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Relief vent sizing and location for long tubular reactors

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Abstract

The DIERS (Design Institute for Emergency Relief Systems) methodology for emergency relief evaluation is recognized as a state-of-the-art procedure for reacting systems operated in batch or semi-batch mode process vessels. By comparison, the evaluation of emergency relief requirements for tubular reactors has received little consideration. This paper addresses the questions of location and size requirements for long tubular reactors. Much of the DIERS methodology is applicable in principle, and this paper suggests where certain key details may be modified for tubular reactor considerations. This paper presents emergency relief evaluation procedures for both tempered and pure gassy reacting systems and provides example illustrations of each case.

1. Introduction

Tubular reactors (sometimes referred to as plug flow reactors) are geometrically quite different from batch reactors. For the purpose of this paper we will consider a typical tubular reactor as a continuous flow reactor system made up of a relatively small diameter pipe but having a very long length-to-diameter ratio (L/D), perhaps ranging from several hundred to several thousand. Such reactors are rarely straight, and often consist of a sequence of straight segments and reversing "hair-pin" turns enclosed in heat exchange or temperature control shells.

Upset conditions which lead to requirements for pressure relief may include any of the following considerations:
(a) Loss of flow and temperature control
(b) Reagent feed error
(c) External fire
(d) Other site specific considerations

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As is often the case for a batch reactor process, the necessary information required to characterize the run-away reaction for emergency relief evaluation will require empirical bench-scale calorimetry tests. This essential part of the DIERS methodology has been adequately described in References [1–5] for example, and is equally applicable to tubular reactors. Benchscale test data should establish whether the reacting system under consideration is tempered (reaction rate may be limited by evaporation of volatile reagents) or generates non-condensable gas (gassy). In either case, the test data for the specific chemical system should provide the relevant reaction rate data required for vent sizing analysis.

This background information is an essential and well-established part of the DIERS methodology and will be assumed as prerequisite for consideration of tubular reactors. However, use of this data for vent size evaluation has emphasized batch reactors and the purpose at hand is to illustrate how the same methods may be adapted to long tubular reactor devices.

2. Features of tubular reactors

There are several features of tubular reactors that are important to recognize at the outset. The device shown in Fig. 1, which is only one example of the concept, serves to illustrate several features. Relative to batch reactors, the in-reactor inventory is usually quite small. Depending on specific material of construction, the pressure rating is usually quite high owing to the usually small diameter. If this is indeed the case, then relative to a batch reactor, the thermal inertia or so-called phi factor may be significant in slowing the rate of a postulated run-away reaction. This point will be addressed later in the paper.

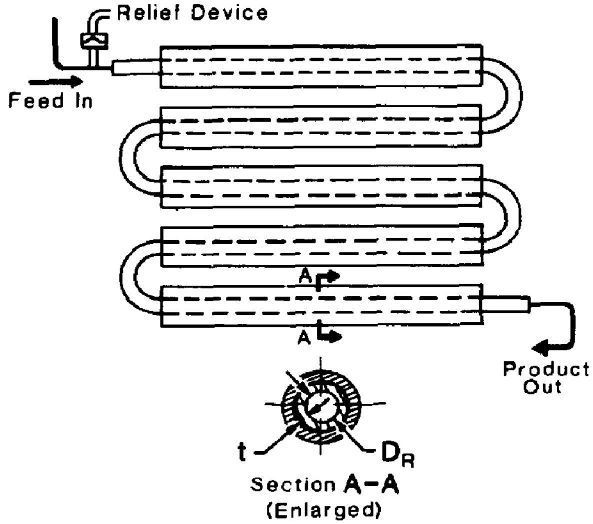

Fig. 1. Illustration of a long tubular reactor.

In spite of the long length of the reactor, locations for emergency relief may be limited due to concentric heat exchange equipment. Therefore, one essential question regarding pressure relief for tubular reactors is the maximum length of reactor which can be protected by a single pressure relief device, when the relief vent area is limited by the cross-section area of the reactor itself.

The last significant characteristic of tubular reactors which will be mentioned here is that the relief vent flow rate will be significantly affected by length of the reactor. Fortunately, prior analysis of the two-phase discharge of long pipes may be utilized to enable the batch reactor vent sizing evaluation models to be adapted to tubular reactors.

3. Discharge rates in long tubes

The two-phase discharge of non-reacting flashing liquids was evaluated in Ref. [6] by means of a detailed numerical model which considered sensitivity to flow regime assumptions as well as numerical nodalization of the pipe line. Pipe lengths of several kilometers were considered. This analysis set the stage for a one-step analytical approximation developed in Ref. [7]. Grolmes and Fauske [7] showed how the long-pipe two-phase discharge coefficient models established in Ref. [8] could be utilized to provide good agreement with detailed numerical models. These methods are adopted here.

A discharge coefficient, or flow reduction factor F, may be defined as

$$F = G/G_{\mathrm{max}} \tag{1}$$

where G is the actual discharge mass flux and G_{max} is the two-phase discharge mass flux for an ideal no-loss geometry. For flashing liquids characteristic of tempered reacting systems, G_{max} may be approximated by [9]:

$$G_{\mathrm{max}} = \frac{\lambda}{v_{\mathrm{fg}}} \frac{1}{\sqrt{CT}} \tag{2a}$$

or the equivalent form when the Clausius–Clapeyron equation holds,

$$G_{\mathrm{max}} = \frac{\mathrm{d}P}{\mathrm{d}T}\sqrt{\frac{T}{C}}. \tag{2b}$$

For non-flashing, two-phase discharge of a gassy system, the more rigorous formulation for G_{max}, found in Ref. [10], may be approximated by

$$G_{\mathrm{max}} = \sqrt{P_0(1-\alpha)\rho_{\mathrm{f}}}\, \frac{\left[\frac{2}{\alpha}\left\{\left(\frac{1-\alpha}{\alpha}\right)(1-\eta) - \ln\eta\right\}\right]^{1/2}}{1/\eta + (1-\alpha)/\alpha} \tag{3}$$

where

$$\eta = [2.016 + [(1-\alpha)/2\alpha]^{0.7}]^{-0.714}. \tag{4}$$

The terms in eqs. (2) and (3) are defined as follows: λ is the latent heat of vaporization, v_{fg} is the difference between vapor specific volume v_g and liquid specific volume v_f, C is the liquid heat capacity, T is the absolute temperature, dP/dT is the slope of the vapor pressure–temperature relation, P_0 is the source pressure, ρ_f is the liquid density, α is the gas volume fraction, and η is the choking pressure ratio (see Ref. [10]).

If the ambient pressure ratio, defined as $\eta_{amb}=P_{amb}/P_0$, is greater than η defined by eq. (4), for gassy systems, G_{max} may be represented by the incompressible form

$$G_{max}=\sqrt{2(1-\alpha)\rho_f(P_0-P_{amb})}. \tag{5}$$

Grolmes and Fauske [7] showed that the discharge coefficient for flashing flow in long horizontal tubes in the subsonic region could be represented by

$$F=\frac{127}{98}\,\frac{\omega^{1/2}(1+\omega)^{-1/3}}{\sqrt{N}} \tag{6}$$

where

$$\omega=P_0\rho_f/G_{max}^2 \quad \text{(eq. 2)}. \tag{7}$$

In eq. (6), the friction length parameter N is defined

$$N=4f(L/D)_{equivalent} \tag{8}$$

and for two-phase turbulent flow a constant value of

$$f=0.005$$

is often assumed for the friction factor.

For a non-flashing two-phase gassy system at void fractions α, approaching zero, the incompressible form of the discharge coefficient F is given by

$$F=(1+N)^{-1/2} \tag{9}$$

where N has the same meaning as defined by eq. (8) above.

Figure 2 shows the above relations for flashing and non-flashing discharge flows in the long L/D range. As a further simplification, one can represent an arbitrary flashing system whose ω value lies in the range of $10<\omega<30$ and for $L/D>1000$, by the relation

$$F=2N^{-1/2}. \tag{10}$$

Equations (10) and (6) are also compared in Fig. 2.

Equations (9) and (10) provide two convenient representations for the vent flow discharge coefficient for systems of interest. Note that for batch type process vessels, the effective discharge coefficient is rarely less than 0.5. However, for long tubular reactors, the effective relief discharge coefficient might well be less than 0.1.

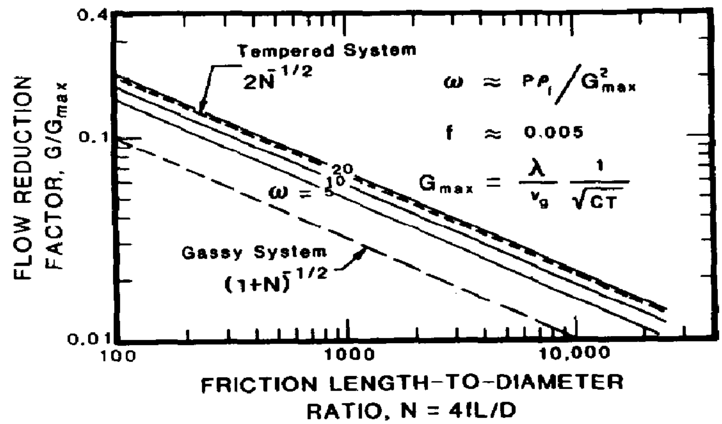

Fig. 2. Flow reduction factor for large friction length-to-diameter ratios representing subsonic discharge of saturated liquid flashing flow, and non-flashing gassy systems. Solid lines represent eq. (6) for flashing systems. Dashed lines represent approximations given by eqs. (9) and (10).

4. Adaptation of DIERS vent sizing relations to the problem of tubular reactors

The analysis presented here takes the approach that defining the maximum length of tubular reactor, which can be relieved by a vent area equal to the reactor flow area, is equivalent to the normal vent size evaluation for a batch reactor. We consider here both tempered as well as pure gassy systems.

It should be noted at the outset that in adapting analytic methods developed for batch reactors to tubular reactors, an implicit assumption of uniformity regarding temperature, concentration and/or extent of reaction within the tubular reactor that in most cases is far from reality. It is also true that, in most cases, the results of the proposed analysis will be more conservative due to real non-uniformities. For example, consider a flow cessation in a tubular reactor with a corresponding run-away reaction. The hottest zone which may be of considerably less extent than the total reactor will determine the system pressure and pressure relief activation. The expansion or relief of this zone will lead to pressure mitigation more quickly than if the total length of the tubular reactor were at uniform conditions. None-the-less, it is felt that the net conservative features of the proposed methods are justifiable in view of the essential absence of reported performance experience on emergency relief of tubular reactors.

4.1 Tempered systems

For tempered systems we cite the well established model of Leung [11], as the reference formulation. We choose to represent Leung's formulation in the following way:

$$A = \beta^* A_0 \tag{11}$$

where A is a required vent area and A_0 is a reference vent area based on reacting system characteristics and β^* is a dimensionless term which collects all parameters important to the allowance for pressure increase after relief activation as:

$$\beta^* = \left[\frac{T^\bullet_{set} + T^\bullet_{max}}{2T^\bullet_{set}}\right]\left[1 + \left(\frac{\rho_f C(T_{max} - T_{set})}{T_{set}(dP/dT)}\right)^{1/2}\right]^{-2}. \tag{12}$$

In eq. (12) the new terms are: T_{set} and T_{max}, which refer to the system temperature corresponding to the relief set pressure and to the maximum pressure to be allowed after relief activation, respectively; $T^\bullet_{set}$ and $T^\bullet_{max}$, which refer to the rate of temperature change (non-vented basis) at the same pressure conditions.

In eq. (11), the reference area A_0 is defined as

$$A_0 = \frac{M\rho_f (C/T_{set})^{3/2} T^\bullet_{set}}{F(dP/dT)^2} \tag{13}$$

where M is the tubular reactor inventory defined as

$$M = \rho_f L_R A_R \tag{14}$$

and A_R is the cross section area of the tubular reactors.

The flashing discharge mass flux of eq. (2b) has already been incorporated into eq. (13). Now, if one sets the vent area A in eq. (11) equal to the reactor area, A_R, then on combining (14), (13) and (11), one finds a relation for the maximum reactor length L_0 which can be vented by a single device which is given in dimensionless form as

$$\frac{L_0}{D_R} = \frac{F(dP/dT)^2}{\beta^* D_R \rho_f^2 (C/T_{set})^{3/2} T^\bullet_{set}}. \tag{15}$$

Equation (15) can be cast in more convenient form by considering the following additional details. First, recall F will be given by eq. (10) and N can be related to L_0 by the expanded form below.

$$F = \frac{2}{\sqrt{\Sigma K_i + 4f L_0/D_R}}. \tag{16}$$

The loss coefficients K_i may represent any relevant fitting factor. For example, one may encounter a 180° pipe bend which may be represented by 50 equivalent length-to-diameter ratios. On this basis the K factor for a 180° bend would be $4 \times 0.005 \times 50 = 1.0$.

A second detail applicable to eq. (15) is to note that most vapor pressure–temperature relations, including mixtures, can be represented over some suitable temperature range by a two-parameter Antoine equation of the form

$$P = \exp(a + b/T) \tag{17}$$

where T is the absolute temperature and a and b are the parameters that fit eq. (17) to specific data. The term $\exp(a)$ will have pressure units and the term b will have units, K.

With the above, the slope of the vapor pressure–temperature equation can be represented as

$$\frac{dP}{dT}=\frac{-bP}{T^2} \tag{18}$$

and with eqs. (16) and (18) we can represent (15) in the form

$$\frac{L_0}{D_R}=\frac{\frac{2}{\sqrt{\Sigma K_i+4fL_0/D_R}}\frac{P_{set}^2 b^2}{D_R T_{set}^4}\left(\frac{T_{set}}{C}\right)^{3/2}}{\beta^* \rho_f^2 \dot{T}_{set}}. \tag{19}$$

In the implementation of eq. (19) the important related terms of T_{set} and $\dot{T}_{set}$ will be obtained from empirical test data in the same manner as for batch reactor applications. Examples will be illustrated later in the paper.

If the actual reactor length L_R is less than the length L_0 defined in eq. (19), then a relief vent size less than the reactor cross section area would be adequate. The previous relations may be manipulated to show that to a first order the relative relief vent area is given by

$$\frac{A}{A_R}=\frac{L_R}{L_0}. \tag{20}$$

The above relation is conservative, and could be further refined by the consideration that the internal flow resistance factor should also be reduced by the ratio $(A/A_R)^2$ which leads to an iteration solution requirement.

4.2 Gassy systems

Gassy systems may be treated in a similar manner. The reference vent sizing approach using the DIERS methodology for gassy system in batch reactors is to balance the volumetric discharge rate Q_d, where

$$Q_d=\frac{FGA}{(1-\alpha)\rho_f} \tag{21}$$

with the volumetric gas generation rate, Q_g, given by

$$Q_g=\frac{M}{M_t}\frac{V_t}{P_m}\dot{P}_{max}, \tag{22}$$

The required vent area for a batch reactor is given by the combination of (21) and (22) as

$$A=\frac{(1-\alpha)\rho_f M V_t \dot{P}_{max}}{FGM_t P_m}, \tag{23}$$

The new terms introduced in the above relations are found in the Q_g term which contains information based on empirical test data (cf. [12]): M_t is the test sample mass, V_t is the free volume in the test equipment, $P^{\bullet}_{max}$ is the maximum pressure rise rate in the test volume V_t, and P_m is the maximum pressure allowed during relief activation.

We assume that the tubular reactor inventory $M=\rho A_R L_R$ and that G is given by eq. (5) for non-flashing flow. The discharge coefficient F is represented by eq. (9) in the modified form $F=(1+\Sigma K_i+4fL/D_R)^{-1/2}$. We further assume the initial void fraction $\alpha=0$.

With the above one finds the maximum length for a single relief device of area A_R given by

$$\frac{L_0}{D_R}=\frac{M_t}{\rho_f^2 V_t P^{\bullet}_{max}}\frac{P_m}{D_R}\left(\frac{2\rho_f(P_m-P_{amb})}{1+\Sigma K_i+4fL_0/D_R}\right)^{1/2}. \tag{24}$$

In the same manner as pointed out for tempered systems, if the actual reactor length L_R is less than L_0, then the relative reduction in vent area is given to a first order by

$$\frac{A}{A_R}=\frac{L_R}{L_0}.$$

Other comments relative to required correction to the internal friction resistance as noted previously also apply here.

The maximum length for a single relief device for a gassy system, as defined by eq. (24), should be conservative relative to other possible evaluations for certain scenarios. For example, we note the possibility that a flow blockage with continued reaction for a gassy system can lead to early relief activation, and therefore allow time for venting the reactor inventory prior to reaching the maximum reaction rate assumed in eq. (24). However, completion of an empty time evaluation will typically require more test information than needed for eq. (24).

5. Illustrative examples

The following examples will illustrate use of the previous tubular reactor evaluation models.

5.1 Tempered system case

We will set up a basis for evaluation of a tempered system as if certain information were available from test data.

5.2 Maximum reaction temperature and phi-factor

It is assumed that an estimate of the heat of reaction can be obtained directly from appropriate calorimetry data in which the reaction temperature increase is measured,

$$\delta = \frac{\phi M_w C \Delta T}{x} \tag{25}$$

where δ is the heat of reaction [J/mol]; ϕ is the test cell thermal inertia, where it is assumed that $\phi = 1.08$; M_w is the molecular weight of the reactant, where it is assumed that $M_w = 120$ g/mol; C is the liquid heat capacity, where it is assumed that $C = 2.1$ J/g °C; ΔT is the measured temperature rise, where it is assumed that $\Delta T = 310$ °C; and x is the reactant weight fraction, where it is assumed that $x = 0.5$.

With the above assumed values, one finds

$$\delta = \frac{1.08 \times 120 \times 2.1 \times 310}{0.5} = 169 \times 10^3 \text{ J/mol.}$$

However, with a known heat of reaction, inversion of eq. (25) allows one to estimate the expected reaction temperature rise for other conditions in which x, or ϕ, may be different from test values. For example, in the ideal limit of $\phi = 1.0$, eq. (25) would define the adiabatic reaction temperature increase.

The value of $\phi = 1.08$ is typical of a good reaction calorimeter. The working definition of the ϕ factor is

$$\phi = 1 + \frac{(MC)_{\text{test cell}}}{(MC)_{\text{test sample}}}. \tag{26}$$

A good approximation of eq. (26) for a steel wall vessel and an organic sample is

$$\phi \approx 1 + 10\,t/D$$

where t and D are the vessel wall thickness and diameter respectively. A typical value for a 1 inch schedule 40 pipe, tubular reactor ($t = 0.133$ inch, $D = 1.049$ inch) would be $\phi = 2.27$. With the previously estimated heat of reaction, $\delta = 169 \times 10^3$ J/mol and other property values previously assumed, eq. (25) inverted for reaction temperature rise for various cited values of the ϕ factor are:

ϕ	ΔT
1.0	335 °C
1.08	310 °C
2.27	148 °C

5.3 Reaction kinetics and P–T relation

The reaction self-heat rate would normally be obtained from calorimetry test data. However, to provide an illustrative example the following expression for reaction self-heat rate is based on assumed kinetics rate constant parameters:

$$\frac{\mathrm{d}T}{\mathrm{d}\phi}=(T_{\mathrm{m}}-T)u\exp[-w/(T+273.15)] \tag{27}$$

where $\mathrm{d}T/\mathrm{d}\phi$ is the reaction self-heat rate (°C/min); T_{m} is the maximum reaction temperature (°C); T is the instantaneous reaction temperature (°C); u is the pre-exponential rate constant parameter, here assumed to be $u=7.266\times10^{7}\ \mathrm{min}^{-1}$; and w is the activation energy parameter, here assumed to be $w=9148$ K.

The kinetics parameters, u and w, represent a hypothetical reaction having an onset temperature of ≈ 76 °C where the self-heat rate will approximately double with every 10 °C temperature increase. A hypothetical pressure temperature relation given by

$$P(\mathrm{psia})=5.9\times10^{5}\exp[-4167.5/(T+273.15)] \tag{28}$$

will have a normal boiling point temperature of 120 °C.

Figure 3 shows the self-heat rate ($\mathrm{d}T/\mathrm{d}\phi$) for three cases along with the pressure temperature relation. The three reaction cases correspond to ϕ factors of 1.0, 1.08 and 2.27 with the corresponding values of T_{max} relative to 76 °C as also indicated in Fig. 3.

Note the profound effect of the assumed ϕ factor for the 1 inch schedule 40 pipe ($\phi=2.27$). In fact, Fig. 3 suggests that if the relief set pressure or reactor design pressure is greater than 150 psia, the reaction path defined by case 3 could perhaps be contained without pressure relief. Also note that the differences in self-heat rate between the adiabatic case, $\phi=1.0$ and the hypothetical calorimeter case, $\phi=1.08$, are small. With respect to case 3 in Fig. 3,

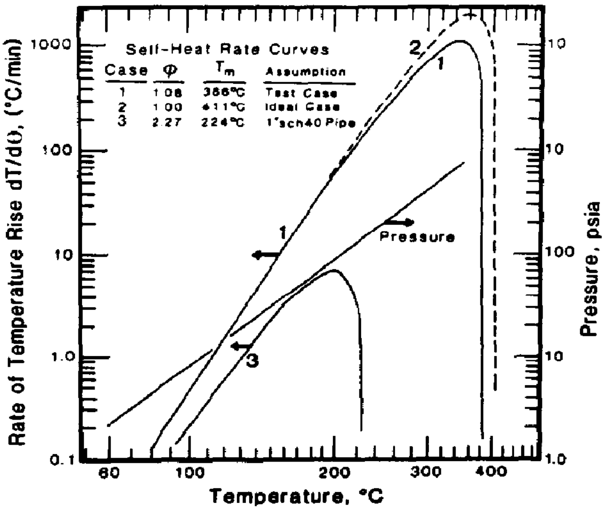

Fig. 3. Self-heat rate(s) and pressure–temperature relations for tempered system example problem.

any similarly large ϕ factor can often result in a significant reduction in pressure relief requirement. However, each case should be examined carefully since the effect of the heavy containment wall is always overstated in the static lumped heat capacity model considered here. The effect of the wall will also be less for flowing systems. However, the potential benefits may justify a more detailed thermal evaluation. These considerations having been noted, the calorimetry test case, $\phi = 1.08$ and $T_m = 386\,°C$, will be used for the example evaluation.

5.4 Evaluation case

Consider the following evaluation case: $P_{set} = 150$ psig or 164.7 psia (1.136×10^6 N/m^2), $P_{max} = 197.6$ psia (20% overpressure) (1.363×10^6 N/m^2), $T_{set} = 236\,°C$ (509 K), $T^{\bullet}_{set} = 171.6\,°C$/min (2.86 °C/s), $T_{max} = 248\,°C$ (521 K), and $T^{\bullet}_{max} = 237\,°C$/min (4 °C/s). In addition, we assume that $\rho_f = 900$ kg/m^3, $C = 2100$ J/kg, $D_R = 2.54 \times 10^{-2}$ m (1 inch) and $\Sigma K_i + 4f\,L_0/D_R = 0.025\,L_0/D_R$.

Using eqs. (15) through (19) and the above parameter list leads to

$$\left(\frac{L_0}{D_R}\right)^{3/2} = \frac{12.65 \times (1.136 \times 10^6)^2 (4167.5)^2 (509/2100)^{3/2}}{2.54 \times 10^{-2}\, 509^4 \beta^* 900^2\, 171.6/60}.$$

The term β^* must be evaluated using eq. (12). If one evaluates the term

$$\frac{\mathrm{d}P}{\mathrm{d}T} = \frac{bP}{T^2} = \frac{4167.5 \times 1.1356 \times 10^6}{509^2} = 18267\ \mathrm{N/m^2\,K}$$

then one finds, using the previous parameters,

$$\beta^* = \frac{(171.6 + 236.9)}{2 \times 171.6}\left[1 + \left(\frac{900 \times 2100(247.7 - 236)}{509\ 18267}\right)^{1/2}\right]^{-2}$$

or

$$\beta^* = 0.184.$$

Therefore, completing the evaluation, one finds

$$\left(\frac{L_0}{D_R}\right)^{3/2} = 46429 \qquad \text{or} \qquad \frac{L_0}{D_R} = 1282.$$

For a 1 inch (2.54 cm) diameter reactor, $L \approx 32.5$ m.

5.5 Gassy systems

An example illustration of the use of eq. (24) for gassy systems can be presented on the basis of the following assumed property, test data, and case specific data:

Case specific: $P_m = 400$ psia or 27.6×10^5 Pa
$P_{amb} = 14.7$ psia or 1.0×10^5 Pa
$D_R = 1$ inch or 2.54×10^{-2} m
$\rho_f = 900$ kg/m^3

Test data: $M_t = 0.01\,\text{kg}$

$V_t = 0.4 \times 10^{-3}\,\text{m}^3$

$P^{\bullet}_{\text{max}} = 400\,\text{psi/min}$ or $6.67\,\text{psi/s}$

Assumption: $1 + \Sigma K_i + 4f\,L/D \approx 0.025\,L_0/D$

With the above, one can evaluate eq. (24) to find

$$\left(\frac{L_0}{D_R}\right)^{3/2} = \frac{0.01 \times 400 \times (2 \times 900 \times 26.6 \times 10^5)^{1/2}}{900^2 \times 0.4 \times 10^{-3} \times 6.67 \times 2.54 \times 10^{-2} \times 0.025^{1/2}} = 31890$$

or

$L_0/D_R = 999$ or $L_0 = 25.4\,\text{m}$.

6. Concluding remarks

We have shown how the conventional vent sizing relations for batch reactors, as often used in implementation of the DIERS methodology, can be adapted to long tubular reactors. In specific instances, a tubular reactor may have a significant thermal inertia effect which can further mitigate the severity of a run-away reaction. This should be evaluated carefully since there may also be cases where the thermal inertia effect is less significant. For long reactors, it is likely in most cases that a non-reclosable relief device will be preferred. One implication of the relatively large friction effect on the discharge flow rate may be the inability to satisfy the requirements of reclosable safety relief valves for minimum upstream pressure drop.

The evaluations presented here indicate a maximum length which can be protected by a single relief device without indicating an explicit preference for location. However, as a practical matter, preference should be given to a relief device location nearest the presumed location of the mixture that is farthest removed from completion of the run-away reaction scenario. This may be, in most cases, at the inlet of the reactor where feed reagents are introduced. In circumstances where the normal product represents a potentially unstable hazard, the exit location may be preferred.

Highly viscous systems may also generate special requirements. While the methods presented here are generally applicable, use of turbulent flow, constant friction factor is not recommended for laminar flow discharge.

Finally, the important aspects of the DIERS methodology requiring system characterization for run-away reaction scenarios through empirical test data are equally important for tubular reactor consideration.

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