

## PRESSURE RELIEF SYSTEM DESIGN FOR VAPOUR OR TWO-PHASE FLOW ?

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Criteria will be presented which allow for a classification of a given reaction system with respect to the expected phase quality of the discharged mass flow. Based on this, for the case of non-foaming, low viscosity liquid phase reaction systems, an improved method for the prediction of the maximum level swell in the reactor using a sophisticated drift flux model will be described. From this, it can be assessed whether a two-phase or a single-phase vapour flow in the ventline will occur. The reliability of the predictions is checked by systematic top-venting experiments.

Depressurisation, chemical reactor, level swell, drift-flux model, emergency relief device, single-phase flow, two-phase flow.

### 1. Introduction

Chemical reactors or other plant components under pressure loaded with vapour/liquid mixtures<sup>1</sup> are equipped with independently operating safety devices such as emergency relief valves or bursting discs to avoid an unacceptable high internal pressure build up, e.g. as a result of a thermal runaway reaction or external heat input. The API RP 520/521 /1/ or the German AD codes A1 and A2 /2,3/ are the standard methods currently applicable to the design of the minimum required relief area of such safety devices. These guidelines, however, apply only to instances of single-phase vapour or liquid flow in the vent line.

During top venting of a reactor under pressure out of the vapour free board space, single-phase vapour discharge is normally immediately followed by a two-phase or multi-phase flow in the vent line. This is a result of entrainment of droplets from the free interphase and/or condensation of vapour in the reactor nozzle, level swell or ebullition eventually along with a superimposed foaming of the reactor contents. In the first two cases a (weak) two-phase discharge flow with a very low liquid mass content generally occurs for a short time only and, thus, the depressurisation rate is not appreciably lowered. In contrast, the slightly later occurring rise of the pool free surface up to the reactor nozzle causes a marked reduction in the flow quality on entry into the relief piping system and, hence, also of the discharged vapour acting as cooling medium. Under identical production conditions the minimum required relief area for controlling the same system pressure increase is in these later cases considerably larger than

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1 For the sake of simplicity only the term vapour is used hereafter. The following comments apply expressively also to gases.

that for the (desired) single-phase vapour flow. It depends on the type of reaction system to be vented and the boundary conditions, such as initial fill level, relief set pressure and vessel geometry. No standardized design method analogous to that available for single-phase discharge flow is presently available for the case of a two-phase discharge.

Despite the complex phenomena occurring in parallel during the depressurisation of a (reaction) system /4/, a conservative design of the required relief area is currently assumed to be possible for two-phase flow. In particular, the design methodology developed by the US "Design Institute for Emergency Relief Systems" (DIERS) /5/ permits acceptable dimensioning of the relief areas even where runaway reactions are involved. The procedure in the design of emergency relief valves for two-phase flow is described in detail, for example, by Friedel /6/.

In this paper we will not discuss the design of the relief areas in the case of two-phase flow, but rather focus on the prediction of the phase quality of the transient flow on entry into the relief system that forms part of the proceeding (calculation) step in a fluid dynamic design. In this context, the physical and geometric boundary conditions, at which two-phase flow is expected to occur in the vent line, after a full swelling up of the vessel contents will be discussed. In other words - for the case of flashing or gas-dissolving liquid phase or boiling or gas-producing reaction systems, the range of parameter combinations is formulated, within which the relief area of safety valves or bursting discs may still be designed with the assumption of vapour flow only by using the API RP 520/521 or the German AD codes A1 and A2.

## **2. Characterisation of vapour/liquid systems with respect to their swelling behaviour during top venting**

Two-phase flow in the case of top venting normally occurs with those systems where rising in the liquid and separation at the free interphase of the vapour produced is hindered. This may be caused by the abrupt inception of the bubble generation and growing in connection with either a gradual rise of the individual bubbles or of bubble clusters, especially in high viscosity liquids, or by a foam layer above the liquid free surface. These effects can cause the reactor to empty almost completely in the event of top depressurisation, even if it contains chemically non-reactive media. In the following, the influence of both effects will be dealt with separately.

### **2.1 Liquid phase viscosity effect on the level swell behaviour**

The free bubble rise velocity in high viscosity liquids is essentially independent of the reactor diameter and substantially lower than that in low viscosity media. This hinders separation of the vapour from the two-phase layer into the vapour space during the short time of top reactor venting. The homogeneously distributed bubbles in the mixture are almost immediately generated and the volume increases with progressing relief time as a result of the normally occurring pressure decay. Due to this, the pool free surface as a whole is lifted. Schemberg /7/ showed with depressurisation experiments carried out with carbon dioxide/ Newtonian Luviskol/ water solutions that with low relief rates and fill levels a discharge of a two-phase mixture via the vent line is always likely, if the liquid viscosity exceeds roughly 100 mPa s. Herewith, this viscosity limit relates to the temperature prevailing when the safety device is activated.

The effect of the viscosity on the swelling behaviour is particularly important in the depressurisation of a continuously progressing polymerisation reaction, since during the thermal runaway the amount of polymer produced increases constantly leading to a fluid system with an effective liquid phase viscosity exceeding that of the monomer many times. At the same time, the viscosity decreases as a result of the rising temperature, thus, again lowering the effective viscosity. A procedure for designing the relief area is described in detail by Friedel et al. /8/ by using the example of a vinyl acetate polymerisation reactor.

## 2.2 Foaming reaction systems

In the case of fluid systems, where foam is produced during depressurisation as a result of bubble formation and penetration through the free interphase, the occurrence of a two-phase flow in the vent line must be regarded as likely, even when the fill level in the reactor at initiation of venting is low. In principle, with a view to foam stability a distinction is made between polyhedral and spherical foam structures. The former type is produced by surface-active substances and exhibits a composite bubble structure above the actual swelling two-phase mixture in the reactor pool. Spherical foam develops from individual bubbles, if the bubbles, e.g. in a high viscosity media, cannot fully separate and penetrate through the free interphase. These spherical bubbles collapse on leaving the liquid surface. Intermediate foaming behaviour is, indeed, possible. For example, in the case of systems with high surface tension and viscosity - such as encountered in the late reaction stages of emulsion polymerisation processes - a very stable structure resembling shaving foam can form during depressurisation, which suddenly fills the reactor free vapour space completely. In the case of an emergency top relief through a ruptured bursting disc the reactor liquid content would then be almost totally discharged. Even with depressurisation of the reactor by an intermittently opening and closing emergency relief valve, a considerable amount of the initial liquid mass would be delivered into the relief containment.

The characterisation of the foaming behaviour of a vapour/liquid system solely by means of macroscopic physical properties is not possible at present, and so for classification we must rely on observations obtained during production process conditions or on dedicated laboratory and pilot plant tests. Experiments have been carried out in a pilot plant to investigate the influence of the foam produced during depressurisation on the amount of mass discharged. A typical result is shown in fig. 1. The total amount of liquid mass discharged, relative to the initial mass, during the complete depressurisation of boiling water and of a water-surfactant solution are plotted against the initial fill level. In the depressurisation experiments with water, a two-phase flow through the relief orifice occurs only when the initial fill level is greater than about 60 %. With the water/surfactant solution this is the case when the fill level exceeds about 15 %. Consequently, the relative mass discharge, under equivalent initial venting conditions, is about 75 % greater in the case of foaming than in the case of water, where at high initial fill levels only about 35 % of the initial amount of liquid is discharged.

In the case of a thermal runaway reaction venting experiment the unknown foaming characteristic of the system is normally compounded by the problem that the reaction kinetics and the fluid dynamic conditions at the set pressure of the safety device are not accurately known. For classification of the foaming behaviour of such reaction systems, depressurisation experiments may be carried out on a 100 ml scale in an upgraded adiabatic VSP (Vent Sizing

Package) reaction calorimeter. Evaluation of own numerous experiments and of data in the literature /5/ support the following finding criterion: in vapour/ liquid systems, in which the relative mass discharge in VSP depressurisation tests is greater than about 60 %, a foam layer above the initial free vapour-liquid interphase must have been produced during depressurisation. Thus, in designing for the relief areas a two-phase flow must always be reckoned with. A design on the basis of the API RP 520/521 or AD codes A1 or A2 would in this case generally lead to underdimensioned relief areas.

As an (empirical) rule of thumb, it can be concluded that single-phase vapour flow during top venting generally only occurs with low viscosity, non-foaming, single-component systems or mixtures of chemically similar substances. But even with these systems, a temporary two-phase flow in the vent line can occur, if an unfavourable combination of relief parameters such as high initial fill level and oversized relief area is met in practice. The individual limits of these parameters are discussed in detail below.

### 3. Fluid dynamic criterion for the occurrence of two-phase flow in the vent line for non-foaming systems

The objective is to derive a criterion for the prediction of the flow type to be expected during venting as a function of the macroscopic physical properties, the geometry of the reactor and the relief line, and the initial venting conditions. According to this, the design follows either the procedures outlined in the standards like API RP 520/521 and AD A1/A2, or in the DIERS methodology. The validity of this equation will be then checked for low viscosity and non-foaming liquid phases by means of results obtained in a large number of depressurisation experiments with different fluid systems.

One basic condition for the vapour-liquid mixture swelling up as far as in the vent line and development of a two-phase flow in the reactor nozzle is that at any time more vapour volume per time unit is produced in the liquid mixture than can separate via the pool free surface, fig. 2:

$$\dot{V}_g > \dot{V}_{sep} \quad (1)$$

For the first concern, the type of vapour formation is irrelevant. The vapour/gas phase can be generated by coming out of solution, a gas-producing chemical reaction or, as assumed here, from flashing. By means of a vapour mass flow balance it can be shown that the amount of vapour produced in the liquid per time unit and the amount of vapour discharged in the vent line are almost identical, ignoring the small change in density of the vapour phase in the vessel:

$$\dot{V}_g = \dot{V}_E = \dot{G}_g A_E / \rho_g \quad (2)$$

$\dot{G}_g$  denotes the vapour mass flux in the relief area  $A_E$  and  $\rho_g$  is the vapour/gas density. The exiting vapour mass flux can be predicted very accurately in case of an ideal nozzle flow as a function of the vessel stagnation pressure and the fluid properties. In practice, the pressure loss in the vent line and in the emergency relief device must of course be taken into consideration.

The volumetric flow rate of the separating vapour is calculated by using the basic drift-flux model of Zuber and Findlay /9/ originally developed for pressure driven fully developed pipe flow and, nevertheless, traditionally used in describing transient reactor contents volume

swelling. It is given by:

$$\dot{V}_{\text{sep}} = A_R u_{gj} \frac{\hat{\alpha}}{1 - C_0 \hat{\alpha}} \quad (3)$$

Here  $A_R$  denotes the vessel cross-sectional area,  $u_{gj}$  the vapour drift velocity and  $C_0$  a distribution parameter ranging between 1.1 and 1.3, depending on the flow pattern during ebullition and the pressure range. The mean volumetric void fraction  $\hat{\alpha}$  at pool free surface in the reactor is approximated by an equation proposed by Grolmes /10/ for a vertical cylindrical reactor:

$$\hat{\alpha} = \frac{2 \bar{\alpha}}{(1 + \bar{\alpha} C_0)} \quad (4)$$

With some algebraic transformations, equations (1) - (4) provide a final equation for the (theoretical) initial average volumetric void fraction  $\bar{\alpha}$ , or more conveniently, the initial fill level  $(1 - \bar{\alpha})$  of the reactor. It is now postulated that for an initial fill level higher than this value the reaction mixture will swell up to the reactor top and, thus, two-phase flow is expected to occur in the vent line. The mathematical relationship reads:

$$\text{Initial fill level}_{2,ph} = 1 - \bar{\alpha}_{2,ph} > 1 - \frac{1}{\frac{2 u_{gj} \rho_g A_R}{\dot{G}_g} + C_0} \quad (5)$$

The primary variables in this correlation are the vapour density, the vapour mass flux and the ratio of reactor cross-section to relief area, which are readily available in practice. Further on, the drift velocity  $u_{gj}$  and the distribution parameter are primary variables describing the phase separation behaviour in the liquid mixture.

The drift velocity can be calculated according to various semi-empirical equations. Zuber and Findlay /8/ proposed a correlation based on the free rise velocity of a single bubble:

$$u_{gj} = 1.53 \left[ \frac{\sigma g (\rho_l - \rho_g)}{\rho_l^2} \right]^{0.25} \quad (6)$$

The distribution parameter  $C_0$  is deduced essentially as a constant varying in dependency of the flow regime between 1 and 1.3. A value of unity corresponds to the case of homogenous flow. An extension of the correlation of Zuber and Findlay is suggested by Kataoka and Ishii /11/. By fitting a large number of results obtained during stationary vapour/gas-liquid flow experiments, in effect, an additional density ratio considering the pressure and a dimensionless number  $N_v$  including the liquid phase viscosity are introduced. For typical reactor sizes used in chemical industry practice, the drift velocity is given by an equation being independent of the channel diameter:

$$u_{gj} = 0.030 \left[ \frac{\sigma g (\rho_f - \rho_g)}{\rho_f^2} \right]^{0.25} \left[ \frac{\rho_g}{\rho_f} \right]^{-0.157} N_v^{-0.562} \quad \text{for } N_v \leq 0.00225 \quad (7)$$

$$u_{gj} = 0.92 \left[ \frac{\sigma g (\rho_f - \rho_g)}{\rho_f^2} \right]^{0.25} \left[ \frac{\rho_g}{\rho_f} \right]^{-0.157} \quad \text{for } 0.1 > N_v \geq 0.00225 \quad (8)$$

$$\text{with } N_v = \frac{v_f \rho_f}{\sqrt{\rho_f \sigma} \sqrt{\frac{\sigma}{g (\rho_f - \rho_g)}}} \quad (9)$$

For fully developed two-phase flow in round tubes, the calculation of the distribution parameter is proposed by Kataoka and Ishii to follow:

$$C_0 = 1.2 - 0.2 \sqrt{\frac{\rho_g}{\rho_f}} \quad (10)$$

Analytically, the distribution parameter is now always less than 1.2.

The suitability of both drift flux models for describing the void fraction and, thus, the swell behaviour of the reactor contents are checked by a large number of experimental data /12,13,14,15,16,17/. In fig.3 the weak predictive accuracy of the Zuber and Findlay model is demonstrated by comparison of the calculated vapour volume fluxes with experimental results. Ideally, the values should lie on or tightly agglomerate along the diagonal. Clearly, with the Zuber-Findlay model the calculated values would be extremely low.

The vapour volume flow predicted by using the model of Kataoka and Ishii correlates by far better with the experimental data, fig. 4. Up to a vapour volume flow of about 0.6 m/s the measured values are nearby the diagonal, thus, indicating good consistency with the experimental data. Beyond this limit, the values scatter and diverge from the diagonal. Altogether, this model has a much more higher predictive accuracy than that of Zuber and Findlay and it should be suitable for the calculation of the drift velocity in the reactor during venting, since here the drift velocities are mostly lower than 0.6 m/s.

#### 4. Experiments

To confirm the validity of the criterion recommended in eq. (5), additional experiments were carried out in two pilot test rigs, fig. 5. These essentially consist of a jacket heated reactor and a catchtank. Relief takes place via a vent line or an overflow line, which are arranged on the vapour space side and into which orifice plates with various bore diameters simulating different relief cross-sections can be inserted and a quick-opening valve for the controlled initiation of venting is installed. In each case, pressure and temperature were measured at different points.

The volumetric void fraction in the reactor head was determined with a conductivity probe and the size and composition of the mass flow was determined by combined measurements with a pitot tube and a gamma densitometer.

Systematic depressurisation experiments were carried out with refrigerants R12, R22, R114, methanol and water as media. As an example, measurements obtained during top venting of refrigerant R12 are shown in fig. 6. The experiments, in which two-phase flow occurred in the vent line, are identified by a shaded symbol. Herewith, this flow pattern was assigned by measurement of the phase composition in the vent line. In the figure also the limiting curve calculated according to eq. (5) for the initial fill level as a function of the ratio of relief area to reactor cross-section has been introduced. This boundary curve falls, as expected, with increasing cross-sectional ratio, i.e. increasing relief area with constant reactor cross-section indicating a higher initial venting rate. With an initial fill level greater than the calculated limit value, a two-phase mixture is discharged at least temporarily through the vent line. It is obvious that the change from two-phase to single-phase flow occurs in the experiments at a somewhat higher fill level than according to the calculation, which means that a slightly safe design would be applied.

The analytically determined limit for the occurrence of two-phase flow in the vent line is confirmed by further results obtained in venting experiments with water and methanol. In Tab. 1 the experimentally deduced minimum initial liquid fill levels, for which two-phase flow is just expected to occur in the ventline, are compared to the predicted values by using eq. (5) in combination with the models of Ishii/Kataoka, Hardekopf /18/ and of Zuber/Findlay, the latter with a distribution parameter of 1.5. This relatively high value is proposed by Grolmes /10/ as a correlating parameter in the course of the development of the DIERS methodology /5/. For the conditions of the depressurisation experiments with water, the predicted initial liquid fill levels are partly lower than the experimental ones. In detail, in the experiments for initial fill levels lower than 60 % vapour flow has already occurred, while with our own and with the Hardekopf model two-phase flow is calculated for fill levels higher than about 55 %. With the Zuber/Findlay model two-phase flow is predicted for values higher than 44 %. All model predictions are in this context over-conservative with respect to the adequate design of relief areas.

In the case of the venting experiments with methanol, the predictive accuracy of our own and of the Zuber/Findlay model is extraordinarily good, especially when comparing the data of the test with the larger relative relief area. For the conditions of the depressurisation experiments with refrigerant R12, however, only by using our own model an acceptable prediction of the minimum fill level for the case of a larger relative relief area is possible. For the experiments with a relative relief area of 0.0088 the result of the Zuber/Findlay equation is about 11 % higher than the experimental value. By using the model of Hardekopf for the organic media R12 and Methanol, the predicted initial fill levels are mostly higher than in the experiments. This implies that the prediction is not conservative in the case of a relief area design, since a vapour flow will be predicted for fill levels at which in the experiments two-phase flow has occurred.

Altogether, the results of our own model fit fairly well to the experimental data and it could be used as a more general criterion for the prediction of the occurrence of two-phase flow in the ventline. The Zuber/Findlay equation tends to a conservative prediction. The model of Hardekopf can only be used for water or aqueous systems.

Medium	Pressure [bar]	Relative relief area [-]	Minimum initial fill level for two-phase flow occurrence			
			Experiment	This work (Ishii/Kataoka)	DIERS (Zuber/Findlay, $C_o = 1.5$ )	Hardekopf /18/
Water	3.5	0.0056	60	55	44	56
Methanol	10	0.0014	65	66	62	74
"	"	0.0025	55	54	53	69
R12	10.7	0.001	83	75	76	83
"	"	0.0027	70	56	59	73
"	"	0.0088	35	34	44	61

**Tab. 1:** Model predictions and experimentally derived minimum initial liquid fill level necessary for occurrence of two-phase flow in the ventline

## 5. Summary

For assessment of the probability of two-phase flow in the vent line during emergency relief of a reactor under pressure charged with a vapour/liquid (reaction) mixture it is necessary first of all to investigate the foaming behaviour of the fluid system. If this has been established by experiments to be the case, for example in a small scale reactor depressurisation experiment under venting conditions or by experience, two-phase flow through the relief area is normally expected. Also, in the case of the dynamic viscosity of the liquid phase exceeding approximately 100 mPa s, as a rule, complete swelling of the reactor contents up to the top, which leads to a two-phase flow in the vent line, is likely due to a hindered separation of the bubbles in the mixture pool. Design of the emergency relief valve or the bursting disc according to the API RP 520/521 or to the German AD codes A1/A2 would then in both cases generally lead to underdimensioned relief areas.

An equation was derived from a volume flow balance, allowing for prediction of the flow type in the relief line as a function of the relative relief area and the maximum initial fill level for boiling systems. It should be used in connection with the drift-flux model of Kataoka and Ishii, which has been demonstrated by comparison with the results of depressurisation experiments. This criterion is only valid for non-foaming low viscosity reaction systems, but since it is a pure fluid dynamic approach, it is independent of the way in which the gas or vapour is generated. In this way, for flashing liquids or gases coming out of solution, or boiling or gas-producing reaction systems, the limits of application of API RP 520/521 and the German AD codes A1/A2 are, thus, established.



## Nomenclature

$A_E$	$[m^2]$	relief area
$A_R$	$[m^2]$	reactor cross-sectional area
$C_O$	$[-]$	distribution parameter
$\dot{G}$	$[kg/(m^2s)]$	mass flux
$\dot{G}_g$	$[kg/(m^2s)]$	vapour mass flux
$g$	$[m/s^2]$	gravitational constant
$u_{di}$	$[m/s]$	drift velocity
$\dot{V}_E$	$[m^3/s]$	discharged vapour volume flow
$\dot{V}_g$	$[m^3/s]$	generated vapour volume flow
$\dot{V}_{sep}$	$[m^3/s]$	separated vapour volume flow
$\bar{\alpha}$	$[-]$	average volumetric reactor void fraction
$\hat{\alpha}$	$[-]$	void fraction at pool free surface
$\rho_l$	$[kg/m^3]$	liquid density
$\rho_g$	$[kg/m^3]$	vapour density
$\sigma$	$[N/m]$	surface tension
$\nu_l$	$[m^2/s]$	liquid viscosity

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## Figures

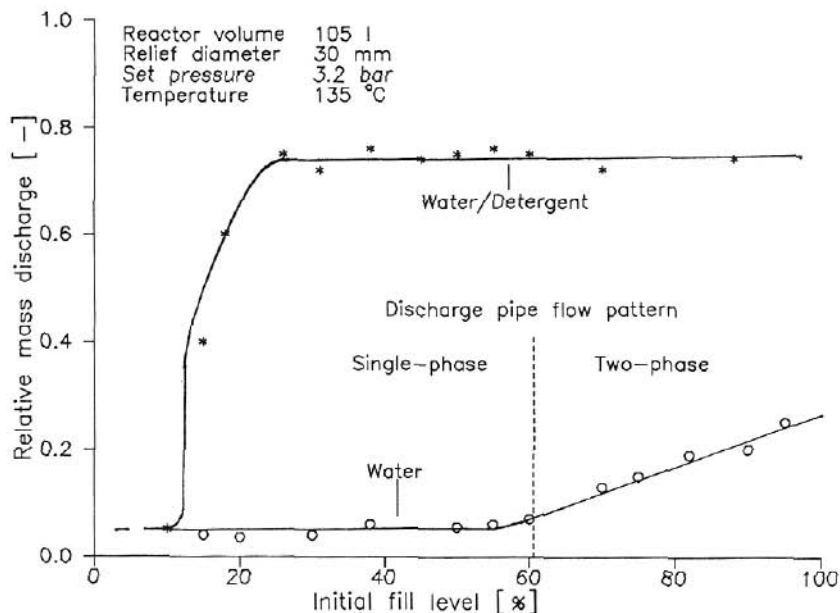


Fig. 1: Relative mass discharge of water and water/detergent and flow type transition as a function of initial fill level

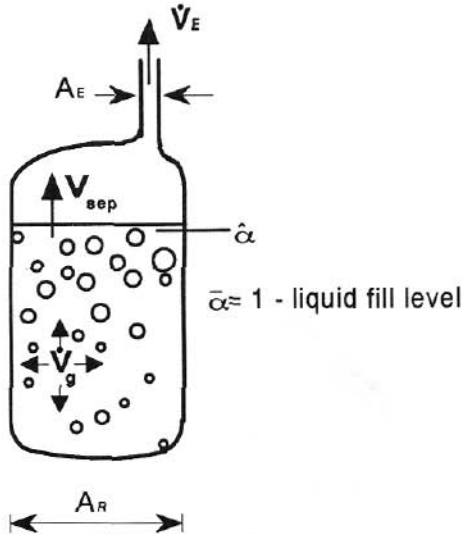


Fig. 2: Reactor geometric dimensions and vapour volume flows used in the flow balance

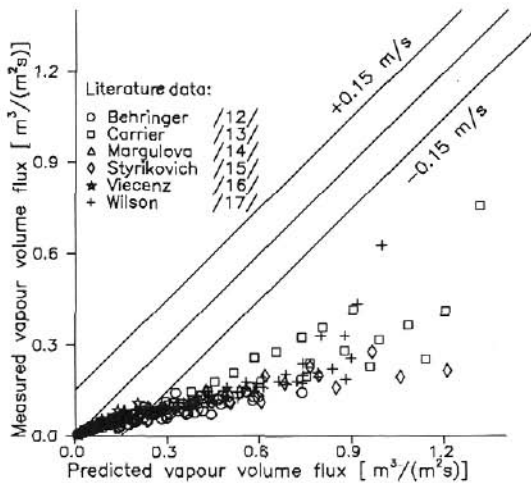


Fig. 3: Predictive accuracy of the Zuber/Findlay drift flux model on the basis of published experimental results at stationary vapour-liquid flow in bubble columns

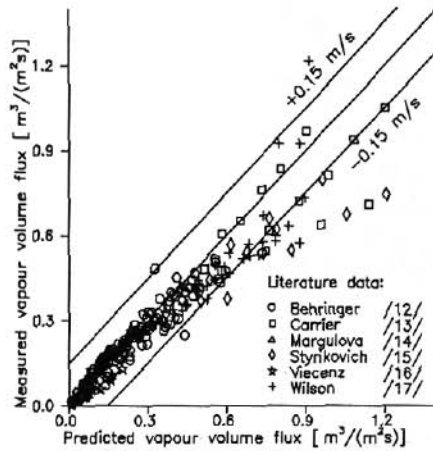


Fig. 4: Predictive accuracy of the Kataoka/Ishii drift flux model on the basis of published experimental results at stationary vapour-liquid flow in bubble columns

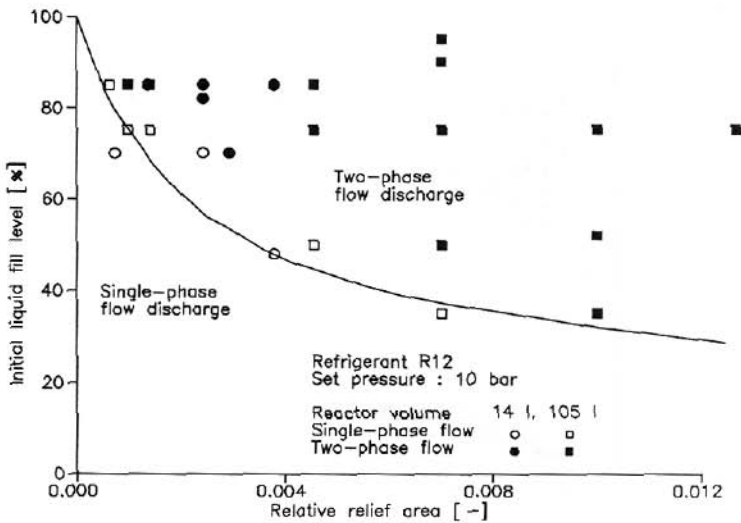


Fig. 6: Initial fill level as a function of ratio between relief area and reactor cross-sectional area in venting experiments with R12 and boundary line between single-phase and two-phase flow discharge regions

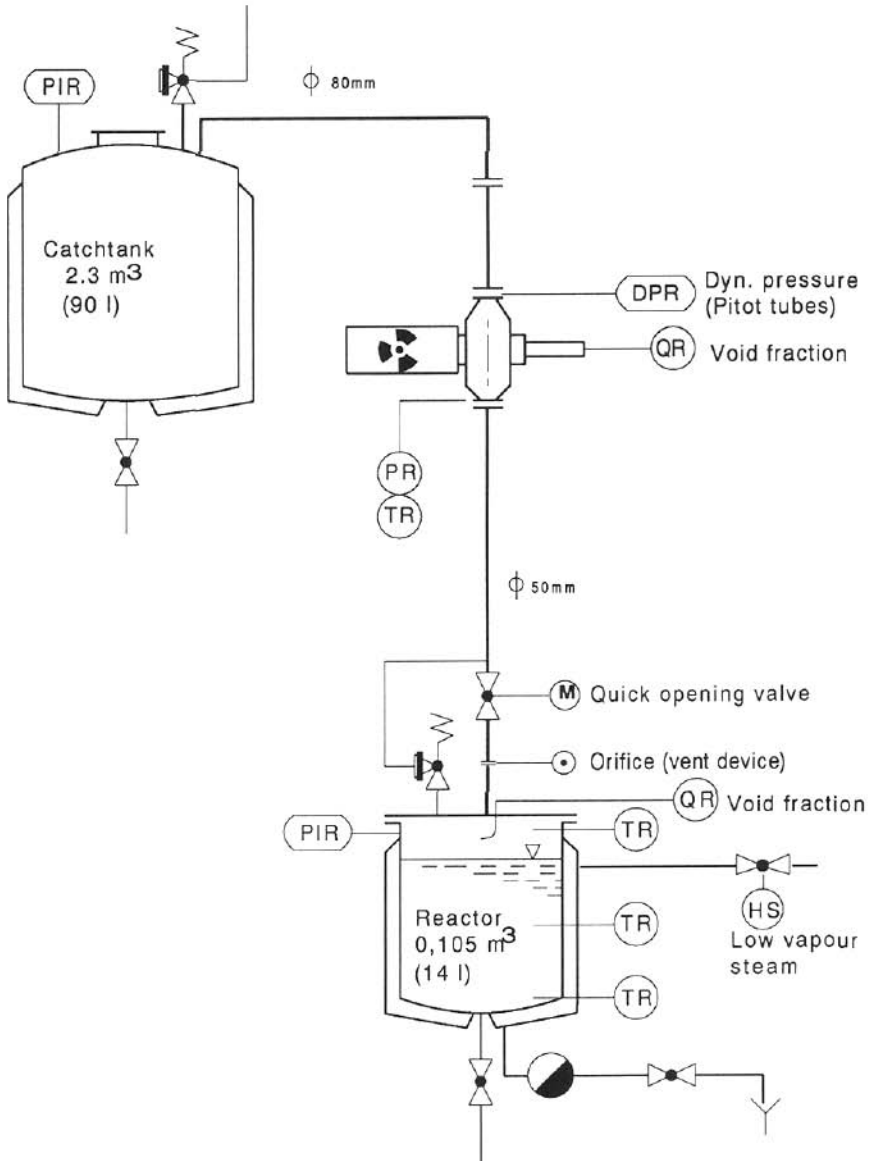


Fig. 5: Test facility for depressurisation experiments (105 l and 14 l)