SIMPLIFIED HEAT TRANSFER MODEL FOR BUFFER STORAGE OF HIGHLY ACTIVE RAFFINATE ON THE SELLAFIELD SITE

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1. INTRODUCTION

High level waste (HLW) management refers to the treatment of a toxic, radioactive effluent stream generated during the reprocessing of spent fuel from nuclear reactors. After a review of relevant literature [1], challenges have been identified regarding this research area, because there is a demand for improved data from, and simulation of, existing equipment that impinge on the safety of HLW treatment plants. These data and simulations can improve the design of future reprocessing plants. In contrast to established simulations of evaporation operations on the Sellafield site, none exist for the buffer storage plant. The general approach to simulation involves the generation of complex CFD models, which require expensive computational power and for long simulation times. By contrast, there is a need for the development of relatively simplified and accurate models requiring little computational power. This discourse research presents a heat transfer model that provides the prediction of the temperature response of the contents of buffer storage tanks (BSTs) containing highly active raffinate (HAR) at Sellafield and does not rely on complex CFD simulations.

The buffer storage tank (BST) plant was originally built for the purposes of in-tank evaporation of fission product raffinate from the original reprocessing plant at Sellafield. There are six 1200 m^3 BSTs, each weigh approximately 52 tonne empty and a full batch of HAL increases this by approximately 1200 tonne, and all were fitted with two steam coils for in-tank evaporation. Evaporation procedures are no longer carried out and these coils have since been converted to cooling coils with an additional helical cooling coil traversing the tank periphery. During times of high demand of HAR evaporative capacity, the BSTs currently act as buffer storage and are each located inside a sealed thick-walled concrete cell to shield the radioactive contents.

The HAR is an effluent stream consisting of concentrated acid with dissolved fission products and has self-heating properties. The basis for safe operation of the BSTs is to control the temperature of the contents below the bubble point of 95^{o}C to avoid potential release of toxic vapours into the atmosphere. The BSTs at Sellafield currently fulfill a purpose different to the intended design. Previous plant data from the BSTs is not useful for the new operational use. Presently it is assumed the BSTs operate adiabatically and so the prediction of a temperature response is considerably faster than that experienced on the plant. Hence there is an urgent requirement to improve the current predictions by using a simplified and relatively accurate heat transfer model for the prediction of the temperature response of the BSTs. This will allow more efficient operation of the buffer storage plant and save operations staff time and resources.

2. THEORY AND MODEL DEVELOPMENT

The model presented herein has been derived from first principles by performing an energy balance across a single BST, and the following assumptions apply: 1-dimensional conduction heat flow; the thermophysical properties are independent of temperature; the HAR is well mixed and so the temperature is the same everywhere in the vessel; the vapour in the ullage region is in thermal equilibrium with the HAR; the heat loss from inside the ullage region is caused by condensation of water, which flows back into the HAR and is equal to the heat lost from evaporation of water from the HAR surface; the cell vent flow is very low and results in a large residence time and the temperature is uniform throughout; the concrete cell wall acts as a perfect insulator and the temperature of its inner

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surface is 1°C less than the cell vent value. Figure 1 is a schematic of the various heat flows occurring within the BST.

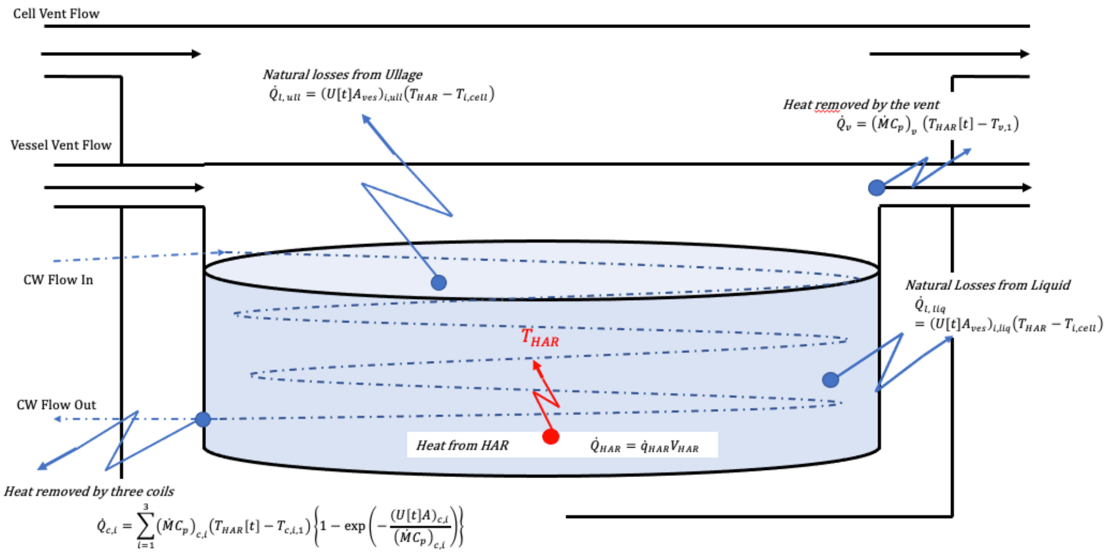

Figure 1. Schematic highlighting the contributing terms to the energy balance over a single BST.

Heat is generated within the BST $\dot{Q}_{HAR}$ and occurs due to the presence of decaying radioactive isotopes dissolved in the HAR mixture. Within the industry this heat is characterized by assigning the material with a heat rating value that depends on the origin of and the type of isotopes in the HAR, and the age of the liquor. This rating $\dot{q}_{HAR}$ is a specific quantity of heat generated per unit volume and, for standard HAR reprocessed from THORP, the heat rating is $235\ \mathrm{W/m^3}$ [2]. Hence the time variation of the temperature of the HAR depends upon the heat generated from the volume of HAR V_{HAR} deposited in the BST, the thermal inertia of the quantity of HAR in the vessel and that of the vessel and coils $\left[(\rho V C_p)_{HAR} + (\rho V C_p)_{steel}\right]$, and the various heat losses from the vessel contents to the vent gas flow through the concrete cell containing the BST.

The following expression represents the response of the HAR temperature $T_{HAR}[t]$ within the vessel due to the heat evolved $\dot{q}_{HAR}V_{HAR}$ minus the summation of the enthalpy losses depicted in figure 1:

$$\left[(\rho V C_p)_{HAR} + (\rho V C_p)_{steel}\right]\frac{\mathrm{d}T_{HAR}[t]}{\mathrm{d}t} = \dot{q}_{HAR}V_{HAR} - \sum_{i=1}^{3}(\dot{M}C_p)_{c,i}(T_{HAR}[t] - T_{c,i,1})\,E_{c,i}[t]$$
$$-(\dot{M}C_p)_v(T_{HAR}[t] - T_{v,1}) - \left((U[t]A_{ves})_{i,liq} + (U[t]A_{ves})_{i,ull}\right)(T_{HAR} - T_{i,cell}) \quad (1)$$

where in equation (1), the volume of HAR is V_{HAR} and $E_{c,i}[t]$ are the thermal effectiveness values obtained from the relationship $E_{c,i}[t] = 1 - \exp\left(-(U[t]A/\dot{M}C_p)_{c,i}\right)$, $U_{c,i}[t]$ are the OHTCs, $A_{c,i}$ are inner surface areas of the three cooling coils, and $\acute{M}_{c,i}$ are the flows and $C_{p,c,i}$ are the specific heats of the cooling water respectively. The last two terms on the right-hand side of the equality sign are the heat losses by the vent gas flow and those combined from the vessel liquor and ullage volumes to the cell vent gas respectively. Note, all of the OHTCs in equation (1) vary with time dependence due to various individual film heat transfer coefficients being dependent upon non-linear temperature differences, which cause equation (1) to become non-linear overall with respect to the temperature $T_{HAR}[t]$. In addition, this expression includes the impact of the thermal inertia of the steel contents of the BST; see the work of Johnson *et al.* [3].

The largest contribution to heat losses is due to the 3 cooling coils in the vessel $\dot{Q}_{c,i}$ and is around 85% of the total. The OHTCs $U[t]_{c,i}$ of the coiling coils based on the outside surface area of each coil $(2\pi d_{O,c,i}L_{c,i})$ are obtained from following expression:

$$\frac{1}{U[t]_{\mathrm{O},c,i}} = \frac{1}{h[t]_{\mathrm{O},c,i}} + \frac{d_{\mathrm{O},c,i} ln\left\{\frac{d_{\mathrm{O},c,i}}{d_{\mathrm{I},c,i}}\right\}}{2k_{w,c,i}} + \frac{d_{\mathrm{O},c,i}}{d_{\mathrm{I},c,i}h_{\mathrm{I},c,i}} \tag{2}$$

The time dependency emanates from the outside film heat transfer coefficient $h[t]_{\mathrm{O},c,i}$ of the coils in equation (2), because the value at any point in time depends upon the difference between the HAR $T_{HAR}[t]$ and the outside coil surface $T_{\mathrm{O},c,i}$ temperatures. This coefficient is calculated from the turbulent free convection correlation provided by equation (3) as follows:

$$h[t]_{\mathrm{O},c,i} = 0.1 \times (\mathrm{GrPr})^{\frac{1}{3}} = 0.1 \times \left(g\beta\left(T_{HAR}[t] - T_{\mathrm{O},c,i}\right)/(\nu\alpha)^2\right)^{1/3} \tag{3}$$

The other two terms in equation (2) represent the conduction across the wall of the coil and the forced convection coefficient $h_{\mathrm{I},c,i}$ of the cooling water flow within the coil.

The BST also suffers natural heat losses to the surroundings from the HAR in the vessel and the ullage region above the HAR. This means other expressions for the OHTC are required, which are far more complex due to the multiple heat transfer mechanisms occurring both in parallel and series. These are highlighted by the resistance network in figure 2.

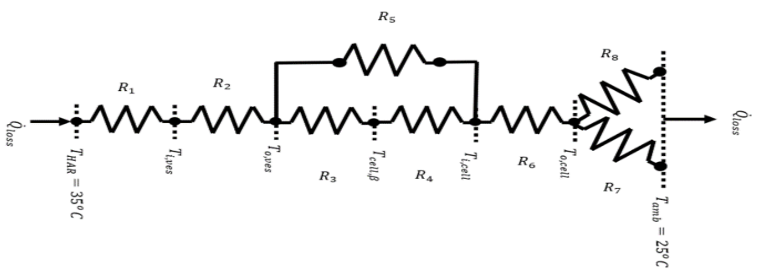

Figure 2. Resistance network for the heat losses from the BST

The heat transfer occurs by free convection currents within the HAR, but for the ullage region it occurs by dropwise condensation of the vapours from the vent gases, which are saturated at the same temperature as the HAR. The amount of condensation will be equivalent to the evaporation from the HAR surface. In both cases, the heat flow is then by conduction through the wall of the BST and then from the outside wall by free convection to the cell vent gas, and also, by radiation to the inside cell wall. Then there is next a further transfer of heat by free convection at the inside surface of the cell from the bulk cell vent gas. The thick concrete cell wall provides additional resistance to heat flow. Following this, heat flows from the outside surface of the cell by free convection and radiation in parallel to the ambient surroundings. However, the combined resistance network across the cell wall to the ambient surroundings is completely dominated by the conduction resistance of the concrete walls of the cell, which are 3 metres thick. Hence the cell can be considered to act adiabatically, that is a perfect insulating material. Hence the overall resistance network illustrated in figure 2 is only evaluated up to the inner cell wall temperature $T_{\mathrm{i,cell}}$ and this is taken to be 1°C less than the vent gas temperature $T_{\mathrm{cell,g}}$.

The OHTC $U_{i,liq}[t]$ in equation (1) is based on the inside surface area $A_{i,liq,ves}$ corresponding to the depth of HAR liquor in the vessel is evaluated by the expression detailed in equation (4), which contains individual film coefficients for free convection and radiation. The free convection film coefficient inside the vessel $h_{i,ves}$ is dependent upon the temperature difference between the HAR liquor and the inner vessel surface, and the free convection coefficients $h_{o,ves}$ and $h_{i,cell}$ depend upon differences between the outer vessel surface and the vent gas temperatures, and between the vent gas and the inner cell surface temperatures respectively. These coefficients are obtained from similar correlations to that in equation (3).

$$\frac{1}{U_{\mathrm{i},liq}} = \frac{1}{h_{i,ves}} + \frac{d_{\mathrm{i},ves} ln\left\{\frac{d_{o,ves}}{d_{i,ves}}\right\}}{2\pi k_{w,ves}} + \frac{A_{i,liq,ves}}{\left[\left(h_{i,cell}A_{i,cell}\right) + \frac{1}{\left(\frac{1}{h_{o,ves}A_{o,ves}} + \frac{1}{h_{o,ves,r}A_{o,ves}}\right)}\right]} \tag{4}$$

Finally, the radiative coefficient $h_{o,ves,r}$ depends on the difference between the outer vessel and the inner cell wall absolute temperatures defined by equation (5), where $\Im_{o,ves \to i,cell}$is the gray body shape factor between the outer vessel and the inner cell surfaces.

$$h_{o,ves,r} = \Im_{o,ves \to i,cell} \sigma\left(T_{o,ves}^2 + T_{i,cell}^2\right) \times \left(T_{o,ves} + T_{i,cell}\right) \tag{5}$$

The OHTC $U_{i,ull}[t]$ in equation (1) is based upon the inside surface area of the ullage region $A_{i,ull,ves}$ and is obtained from a similar expression to equation (4), but now the inside coefficient is caused by dropwise condensation and this depends upon the temperature difference between the saturated gas in the ullage volume and the inner vessel surface.

The time variations of the values of $U_{i,liq}[t]$ and $U_{i,ull}[t]$ emanate from the varying HAR temperatures, which causes all the film coefficients and all surface temperatures to also vary with time. Hence the evaluation of all these coefficients requires solution to sets of simultaneous non-linear algebraic equations representing the heat flow from the vessel to the cooling coils and the cell vent gases. The residence time of the vent gases in the cell is a matter of hours and so it is assumed that the cell temperature is uniform throughout the cell.

A numerical solution of equation (1) provides a prediction for the temperature response of the contents of the BST. Having established an appropriate time interval, it is possible to numerically determine the change in the temperature of the BST contents using Euler's approximation formula (6).

$$T_{HAR}[t_{i+1}] = T_{HAR}[t_i] + \frac{\mathrm{d}T_{HAR}}{\mathrm{d}t} \times \Delta t \tag{6}$$

The temperature profile generated from the Euler approximation is first order accurate with the size of the step. In order to be confident of convergence, a predictor-corrector numerical solution procedure is also used; the trapezoidal formulation (7). Provided the results from Euler's approximation are within tolerance of those generated from the trapezoidal predictor technique one can claim convergence.

$$T_{HAR}[t_{i+1}] = T_{HAR}[t_i] + \left(\left.\frac{\mathrm{d}T_{HAR}}{\mathrm{d}t}\right|_i + \left.\frac{\mathrm{d}T_{HAR}}{\mathrm{d}t}\right|_{i+1}\right) \times \frac{\Delta t}{2} \tag{7}$$

Both numerical solutions require iterations for each time step due the evaluations of the OHTCs and the various film heat transfer coefficients. These sets of equations are re-arranged by the precedence ordering technique in order to facilitate robust solution algorithms for each OHTC [4]. The predictions by both techniques have been developed as a VBA code for solution in Excel.

3. RESULTS AND DISCUSSION

Figure 3 (a) presents the very similar temperature response predictions from simulating a full tank of HAR rated at 235 W/m^3 by both Euler and trapezoidal predictor-corrector formulae. The tank's contents reach a temperature of approximately 70ºC over the course of 30 days. Fig 3 (b) presents very reasonable predictions of recorded data from a BST containing 810 m^3 of HAR material with heat rating at 70 W/m^3 and with an equilibrium temperature of 23.4ºC compared with that on plant of 22.5ºC. This overprediction provides an acceptable conservative projection, without the recourse of adopting expensive CFD simulations. Fig 3 (c) provides the temperature response predictions for varying volumes of HAR with loss of the cooling water supply for a HAR rating of 235 W/m^3. A HAR volume of approximately 200 m^3 will barely the change temperature. However, with increasing volumes higher equilibrium temperatures occur and the highest is around 87ºC for a volume of 1171 m^3.

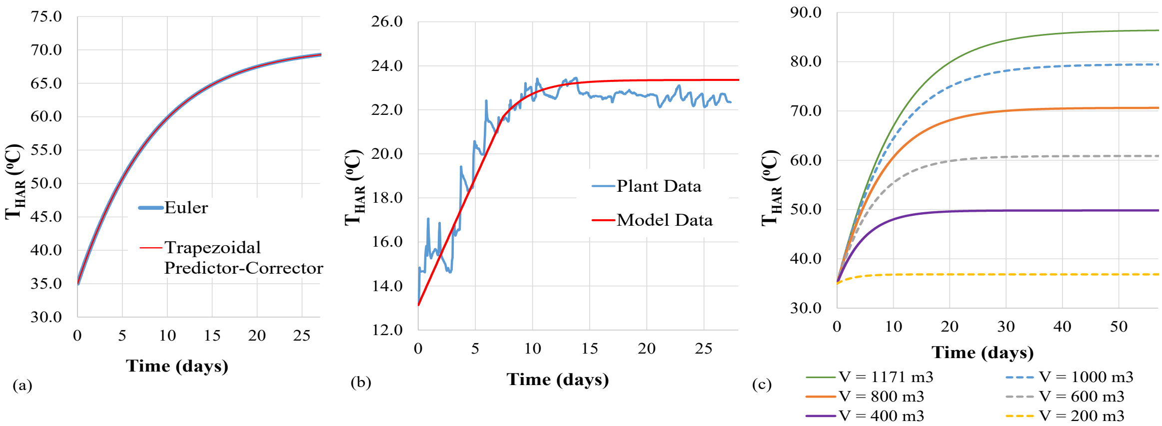

Figure 3. (a) Temperature-time profiles for a full tank of HAR rated at 235 W/m^3 by both solutions at a Δt of $5000\ s$. (b) Temperature-time profiles from measured plant data and model predictions. (c) Temperature-time predicted profiles for varying HAR volumes and loss of cooling water flow at a heat rating of 235 W/m^3.

The current approach to operations on the Sellafield site is to use the zero heat loss (ZHL) method, which results in a linear temperature-time relationship [2]. Fig 4 (a) is a plot of temperature responses by the ZHL and model predictions for a full tank of HAR rated at 235 W/m^3 with the cooling water active at 0.5 m^3/h per coil and the vessel and cell vent flows active. The linearity of the ZHL relationship diminishes when allowing for heat losses and reveals the significant conservation associated with the ZHL assumption. The bubble point of 95ºC is reached after approximately 11 days by the ZHL approach whereas the model prediction reaches an equilibrium temperature of 70.4ºC after approximately 35 days. A further operational parameter, the ejector strike temperature[1] of 67ºC, is predicted by the ZHL method to be reached in approximately 6 days, but the proposed model predicts almost double this time (approximately 15 days). Also plotted in fig 4 (a) are the predictions of the cascade of temperatures through the BST system with time: the outside surface of the vessel, the bulk cell and the inside of the cell temperatures. These data cannot be predicted by the fundamental and simple ZHL approach.

Fig 4 (b) is a plot of temperatures for a full BST of 1171 m^3 of HAR, a heating rate of 3500 W/m^3 a cooling water flow of 0.5 m^3/h per coil and both vent and cell flows activated. At this higher heat loading, the temperature-time plot predicted by the proposed model exhibits linearity and follows closely the ZHL profile. The similarity in predictions is caused by the heat released by the HAR being orders of magnitude larger than the amount removed by the various heat losses. At lower heat loadings, the difference in magnitude between the heat generated by the HAR and the heat removal is not so large and the ZHL is no longer valid. In summary it becomes clear that, at low to medium heat loadings of HAR, the proposed model provides a useful predictor of the temperature response behaviour of the contents of the BST. Despite this, the ZHL approach fails to provide details of intermediate temperatures

[1]The ejector strike temperature is the maximum temperature the motive fluid (steam) can reach in an ejector device. If steam exceeds this temperature this causes low motive flow and a phenomenon known as breaking the shockwave (striking) occurs. This deteriorates the ejector performance and can lead to wastage of steam and reduction in operation efficiency.

as well as Q-values and overall heat transfer coefficients which can be extracted from the output of the proposed model.

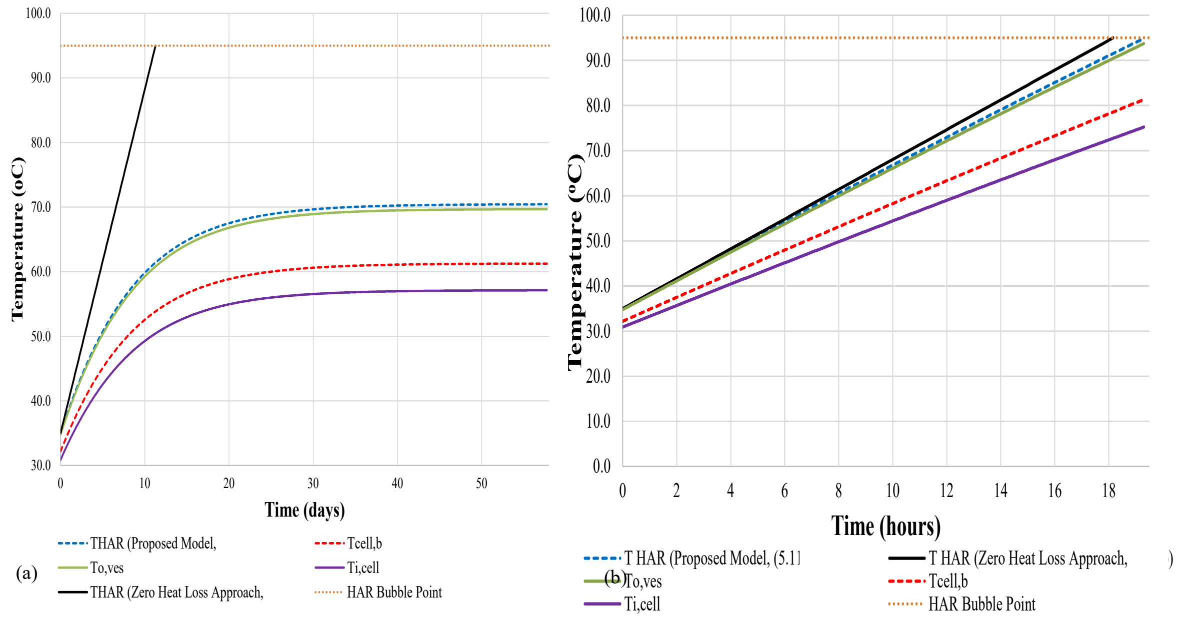

Figure 4 (a) Temperature-time plots predicted by the proposed model and the ZHL approach used in current operations. b) Plots predicted at higher heat loading by both ZHL and the model.

4. CONCLUSIONS

A simplified heat transfer model is presented that predicts the temperature response of the contents of BSTs containing HAR at Sellafield. It was developed by an energy balance around a single BST. Expressions for the non-linear overall heat transfer coefficients were derived and solved using precedence ordering techniques at each time interval [4]. The solutions presented herein have been generated by numerical procedures with demonstrated convergence. In addition, the solutions have been demonstrated to show strong agreement with measurements on plant. The results from the model highlights that, in certain scenarios, the plant can safely store without reaching the ejector strike temperature for approximately 67 % longer than is predicted by current practice (ignoring heat loss).

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