Chapter 23

Chemical Plant Safety

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23.1 Safety Problems in Chemical Engineering

Safety problems arising from unit operations in chemical engineering depend on the mechanical, thermal or chemical process to be performed. Especially in thermal and chemical processes often high pressures have to be mastered and with exothermal reactions the conditions of the heat balance have to be carefully observed to avoid an unallowable temperature increase or a pressure excursion.

Chemical reactions can be controlled by mass transport, heat transfer or in a catalytic way. Heat transfer and reducing mass flow rate of the reacting components may be sometimes too slow to avoid an excursion of the reaction and then a pressure relieve by vent valves can be an additional measure to keep the system within safe borders. If the reacting components are liquids flashing may occur after opening the vent valves and a two-phase flow mixture leaves the nozzle. Under certain conditions - e.g. with poisoning or explosive components - it is not possible to blow the two-phase mixture into the free atmosphere. Pressure subpression systems or containment vessels have then to be used. Pressure subpression can be performed by injecting the two-phase mixture into a cold liquid, where the gas phase is condensed and absorbed.

For the safe layout of a plant or an apparatus under these venting conditions several two-phase flow phenomena have to be well understood. For pure substances the following phenomena have to be taken in account

- flashing
- thermodynamic non-equilibrium between liquid and gas
- phase separation and entrainment in the vessel
- critical mass flow rate in the nozzle
- pressure drop under high velocities and
- chugging phenomena during condensing in a pressure subpression system.

In a multi component system, where some substances are more volatile than the others, additional questions arise, like

- how differs the flashing process from single components,
- stays the more volatile component in the solution or is time enough for destillation,
- may the more volatile component form early nuclei for boiling and flashing.

Finally with reacting components, the reaction cinetics and the heat and mass transfer between the liquid and the gas phase has to be known.

For pure substances, some two-phase flow phenomena concerning these safety problems, are known from nuclear engineering, however, there the experiments and theoretical deliberations mainly were made for water substance. In some cases also tests were done with refrigerants. For organic liquids, however, very few is known about flashing, critical two-phase flow and quenching during fast transients.

The situation becomes even worse for safety studies. One can perhaps start from the assumption that during fast depressurizations there is not enough time to allow the more volatile components to go out of solution. Up to what extent these components, however, may accelerate the flashing process by forming nuclei has to be studied for each system separately. For constant pressure usually the reaction cinetics are wellknown. The question is, whether they behave the same under fast transients. For calculating the heat and mass transfer under accident conditions, the present status of knowledge also allows only to start from steady state experience.

23.2 General Safety Strategy

Like in all technical disciplines also in chemical engineering the safety strategy follows the principle which may be called the defence in depth. This reaches from the manufacturing of the components, over repeating tests, up to the operation procedure.

Already the mechanical layout of components relevant for safety, is made in a way, that in addition to careful and detailed calculations large safety margins are observed. Great care is taken for material selection, manufacturing and material tests. The probability for a component failure is minimized.

Safety systems for example emergency coolers, or relieve valves are usually provided in a redundant number to improve the availability of the safety systems in case one subsystem should fail and to decrease the probability of a catastrophic failure.

The engineer very often is in the position, that he has to lay out an apparatus or a machinery without knowing all details of physical phenomena to be expected during operation. In safety deliberations a way out of the arising problems sometimes seems to be offered by performing the calculations with conservative assumptions and worst case initial conditions. Conservative means, that the worst physical behaviour is assumed with respect to safety effects.

A certain assumption may be conservative in one way and may be not in the other. For example laying out a safety relieve valve in a conservative way, flow conditions have to be assumed in the nozzle, which give minimum critical flow rate. On the other hand, calculating the depressurization in a vessel after a break of a pipeline, maximum critical flow rates have to be taken in account, to predict the forces acting onto the internals due to pressure pulses in a pessimistic way. So one easily can run out of a logical correlation system. Finally one always can argue what conservatism is conservative enough.

Basing safety calculations on conservative assumptions, certainly give early results. Doing the calculations with best estimate initial and boundary conditions needs a detailed and well based knowledge of the physical phenomena which may require a long and expensive research. Finally, however, an optimal layout of safety systems has to be based on a good and fundamental understanding of the physical phenomena.

23.3 **Flashing and Phase Separation**

Pressure waves in a liquid near the saturation line may cause stable or unstable conditions. A sudden pressure increase always stabilizes the thermodynamic state, because the saturated liquid or vapour will be changed to subcooled liquid or to superheated vapour (see Figs. 23.1 and 23.2). Depressurization, however, causes unstable conditions, i.e. the saturated liquid will become superheated and the saturated vapour subcooled.

The question now is, how fast this unstable situation can be stabilized again by boiling or condensing. For safety deliberations mainly pressurized liquids, are of interest and therefore the boiling delay and the maximum thermodynamic dis-equilibrium has to be known to calculate the flashing phenomena correctly.

Observations of the pressure and temperature behaviour during a depressurization - also called blowdown - measured in a vessel originally filled with a refrigerant partially in liquid and partially in gaseous phase under saturated conditions, is shown in Fig. 23.3. After opening a relief valve the pressure decreases rapidly for a short period, however, then increases again. After reaching a maximum the pressure is lowered more slowly, however, continuously.

The temperature in the liquid and in the gas phase shows at the beginning a remarkable deviation from saturation conditions and not before the moment, when the pressure reaches the maximum thermodynamic equilibrium between the phases can be observed. In the period B - C, there is an evaporation delay in the mixture and the temperature of the liquid stays almost at the value of the initial conditions. At the point C bubble growth starts in the mixture due to flash evaporation. During the period C - D the flashing mixture moves upwards and the superheating of the liquid slowly decreases. At point D the flashing boundary reaches the outlet nozzle at the top of the vessel which originally was not completely filled with liquid and now instead of single phase vapour a two-phase mixture flows through the nozzle. The maximum volumetric flow rate out of the nozzle is decreasing by this

High pressure wave	subcooled liquid superheated vapour	stable
Low pressure wave	superheated liquid subcooled vapour	unstable

Figure 23.1: Stable and unstable behaviour with pressure waves.

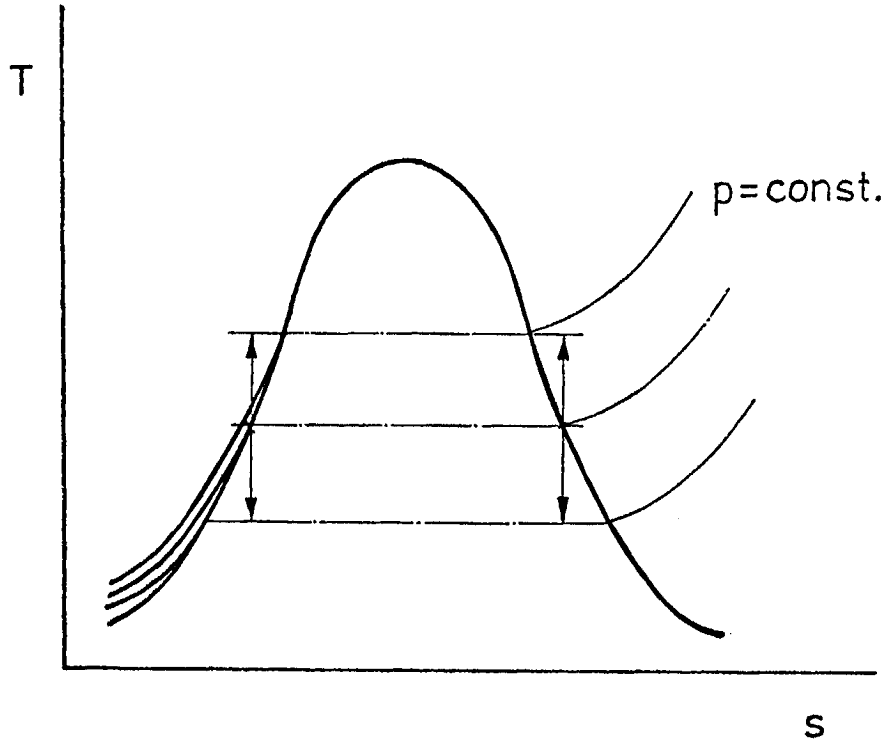
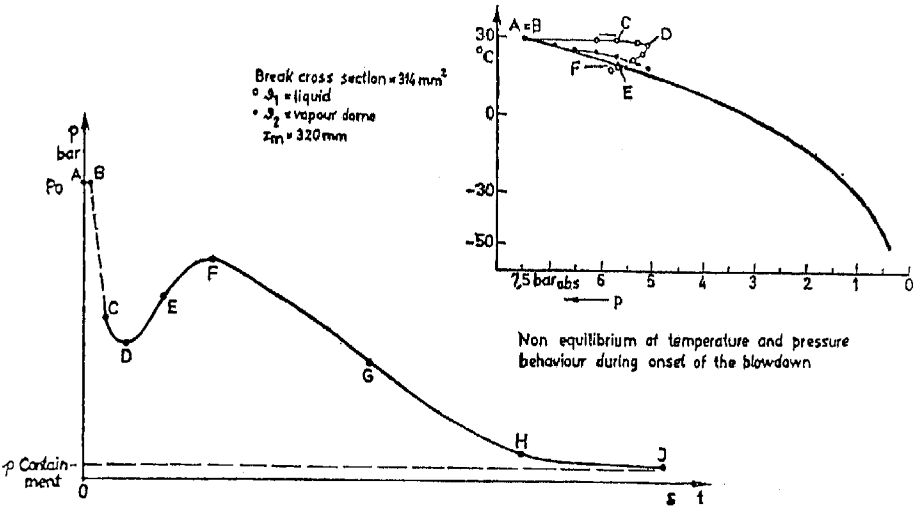


Figure 23.2: Pressure waves and stability.

effect. In the period D - F due to non-equilibrium conditions the flash-evaporated volume prevails the out streaming volume, which allows the pressure to rise again. At the point F the evaporated volume by flashing is equal to the out streaming volume, which can be deduced from the maximum in the pressure curve. In the period D - F the thermodynamic dis-equilibrium is rapidly reduced due to the evaporation by flashing. The period



A : initial conditions

B-C : evaporation delay in the mixture
onset of vapour decompression

At point C : onset of bubble growth in the mixture due to flash-evaporation

C-D : Flashing mixture moves upwards

At point D : Flashing boundary reaches the outlet pipe at the top of the vessel.
Onset of two-phase outflow.

D-F : Due to the non-equilibrium effects the flash-evaporated volume prevails the outstreaming volume

At point F : Flash evaporated volume = outstreaming volume

F-H : Continuous pressure decrease due to homogeneous outflow

At point H : Mixture level drops below the altitude of the outlet pipe

J: Pressure equilibrium is reached

Figure 23.3: Thermodynamic disequilibrium during depressurization.

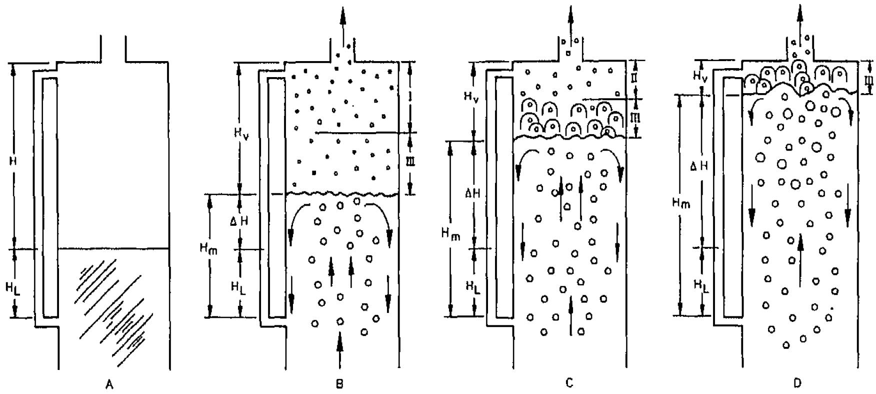


Figure 23.4: Schematic description of hydrodynamic effects with phase separation.

F - H shows continuous pressure decrease with homogeneous outflow and at the point H the mixture level drops below the position of the outlet nozzle. Finally at I pressure equilibrium with the atmosphere is reached.

Schematically the hydrodynamic effects during depressurization and blowdown are illustrated in Figure 23.4. Besides the thermodynamic dis-equilibrium the phase separation, that is the rising velocity of the vapour and also a fall back of the liquid, plays an important role for defining the thermodynamic and fluiddynamic conditions at the outlet. As long as the mixture level or swell level is far away from the outlet nozzle, the carry over of droplets also influences the quality at the outlet.

Measuring the void fraction over the height of the vessel during the blowdown, gives the conditions shown in Figure 23.5. By carrying over liquid droplets the void fraction in the upper part of the vessel decreases where originally only vavpour was present. In the lower parts of the vessel - originally filled with liquid - the void fraction increases continuously with time.

There are several models in the literature for predicting void fraction and phase separation in a pool, when vapour is blown through from below and is rising in the liquid (see Figure 23.6). The references in this figure are discussed in detail by Viecenz (1980)and the reference numbers there refer to the list of literature (see Viecenz (1980). Most of these models are for steady state conditions.

Viecenz (1980), based on careful measurements, by combining and extending the models in the theory developed an own simple correlation for the mean void fraction in the vessel which is shown in Figure 23.7 and which predicts not only his own experiments but also measurements by Behringer (1972), Margulova (1953) and Wilson (1961) as well.

Viecenz (1980) observed two regions where the phase separation

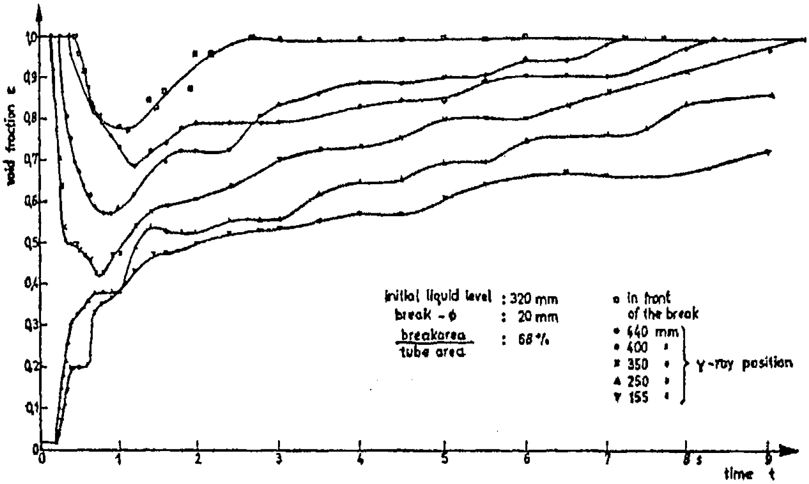


Figure 23.5: Transient void-fraction distribution at different eights of the vessel and in front of the break.

Autor	Medium	Geometrie/ Meßdaten	empirische Beziehung fur den mittleren vol Dampfgehalt in der Mischung	Stromungsformunterscheidung	Druckbereich (bar)
Wilson /17/	H_2O - H_2O Dampf	d_{Beh}: 480 mm 100 mm Sinterplatte eigene Daten /17/	$\langle\bar{\varepsilon}\rangle = C\cdot(Fr)^a\cdot(We)^{0,1}\left[\frac{\rho''}{\rho'-\rho''}\right]^{0,17}$ C = 0,68 a = 0,31 C = 0,88 a = 0,20	$\frac{u_0''}{\left[g\sqrt{\frac{\sigma}{g(\rho'-\rho'')}}\right]^{1/2}} \geq 2$ $\frac{u_0''}{\left[g\sqrt{\frac{\sigma}{g(\rho'-\rho'')}}\right]^{1/2}} > 2$	20 ÷ 41
Sterman Dementiev Lepilin /18/	H_2O- H_2O Dampf	d_{Beh}: 51 mm 69 mm 85 mm 200 mm Verdampfer Sinterplatte Behringerdaten /23/ Margulovadaten /19/	$\langle\bar{\varepsilon}\rangle = C\cdot(Fr)^a\cdot(We)^{0,25}\left[\frac{\rho''}{\rho'-\rho''}\right]^{0,17}$ C = 1,07 a = 0,40 C = 1,90 a = 017	$\frac{u_0''}{\left[g\sqrt{\frac{\sigma}{g(\rho'-\rho'')}}\right]^{1/2}} < 3,7$ $\frac{u_0''}{\left[g\sqrt{\frac{\sigma}{g(\rho'-\rho'')}}\right]^{1/2}} \geq 3,7$	1,07 ÷ 190
Margulova /19/	H_2O - H_2O Dampf	d_{Beh} = 200 mm Sinterplatte eigene Daten /19/	$\langle\bar{\varepsilon}\rangle = (0,576 + 0,00414\,p\,[atm])\cdot(u_0'')^{0.75}$	keine	91 ÷ 190
Kurbatov /20/	H_2O - H_2O Dampf	d_{Beh} = 51 ÷ 200 mm Verdampfer/Sinterpl Behringerdaten /23/ Margulovadaten /19/	$\langle\bar{\varepsilon}\rangle = 0,67\cdot[Fr]^{1/3}[We]^{1/6}\left[\frac{\rho'}{\rho'-\rho''}\right]^{1/3}\left[\frac{v'}{v''}\right]^{2/9}$	keine	1,07 ÷ 190
Labuncov /21/		d_{Beh} = 17 ÷ 748 mm	$\langle\bar{\varepsilon}\rangle = \left(1+\frac{u_{BLS}}{u_0''}\right)^{-1}$ $u_{BLS} = u_{BL}\;\psi_L$ $u_{BL} = 1,5\left(\frac{g\cdot\sigma\cdot(\rho'-\rho'')}{\rho'^2}\right)^{1/4}$ $\psi_L = 1,4\left(\frac{\rho'}{\rho''}\right)^{1/5}\left(1-\frac{\rho''}{\rho'}\right)^5$	Bo > 500	1 ÷ 196
Mersmann /22/	Luft H_2O H_2O-Hg Toluol H_2O		$\langle\bar{\varepsilon}\rangle(1-\langle\bar{\varepsilon}\rangle)^n = 0,14\cdot u_0''\left(\frac{\rho'^2}{g\sigma(\rho'-\rho'')}\right)^{1/4}\cdot\left(\frac{\rho'}{\rho'-\rho''}\right)^{1/3}\cdot\left(\frac{\sigma^3\rho'^2}{\eta'^4(\rho'-\rho'')g}\left(\frac{\rho''}{\rho'}\right)^{5/3}\right)^{1/24}$	$n = f\left(\frac{\rho'}{\rho''}\right)$ Diagramm	

mit $Fr = \frac{u_0''^2}{g\sqrt{\frac{\sigma}{g(\rho'-\rho'')}}}$; $We = \frac{\sqrt{\frac{\sigma}{g(\rho'-\rho'')}}}{d_{Beh}}$; $Bo = g\cdot d_{Beh}^2\cdot\left(\frac{\rho'-\rho''}{\sigma}\right)$; ψ_L = Korrekturfaktor fur Wechselwirkung zwischen den Blasen nach Labuncov

Figure 23.6: Equation for phase separation

behaves somewhat different which seems to be influenced by the Froude-number (see Figure 23.8). The correlation for predicting the void fraction, therefore must be used with two different sets of the empirical factor C and the exponent n, depending whether the Froude-number is greater or smaller than 3. Also the behaviour

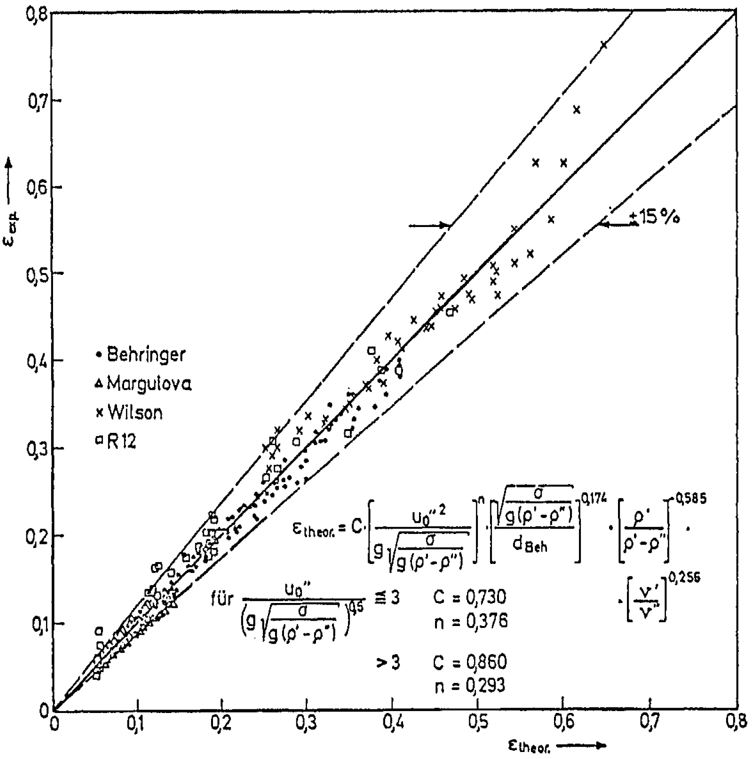


Figure 23.7: Comparison of experimental data measured at the IfV (Freon 12) and data of Margulova, Wilson and Behringer with empirical equation.

of the drift velocities undergoes a change at a Froude-number of approximately 3. As illustrated in Figure 23.9, also the drift velocities can be correlated by a similar equation like the void fraction. In addition, however,the diameter and the height of the vessel as well as the mean bubble diameter have to be taken into account in the correlation. In the Froude-number u_0'' is the superficial velocity of the vapour in the vessel if no liquid at all would be present.

For calculating the phase separation and the depressurization during blowdown, the correlation by Viecenz (1980) in a somewhat different way can be used too, however, there has to be performed an iterative calculation procedure between the predictions of the phase separation, the critical mass flow rate in the nozzle and the depressurization.

If the pressurized system is not a geometrically simple vessel but consists of a complicated arrangement of apparatus, pipes, pumps and valves, predicting the blowdown becomes much more complicated because additional phenomena occur. The pump may - intentionally or accidentally - not be stopped and then its head ratio can influence the conditions in the system. The effect of

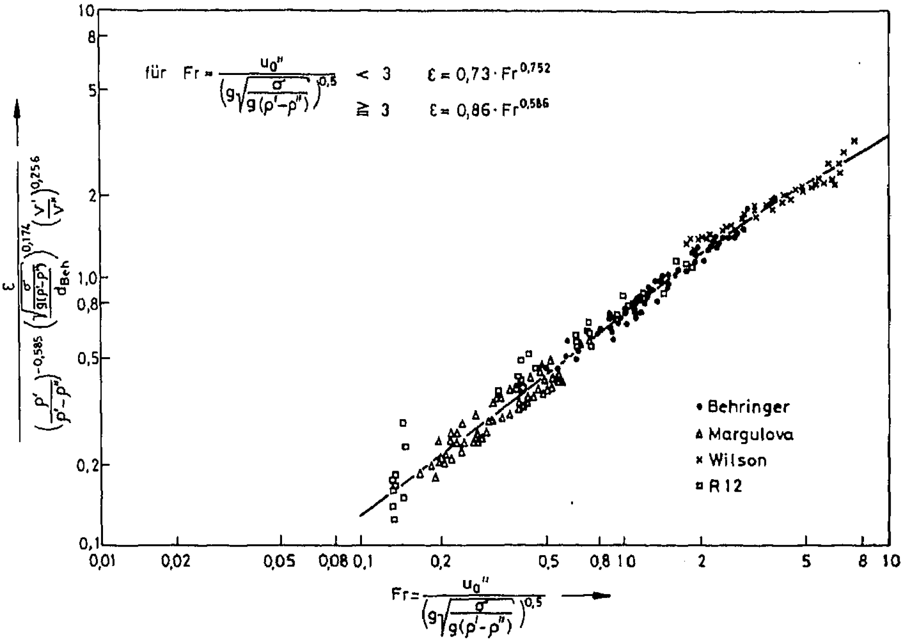


Figure 23.8: Void fraction as a function of shape of flow.

void fraction on the homologeous head ratio measured in water (Olson (1974)) is illustrated in Figure 23.10. It can be seen that the head ratio at a void fraction of approximately 0.2 starts to decrease rapidly, however, the pump still can transport a certain amount of mass.

In a complicated arrangement of different apparatus and other components, the flow at the nozzle is not continuous and the flow pattern may change suddenly as illustrated in Figure 23.11. This is due to the fact, that the different apparatus depending on the pipeline connections are not blown out and evacuated simultaneously and therefore periods of high void may suddenly change with almost pure liquid flow at the nozzle or at the break. So the flow pattern vary between annular-, dispersed- and pure gas flow.

In the pipes connecting the various apparatus liquid carried in the vapour flow may be de-entrained which also influences the thermo- and fluiddynamic conditions at the nozzle. However, also the opposite phenomenon occurs, namely that liquid flowing in form of an annulus at the wall, is entrained by the high shear stresses resulting from the high vapour velocity. Measurements by Langner (1978) demonstrated, that the rate of entrainment becomes larger with increasing acceleration i.e. with a faster depressurization and a shorter blowdown (see Figure 23.12).

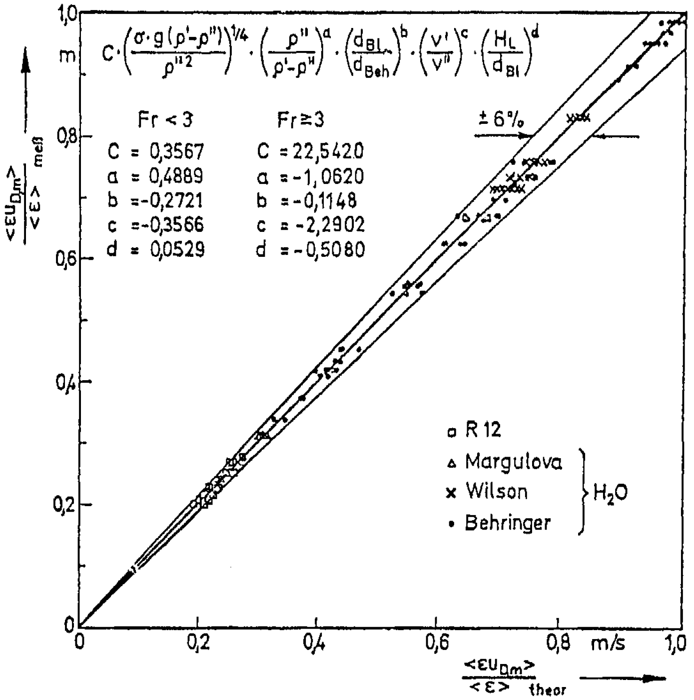


Figure 23.9: Comparison of experimental drift velocities with an empirical equation for data measurement at ifV (Freon 12) and data by Margulova, Wilson and Behringer.

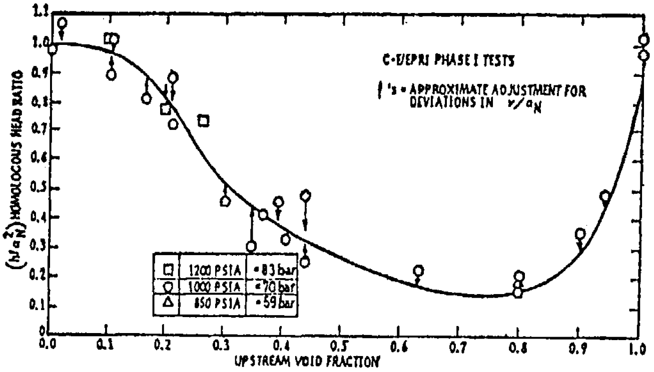


Figure 23.10: Effect of void fraction on homologeous head ratio for rated speed and near rated flow (N_s = 1200).

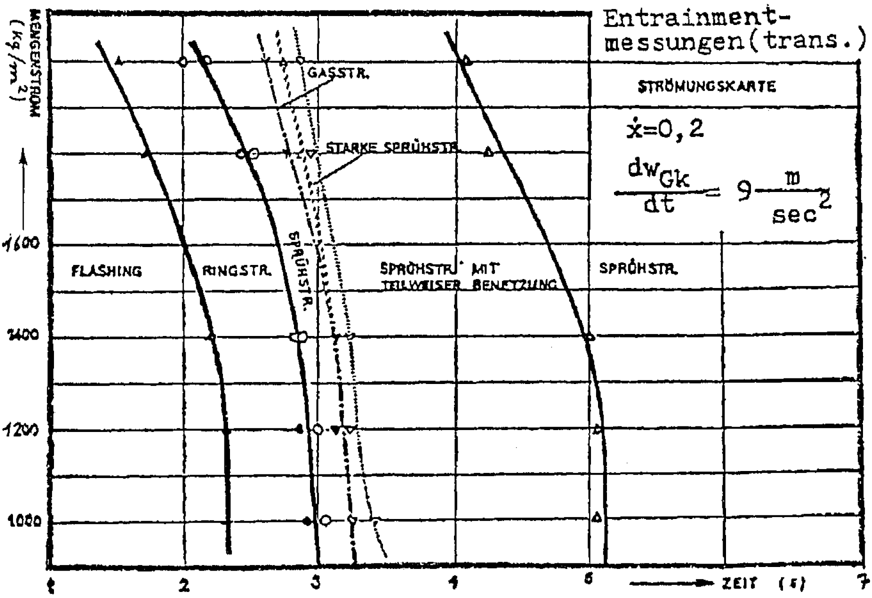


Figure 23.11: Flowpattern during blowdown.

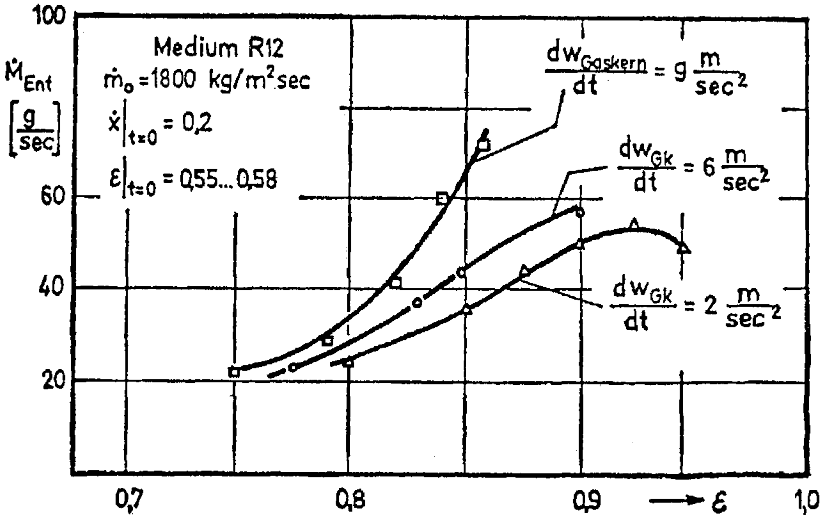


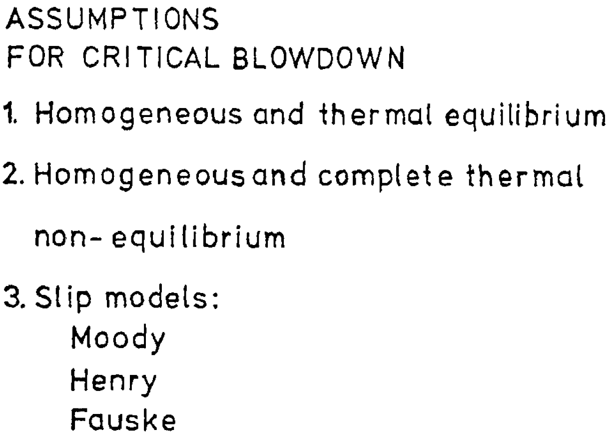
Figure 23.12: Entrainment during blowdown.

23.4 **Critical Flow**

From a certain pressure ratio on in single phase gas flow the velocity in the nozzle remains constant at the value of the sonic velocity, which is also called Laval-velocity. This maximum velocity can easily be deduced from the laws of thermodynamics. In two-phase- gas liquid- mixtures the situation is more complicated. It's not fully correct to speak about a sonic velocity because the travelling velocity of small pressure disturbances - which is the sonic velocity - is different in the liquid and in the gas phase. One usually speaks in two-phase flow about a critical mass flow rate. The velocity in each phase, the quality and the density distribution influences the critical mass flow rate strongly. At critical and super critical pressure ratios there is a strong and sudden depressurization in a short nozzle and thermodynamic dis-equilibrium may also play an important role.

The flow conditions in a nozzle with critical flow are difficult to be measured and therefore theories describing the flow phenomena and predicting the critical flow rate content a number of assumptions. Three classes of assumptions are demonstrated in Figure 23.13. The first assumption - homogeneous and thermal equilibrium flow - imputes that the thermodynamic and the fluid-dynamic state is completely equalized between the phases. The other extreme is the assumption of frozen flow without any momentum and heat exchange between the phases. Between these two situations any condition is conceivable for example homogeneous flow, but complete thermal dis-equilibrium or slip models, as presented by Moody (1965), Henry (1970) or Fauske. It would go beyond the frame of this chapter to discuss in detail critical flow slip models. Here only a few of the important physical phenomena, influencing the critical flow, shall be discussed.

Let us first consider a short nozzle with a small L/D ratio. In case of a very fast depressurization and with saturated or slightly subcooled liquid at the entrance of the nozzle, the



ASSUMPTIONS
FOR CRITICAL BLOWDOWN

1. Homogeneous and thermal equilibrium
2. Homogeneous and complete thermal non-equilibrium
3. Slip models:
   - Moody
   - Henry
   - Fauske

Figure 23.13: Assumptions for critical blowdown.

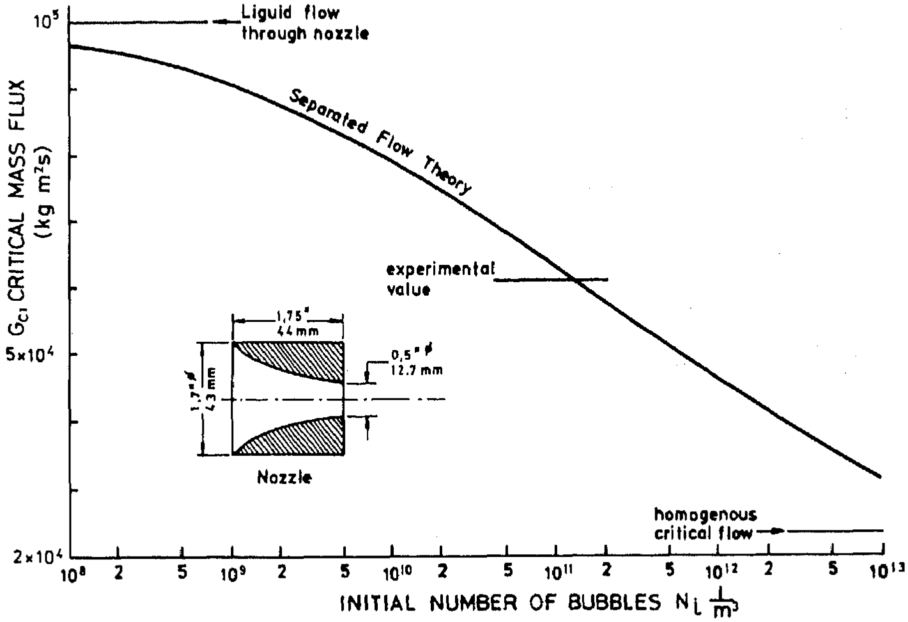


Figure 23.14: Effect of nucleation site density on prediction of short nozzle data of Sozzi-Sutherland.

number of the nuclei being present in the liquid, plays an important role for flashing by expansion and by this for the critical flow rate. Figure 23.14 shows that assuming pure liquid phase pathing through the nozzle the highest mass flow rate could be gained. With increasing initial number of the bubbles or nuclei per unit of volume the critical flow rate decreases and the minimum value would be predicted by a theory assuming homogeneous and equilibrium flow.

From this flashing behaviour it is easily understood that the length of the nozzle influences the critical mass flow rate strongly too, because with increasing nozzle length the depressurization along the flow path is less rapid and there is more time available for flashing. Boiling and nucleation is less effective with increasing subcooling at the nozzle inlet as demonstrated in Figure 23.15 and therefore the critical flow rate increases with smaller subcooling and higher quality (Henry (1979)).

With a two-phase mixture already being present at the inlet of the nozzle, the nucleation problem is ruled out, however, still one can not expect that the rapid expansion through the nozzle follows a fully thermal equilibrium path. The phases have different densities and the pressure gradient will tend to accelerate the lighter vapour phase more than the liquid. The resulting temperature, free energy and velocity differences cause interface transfer of heat, mass and momentum. These interface processes determine the thermodynamic and fluiddynamic paths followed by each phase in the expansion.

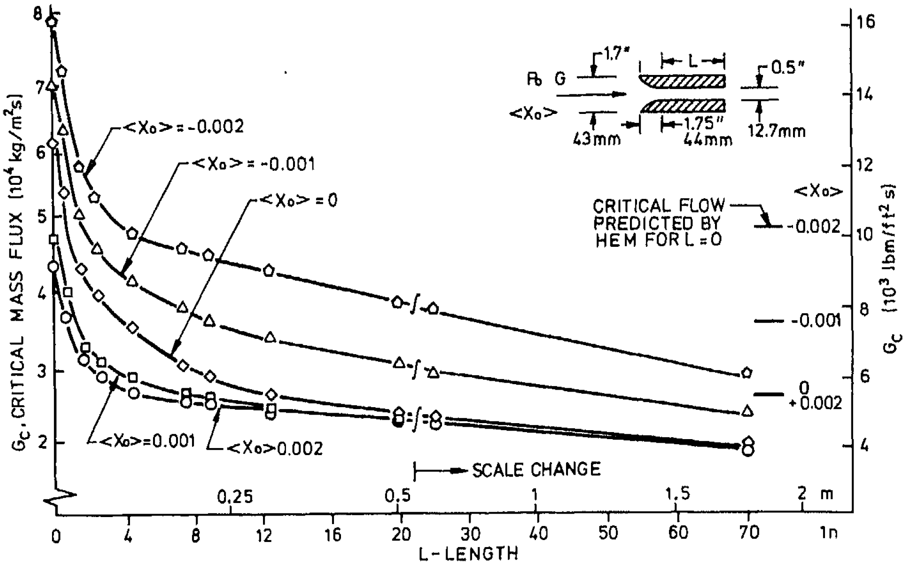


Figure 23.15: Effect of flow length on critical two-phase flow through 0.5 inch diameter tubes (Sozzi-Sutherland)

If there is a long and narrow pipe between the pressure vessel, undergoing the blowdown and the nozzle, the situation for critical flow rate can change completely and other phenomena then discussed up to now, may govern the flow conditions. Friction may play now an important role and its influence may become so large that the flow rate at the nozzle is not determined by the critical flow conditions there, but by the resistance in the pipe between the pressure vessel and the outlet to the atmosphere.

The mass flow rate in the nozzle is not only important for predicting the depressurization and the loss of liquid in the vessel, but also for calculating the momentum forces acting on the outstream geometry. If long pipes are not well fastened the momentum of the outstream jet may push them out of their holding structure and the therefrom resulting whiping effect may destroy other pipelines and escalate the accident.

23.5 Pressure Suppression

Poisoning gases or gas liquid mixtures must not be blown out into the atmosphere. With explosive components it must be guaranteed that by mixing with air a rapid rarefication takes place that no clouds would be formed which can later on explode.

The expansion, distribution and the mixing of a vertical free jet in the air is a function of the jet momentum and the phase distribution at the nozzle outlet of the flashing effects in the jet and of the buoyancy forces between the mixture and the air. With increasing momentum of the jet at the nozzle outlet, the jet diameter is smaller along its path and it has a stronger penetration into the air. The mass mixed with air and so forming an explosive system is proportional to the third power of the nozzle diameter and inverse proportional to the 1,5th power of the jet outlet momentum (Gärtner (1978)). With increasing distance from the nozzle, buoyancy forces in the jet gain more influence. With hydrocarbons one has to be aware of the fact, that the density of their gases is usually higher than that of air and therefore if the momentum in the jet is used up the mixture starts to flow down again. At this point the concentration of hydrocarbons in the mixture should be so low that ignition can be avoided.

Another possibility to prevent additional impacts from the accident to the environment is the quenching of the mixture flowing out from the vessel in a pool of cold and not burnable liquid where the gas is condensed. Direct contact condensation, as demonstrated in Figure 23.16, may cause serious instabilities with respect to pressure and to the liquid level. Only at low vapour fluxes there is a smooth interface between both phases and no chugging occurs.

With steam blown into a water pool under certain flow rates and at low pressures very strong condensation pulses, as demonstrated in Figure 23.17, were observed (Chan et al (1977/8)). There is a close and dynamic coupling between the compressible volume in the vessel and in the pipe where the vapour comes from and the liquid pool where it is condensed. These pressure pulses act onto the walls of the pressure subpression chamber and may cause some damage there. To calculate the local forces onto the wall from the measured pressure pulses is not quite simple because the response of the wall has to be known especially with respect to its flexibility. Highest forces are predicted assuming a

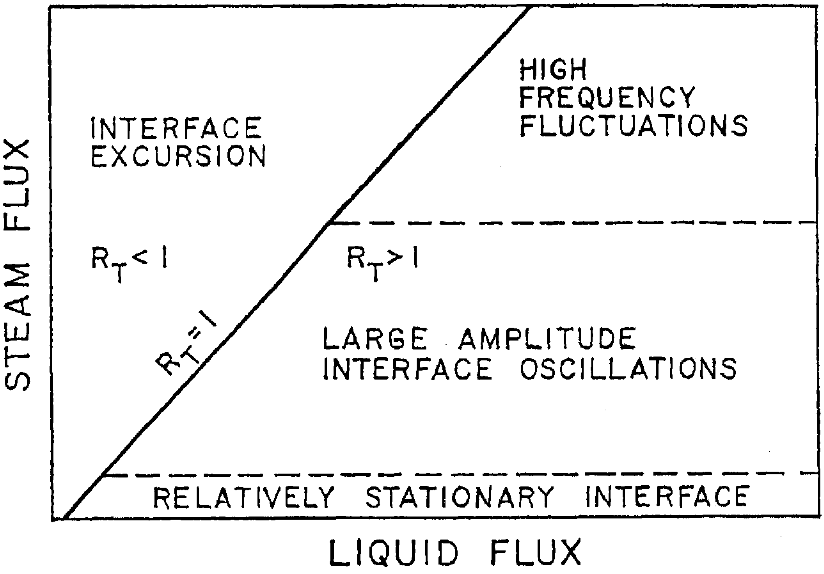


Figure 23.16: "Universal" regime map for direct contact condensation.

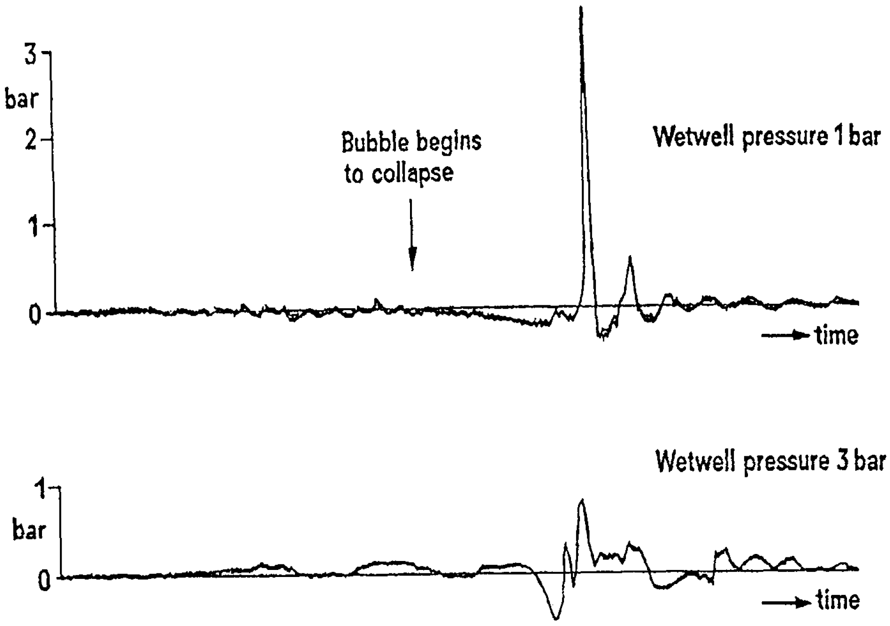


Figure 23.17: Pressure subpression system strong condensation pulses in a stiff and narrow tank at atmospheric and at elevated pressure.

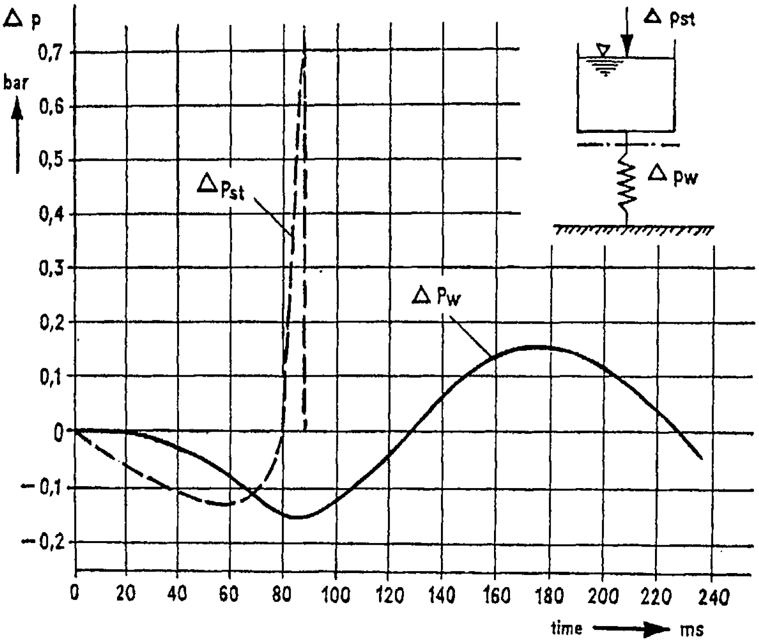


Figure 23.18: Bubble collapse: Calculated pressure transient p_{st} for a rigid tank and transformation p_w on a flexible wall.

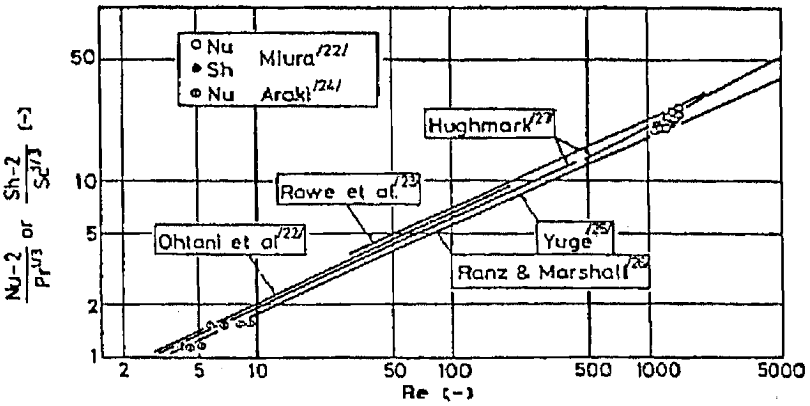


Figure 23.19: Relationship between Re and $(Nu-2)/Pr^{1/3}$ or $(Sh-2)Sc^{1/3}$ Comparison by Miura et al. (1977).

rigid wall (see Figure 23.18). The condensation pulses become smaller with increasing pressure in the pressure subpression system.

23.6 Heat and Mass Transfer

For predicting chemical reactions during a blowdown, but also for estimating the probability of explosive clouds formation, the heat and mass transfer between the phases should be known. Unfortunately our knowledge in this field - especially under fast transient conditions - is very poor (Mayinger (1978)).

Information is available in the literature about interfacial transfer in annular flow and in dispersed flow. The more interesting flow pattern for accident deliberations is the dispersed flow, where small liquid droplets are transported in a gas core. Experiments and theories in the literature show a simple relationship between the Nusselt- or Sherwood-number and the flow conditions, as demonstrated in Figure 23.19. The heat and mass transfer is quite moderate especially for small droplets and even if under transient conditions the Nusselt- or Sherwood-numbers would be greater by one order of magnitude compared to the values in Figure 23.19, the time would be too short for a considerable heat transport or chemical reaction.

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