c_p)_j}$ and $b = \frac{(UA')_p T_p + (UA')_{jloss} T_\infty}{(\dot{M}c_p)_j}$. Integrating eq. (5) to find the jacket temperature at position x along the jacket, $T_j(x)$, yields eq. (6):

$$T_j(x) = \frac{b}{a} + \left(T_{j1} - \frac{b}{a}\right) \exp(-ax) \quad (6)$$

$$\begin{cases} T_j(0) = T_{j1} \\ T_j(L) = T_{j2} \end{cases}$$

Applying the boundary conditions reveals the following expression for the temperature drop along the length of the jacket:

$$T_{j1} - T_{j2} = \left(T_{j1} - \frac{b}{a}\right) (1 - \exp(-aL)) \quad (7)$$

$$T_{j1} - T_{j2} = \left(\frac{(T_{j1} - T_p) + (T_{j1} - T_\infty) \frac{(UA)_{jloss}}{(UA)_p}}{1 + \frac{(UA)_{jloss}}{(UA)_p}} \right) \left(1 - \exp \left(-\frac{(UA)_p + (UA)_{jloss}}{(\dot{M}c_p)_j} \right) \right)$$

Two dimensionless parameters revealed in eq. (7) recur frequently throughout the derivation. The first is a dimensionless OHTCA, representing the ratio of jacket loss and process OHTCAs, $(\tilde{U}A) = \frac{(UA)_{jloss}}{(UA)_p}$. The second is the complement to the exponential term, which resembles the thermal effectiveness,^{1,2} $E = 1 - \exp \left(-\frac{(UA)_p}{(\dot{M}c_p)_j} \right)$, but includes the additional jacket loss OHTCA. This parameter will be labeled the *modified thermal effectiveness*,

$E_{mod} = 1 - \exp\left(-\frac{(UA)_p + (UA)_{jloss}}{(\dot{M}c_p)_j}\right)$. For the case of constant flow rate in the jacket, and assuming the physical properties of the fluids do not evolve significantly with time, these two dimensionless parameters can be considered constant, but could equally be expressed as functions of temperature or reactant conversion.³ Adopting this nomenclature, eq. (7) reduces to:

$$T_{j1} - T_{j2} = \frac{E_{mod}}{1 + (\tilde{U}A)} \left((1 + (\tilde{U}A))T_{j1} - T_p - (\tilde{U}A)T_\infty \right) \quad (8)$$

This identity for the temperature drop along the length of the jacket enables the characterization of the rate of heat transfer from the the jacket:

$$-\dot{Q}_j = k \left((1 + (\tilde{U}A))T_{j1} - T_p - (\tilde{U}A)T_\infty \right) \quad (9)$$

where $k = \frac{(\dot{M}c_p)_j E_{mod}}{1 + (\tilde{U}A)}$. A similar procedure is repeated to define the rate of heat transfer across the jacket-process wall. The jacket-side energy balance rearranges to give:

$$\begin{aligned} \dot{Q}_{j-p} &= -\dot{Q}_j - \dot{Q}_{jloss} \\ \dot{Q}_{j-p} &= (\dot{M}c_p)_j(T_{j1} - T_{j2}) - \int_0^L (UA')_{jloss} (T_j(x) - T_\infty) dx \end{aligned} \quad (10)$$

Substituting the local jacket temperature, $T_j(x)$, expression from eq. (6) yields:

$$\dot{Q}_{j-p} = (\dot{M}c_p)_j(T_{j1} - T_{j2}) - \int_0^L (UA')_{jloss} \left(\frac{b}{a} + \left(T_{j1} - \frac{b}{a} \right) \exp(-ax) - T_\infty \right) dx \quad (11)$$

Solving the integral on the right hand side gives:

$$\begin{aligned} \dot{Q}_{j-p} &= (\dot{M}c_p)_j(T_{j1} - T_{j2}) + (UA)_{jloss} \left(T_\infty - \frac{b}{a} \right) \\ &\quad - \frac{(UA')_{jloss}}{a} \left(T_{j1} - \frac{b}{a} \right) (1 - \exp(-aL)) \end{aligned} \quad (12)$$

The product of the final two bracketed terms is recognisable as the temperature drop along the length of the jacket, previously stated in eq. (7). Factorizing all $T_{j1} - T_{j2}$ terms

gives:

$$\dot{Q}_{j-p} = \left((\dot{M}c_p)_j - \frac{(UA)_{jloss}}{a} \right) (T_{j1} - T_{j2}) + (UA)_{jloss} \left(T_\infty - \frac{b}{a} \right) \quad (13)$$

Substituting for the a and b terms provides the following expression for the rate of heat transfer from the jacket to the process:

$$\dot{Q}_{j-p} = \frac{(UA)_{jloss}(T_\infty - T_p) + (\dot{M}c_p)_j(T_{j1} - T_{j2})}{1 + (\tilde{U}A)} \quad (14)$$

Since the temperature drop along the length of the jacket has been defined in eq. (8), the jacket outlet temperature can be eliminated from the model. The equation for the rate of heat transfer across the jacket-process wall is then factorized in terms of the three key temperatures, T_{j1} , T_p and T_∞ for readability:

$$\dot{Q}_{j-p} = kT_{j1} - \frac{(UA)_{jloss} + k}{(1 + (\tilde{U}A))} T_p - \frac{k(UA) - \tilde{(UA)}_{jloss}}{(1 + (\tilde{U}A))} T_\infty \quad (15)$$

The final rate of heat transfer relevant to the jacket-side balance requiring definition is the rate of heat loss from the outer jacket wall. This can be obtained either from the sum of the two rates of heat transfer previously defined, $\dot{Q}_{jloss} = -(\dot{Q}_j + \dot{Q}_{j-p})$, or by repeating the integration procedure for the identity of the thermal losses from the jacket defined in eq. (1). The resulting expression is given in eq. (16):

$$\dot{Q}_{jloss} = k(\tilde{U}A)T_{j1} + \frac{(UA)_{jloss} - k(\tilde{U}A)}{1 + (\tilde{U}A)} T_p - \frac{k(\tilde{U}A)^2 + (UA)_{jloss}}{1 + (\tilde{U}A)} T_\infty \quad (16)$$

Expanding the process-side energy balance

Having defined all the rates of heat transfer relevant to the jacket-side energy balance, the relevant heat transfer identities can be substituted into the process-side energy balance to solve for the rate of heat accumulation in the process, $\dot{Q}_p = \dot{Q}_{j-p} + \dot{Q}_s - \dot{Q}_{ploss} - \dot{Q}_c$.

$$\begin{aligned}
\sum_{n=1}^N (M c_p)_n \frac{dT_p}{dt} = & k T_{j1} - \\
& \left(\frac{k + (UA)_{jloss}}{(1 + (\tilde{U}A))} + (UA)_{ploss} \right) T_p + \\
& \left(\frac{(UA)_{jloss} - (\tilde{U}A)k}{(1 + (\tilde{U}A))} + (UA)_{ploss} \right) T_\infty - \\
& (\dot{M}c_p)_c (T_{c1} - T_{c2}) + \dot{Q}_s
\end{aligned} \tag{17}$$

This ODE governs the evolution of process temperature. While incorporating the thermal losses from the outer jacket wall complicates the derivation of the model, and requires an additional OHTCA to be characterized, solving the ODE numerically is no more computationally expensive than the classical adiabatic models. For constant flow rates in the jacket and condenser, and assuming no significant evolution in the thermophysical properties with time, each of the temperature multipliers remain constant with time. Furthermore, the model simplifies with various commonly applied assumptions. For the case of (1) no heat generation within the process, $\dot{Q}_s \rightarrow 0$, (2) no condenser on the process side, $\dot{M}_c \rightarrow 0$, and (3) a perfectly insulated outer jacket wall, $(UA)_{jloss} \rightarrow 0$, the model reduces to the two-coefficient *semi-adiabatic* model presented in a previous article by the current authors:²

$$\begin{aligned}
\sum_{n=1}^N (M c_p)_n \frac{dT_p}{dt} = & (\dot{M}c_p)_j E T_{j1} - \\
& \left((\dot{M}c_p)_j E + (UA)_{ploss} \right) T_p + \\
& (UA)_{ploss} T_\infty
\end{aligned} \tag{18}$$

The identities and correlations for each of the rates of heat transfer identified in Figure 1 are summarized in Table 1.

Heat generation in the process

Heats of reaction, mixing, phase change or mechanical agitation may be significant during standard reactor operation. Reactions are particularly significant due to the well established exponential dependence of reaction rate on process temperature from the Arrhenius equation. Using the example of a first order liquid phase reaction, the rate of heat generation can be calculated as follows:

$$\dot{Q}_s = V_p k_A C_A \exp\left(-\frac{E_a}{RT_p}\right) \Delta H^{rxn} \quad (19)$$

The introduction of an exponential function of process temperature into the ODE indicates that numerical errors arising during the solution of eq. (17) are likely to propagate more quickly, especially when coupled with expressions for the evolution of concentration with time and the evolution of thermophysical properties with conversion.³ It may become necessary to use a higher order numerical method and/or a shorter time-step to perform the numerical integration. A similar numerical sensitivity will likely be observed during crystallisation due to the power law dependencies of crystal nucleation and growth on supersaturation.

Table S1: Correlations for the various rates of heat transfer identified in Figure 1

Property	Identity	Equation
\dot{Q}_j	$\int_0^L (\dot{M}c_p)_j \frac{dT_j(x)}{dx} dx$	$-k \left((1 + (\tilde{U}A))T_{j1} - T_p - (\tilde{U}A)T_\infty \right)$
\dot{Q}_{j-p}	$\int_0^L (UA')_p (T_j(x) - T_p) dx$	$kT_{j1} - \frac{(UA)_{jloss} + k}{(1 + (\tilde{U}A))} T_p - \frac{k(UA) - (\tilde{U}A)_{jloss}}{(1 + (\tilde{U}A))} T_\infty$
\dot{Q}_{jloss}	$\int_0^L (UA')_{jloss} (T_j(x) - T_\infty) dx$	$k(\tilde{U}A)T_{j1} + \frac{(UA)_{jloss} - k(\tilde{U}A)}{1 + (\tilde{U}A)} T_p - \frac{k(\tilde{U}A)^2 + (UA)_{jloss}}{1 + (\tilde{U}A)} T_\infty$
\dot{Q}_p	$\sum_{n=1}^N (\dot{M}c_p)_n \frac{dT_p}{dt}$	$kT_{j1} - \left(\frac{k + (UA)_{jloss}}{(1 + (\tilde{U}A))} + (UA)_{ploss} \right) T_p +$ $\left(\frac{(UA)_{jloss} - (\tilde{U}A)k}{(1 + (\tilde{U}A))} + (UA)_{ploss} \right) T_\infty -$ $(\dot{M}c_p)_c (T_{c1} - T_{c2}) + \dot{Q}_s$
\dot{Q}_{ploss}	$(UA)_{ploss} (T_p - T_\infty)$	$(UA)_{ploss} (T_p - T_\infty)$
\dot{Q}_c	$(\dot{M}c_p)_c (T_{c2} - T_{c1})$	$(\dot{M}c_p)_c (T_{c2} - T_{c1})$

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