Predictive Accuracy of Design Methods for the Relief Cross-Sections on Chemical Reactors in the Case of Two-Phase Flow Discharge*

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1 Introduction

For the calculation of the minimum relief cross-section of rupture discs or safety valves on chemical reactors, several so-called simplified methods are described in the literature. An essential feature of these methods is that during relief only a critical two-phase flow consisting of a gas and liquid is assumed. The design criterion is the allowable pressure in the reactor after the opening of the relief device. The discharged total mass flow rate and the composition of the two-phase flow in the relief device then are predefined.

The application of the methods is based on the type of pressure increase in the reactor in the case of an exothermic, thermal runaway reaction. For vapor pressure systems, the pressure rises as the temperature of the reaction mixture increases by the progressive chemical conversion. For gas systems, the pressure rises above the operation pressure from the production of an inert gas, for example, in the course of a decomposition reaction. Reaction systems which exhibit both behaviors simultaneously or successively are considered as hybrid. The simplified methods are specific to the respective reaction type according to the above classification.

Without exception, only the methods and their accuracy developed for vapor pressure reaction systems are presented. A description of the self-adjusting thermodynamic and fluid dynamic states during the pressure relief process, in principle indispensable for a sound appreciation of the obtained results, is given, e.g., by L. Friedel and G. Wehmeier [1].

In the following, the most common methods for calculating the minimum relief cross-section are discussed. Furthermore, the predictive accuracy of these methods will be assessed. For this, either the (formal) reaction kinetics or the characteristic reaction state at prescribed pressures during relief must be available.

2 Design Methods

The methods from the literature can be divided into simplified and transient methods. A simplified method is according to L.S. Kirch et al. [2] an approach for the calculation of the relief diameter which does not contain any dynamic simulation of the pressure relief course and which, therefore, needs no formal reaction kinetics. That means, these methods include some simplifications to reduce the necessary characteristic input values and to simplify the complex calculation. An overview of the commonly used simplified methods is shown in Tab. 1.[1)] They are based on an energy and a mass balance for the reactor at the set pressure and partly on the maximum pressure. Without exception in all the methods as an assumed precautionary measure, a homogeneous liquid level swell of the reaction mixture is included. Indeed, the mass flow quality of the two-phase mixture then can be calculated from the masses of the liquid and the vapor in the closed reactor. This quality is identical for all methods and is assumed to remain constant during the whole relief process like the mass flow rate which, however, may differ for each method. In accordance with the degree of the submodel development, the mass and energy balances included in each method have been extended by considering an incomplete discharge of the reactor contents or an energy release due to the vented vapor. For example the original method by W.J. Boyle [3], which is solely based on mass balance, has been extended by H.K. Fauske [5] by the introduction of the critical mass flux of a two-phase flow. This method, furthermore, contains now the additional assumption that after reaching the interim maximum pressure and the associated temperature increase during the following relief phase, a complete phase separation occurs at the free boundary interface in the reactor dome. This implies that the reactor contents would not further swell up. In comparison to the original method by W.J. Boyle, the necessary relief area will be, thus, reduced, because it is no longer assumed that the reactor is emptied completely during the pressure relief course.

Table 1. Quasi-stationary emergency relief device area design methods for tempered reaction systems.

Method		Relief area
W.J. Boyle,	1967	$m_0/(\dot{G}\,\Delta t)$
H.K. Fauske,	1984	$m_0 \left(\frac{dT}{dt}\right)_s / (\dot{G}_{crit}\,\Delta T)$
H.K. Fauske,	1989	$m_0 \left(\frac{dT}{dt}\right)_s (\alpha_D - \alpha_0) / (2\,\dot{G}_{crit}\,\Delta T\,(1 - \alpha_0))$
H. K. Fauske,	1995	$0{,}12\ V_R \left(\frac{dT}{dt}\right)_{max}$ foaming/viscous reaction systems
		$0{,}013\ V_R \left(\frac{dT}{dt}\right)_{max}$ low viscous reaction systems
J.C. Leung,	1986	$m_0 \left(\frac{1}{2} c_{vf} \left[\left(\frac{dT}{dt}\right)_s + \left(\frac{dT}{dt}\right)_{max}\right]\right) \Big/ \left(\dot{G}_{crit} \left(\left(\frac{V_R}{m_0}\frac{h_{fg}}{v_{fg}}\right)^{\frac{1}{2}} + (c_{vf}\,\Delta T)^{\frac{1}{2}}\right)^2\right)$
J.E. Huff,	1982	$\frac{1}{\dot{G}_{crit}} \left(\frac{m_0}{\Delta t} - \frac{V_R\,h_{fg}}{2\,\dot{Q}\,v_{fg}\,\Delta t^2} \left[\left(1 + 4\,m_0\,\Delta t \frac{\dot{Q}\,v_{fg}}{V_R\,h_{fg}}\right)^{\frac{1}{2}} - 1\right]\right)$

Contrary to the simplified methods shown in Tab. 1, in the equation given by J. Fründt [9], which is also based on a mass and simplified energy balance, the assumption of a homogeneous level swell is no longer maintained, as this would lead

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1) List of symbols at the end of the paper.

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in the case of nonfoaming systems with low liquid phase viscosity to a largely oversized relief area. The mass flow quality is instead calculated by assuming a churn turbulent level swelling by introducing a characteristic bubble swarm rise velocity for the bubbles in the bubbling reaction mixture.

For the calculation of the critical mass flux alternatively the Homogeneous Equilibrium Model, e.g., according to VDI-Wärmeatlas [10], the so-called Equilibrium Rate Model by H.K. Fauske [4], the ω-Method according to J. C. Leung [11] or the Delayed Homogeneous Flow Equilibrium Model according to R. E. Henry and H. K. Fauske [12] is used as sub-model. A more detailed description of the design methods can be found in L. Friedel and S. Korfmann [13,14].

The presented predictive accuracy of the simplified methods is based on the results of relief experiments in comparatively small reactors with volumes between 2 and 280 L as well as in a production reactor with a volume of 10 m^3. The experimentally validated relief areas of the rupture discs or orifices have been additionally predicted by using the modified version VENT of the computer code SAFIRE [15]. This program allows for a time dependent solution of the mass and energy balance for the closed and vented reactor and is essentially based on the same sub-models as used in the simplified methods, e.g., for liquid level swell and phase separation, critical mass flux as well as heat release. In addition, alternative sub-models have been used during the recalculation if appropriate to the actual phase separation behavior. Both types of methods, indeed, assume an immediate establishment of the thermodynamic equilibrium between the phases. Hence, no intermediate superheating or subcooling is considered; the state properties follow the saturation conditions.

With respect to an integrated design of the vent line system and particularly of the containment, the calculated mass flow quality and the total mass flow rates are also shown and assessed against each other. As a precaution, it is already emphasized here that these averaged values are just figures without any actual physical relevance. This mass flow is either higher or lower but never equal to this equivalent momentaneous flux.

3 Experiments and Predictive Accuracy

For the inter-comparison of the relief diameter calculated with the simplified and the transient methods, a prerequisite is that an equal heat release due to the runaway reaction at the set pressure is used in the methods. For this reason, the actual temperature and pressure rise rates as well as the vapor pressure gradient measured during the relief experiments have been used for the predictions. This is contrary to the recommendations for the application of the simplified methods. Indeed, only in this way could the noticeable heat losses of the laboratory reactors be taken into account. For the prediction of the relief area with the transient method, the formal reaction kinetics derived from an experiment in an adiabatic calorimeter had been adopted and the heat losses of the respective swell reactors had been considered. For the calculation of the relief areas on production size reactors one should keep within the specific recommendation of the authors.

In the experiments, the (first or true) critical flow state always appeared in the orifice used for simulating a rupture disc and for the control of the discharge rate. For the prediction with the simplified methods a discharge coefficient of one has been introduced. This implies that the orifice or the rupture disc represents the substantial flow resistance. However, the prediction of the pressure relief process with the transient method relies on a discharge coefficient being constant during the whole venting process and between 0.5 and unity to take into consideration the mass capacity reduction due to friction in the vent lines of various lengths, although a critical flow condition occurred only temporarily in the orifice.

The experimental results are based on the release of a non-catalyzed methanol/acetic anhydride esterification and a radical initiated vinyl acetate emulsion polymerization runaway reaction. In detail, the esterification reaction was carried out in a VSP/Phi-Tec reaction calorimeter and in reactors with volumes of 1.7, 14, and 105 L. Additionally, the results from experiments at the Wilhelm-Jost-Institute e.V. in a reactor with a volume of 280 L [16] and at the Christian-Michelsen Research Institute in a reactor with a volume of 10 m^3 [17] were accessible. The liquid phase in the reaction mixture is relatively low viscous and a so-called homogeneous (chemical) conversion took place. The emulsion polymerization has also been investigated in the VSP/Phi-Tec reaction calorimeter and in a reactor with a volume of 10 L. This reaction system contains a highly viscous liquid phase. So the influence of the viscosity of the liquid phase on the liquid level swell and phase separation and, therefore, the impact on the design of the relief area could be investigated. The emulsion polymerization can be classified as a heterogeneous liquid/liquid reaction.

In Tabs. 2 and 3, the parameters of the pressure relief experiments are shown. The interim pressure rise above the relief pressure during the relief phase was between 2 and 140 %. In this context, any prediction for an experiment with an interim over pressure of more than about 30 % is a non-authorized extrapolation of the model. For the predictions, only recently published design methods, i.e., the equations by H. K. Fauske (3 methods), J. C. Leung, and J. Fründt have been used, implicitly assuming a higher degree of inclusion of new findings. The only exception is the model by J. E. Huff in honor of his long standing activity in this field and his contribution to the development of codes. Additionally, the results produced with the code SAFIRE/VENT are shown for comparison purposes, by using case by case the so-called churn turbulent flow phase separation model according to I. Kataoka and M. Ishii [18] and to DIERS and the homogeneous flow level swell model originally included in SAFIRE.

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Table 2. Experimental and predicted relief diameter as a function of the relief condition for a non-catalyzed methanol/acetic anhydride exterification runaway reaction.

Reactor volume [l]/ Initial liquid level [%]	Relief condition Pressure [bar] Pressure overshoot [%]	Relief diameter [mm]	Predicted relief diameter [mm] Fauske (1984)	Fauske (1989) α_D=1/0,5	Fauske (1995)	Leung	Huff	Fründt u_∞=0/0,05 m/s	SAFIRE/VENT Phase separation model Churn turbulent flow Kataoka/ Ishii	SAFIRE/VENT DIERS	Discharge coefficient [–]
			Discharge coefficient = 1 [–]								
1.7/60	11.0/ 9	2.5	14.8	10.5/ 4.3	13.2	5.8	4.3	7.0/ 4.1	3.1	3.2	
14 /70	11.0/ 5	10.0	53.2	37.6/ 20.1	32.8	19.6	17.8	21.1/ 17.4	14.1	16.8	
14 /70	11.0/ 17	7.1	36.2	25.6/ 13.7	41.1	18.5	14.4	20.1/ 16.4	11.0	12.0	1
105 /62	7.5/ 17	27.1	81.2	57.4/ 25.2	70.9	56.5	55.7	45.3/ 34.7	40.2	37.8	
105 /62	6.1/ 2	27.1	215.0	152.0/ 66.9	59.9	126.3	128.5	90.2/ 80.1	44.7	45.4	
105 /62	8.0/115	20.0	31.4	22.2/ 9.8	75.8	38.0	46.2	37.7/ 29.0	22.1	22.7	
280 /90	2.1/100	15.0	8.6	6.1/ 4.3	29.4	19.9	23.2	25.2/ 15.0	17.9	16.1	0.8
280 /90	2.0/140	13.0	12.7	9.0/ 6.3	22.3	19.4	21.6	21.4/ 11.3	16.9	16.3	0.5
10^4 /80	7.4/ 30	150.0	376.2	266.1/162.9	545.8	298.2	294.0	299.0/257.6	163.2	178.0	0.9

Table 3. Experimental and predicted relief diameter as a function of the relief condition for a radical initiated vinyl acetate runaway emulsion polymerization.

Initial liquid level [%]	Relief condition Pressure [bar]/ Pressure overshoot [%]	Relief diameter [mm]	Predicted relief diameter [mm] Fauske (1984)	Fauske (1989) α_D=1/0,5	Fauske (1995)	Leung	Huff	SAFIRE/VENT Phase separation model Kataoka/ Ishii	SAFIRE/VENT Homogen	Discharge coefficient [–]
			Discharge coefficient = 1 [–]							
40	5.9/ 5	8	33.5	23.7/16.8	56.5	13.8	11.5	2.9	9.5	0.8
80	3.1/19	6	10.0	7.0/ 4.3	17.3	7.3	7.9	4.1	7.9	0.6
60	4 /23	6	10.7	7.6/ 3.1	25.9	7.3	8.3	4.9	7.2	

In general, with the methods of H. K. Fauske (rupture disc) diameters are calculated which are approximately two to eight times larger than the values actually needed according to the experiments (Tab. 2). This results from the case that with these methods it is conservatively assumed a complete reaction mixture discharge during the relief phase when the highest pressure is reached. With the method of H. K. Fauske (1989), it is also possible to take into account that actually only a part of the reaction mixture is discharged. This can be mathematically realized by defining a final liquid level or vapor fraction in the reactor after ending of the relief process. In the calculation extreme values of unity and 0.5 have been used for the volumetric vapor fraction α_D in the reactor at the beginning of the complete phase separation. Indeed, this value must be provided by the user on the basis of his experience. When using a value for α_D equal to unity, diameters will be calculated which are approximately two to five times larger than the experimental result. With values of α_D smaller than unity, a partial phase separation during the level swell and, therefore, a lower mixture discharge will be presumed. This leads here to more realistic results for the relief diameter, but it can also lead in individual cases to an undersizing if too small values for α_D are used. Indeed, the undersized diameters in this tabulation are due to the irregular extrapolation of the method. The diameters predicted with the method by H. K. Fauske (1995) are still two to six times larger than the actual diameter, even though here a partial phase separation has been taken into account. However, in this method the initial filling level in the reactor can not be changed and a completely filled reactor is the basis in the method.

The design methods which are only based on a mass balance [4–6] include an extent of validity for the interim pressure increase above the set pressure of up to 20 or 30 %. In some experiments these values have been exceeded, so that in these cases the prediction with the quasi-stationary methods formally was not permitted. This led to the consequence that the otherwise regular distinct overprediction of the relief cross-section, according to the relief condition, led to an underprediction of the relief cross-section.

With the methods by J. E. Huff and J. C. Leung approximately equal and systematically smaller relief diameters are calculated than with the methods by H. K. Fauske. The predicted diameters are, however, still oversized. The comparatively lower oversizing can be explained by the fact that the energy discharged with the vented vapor phase is in addition taken into account.

The calculation of the relief diameter in accordance with the method of J. Fründt was carried out using a characteristic bubble swarm rise velocity of 0 and 0.05 m/s. This value must also be provided by the (expert) user. For an insignificant rise velocity, which corresponds to a homogeneous level swell, diameters in the range of the results predicted with the methods by J. C. Leung and J. E. Huff are predicted. If a velocity of 0.05 m/s is used, clearly smaller diameters are calculated, which are, however, still about by factor of three larger than the experimentally deduced diameters. By indifferent use of this bubble rise velocity, an undersizing of the relief diameter in the case of an, in this method, unlimited pressure overshoot can also result.

The relief diameters calculated by using the code SAFIRE/VENT with the assumption of a partial phase separation independent of the unlimited size of the interim pressure overshoot are always only insignificantly larger than the experimentally derived values and closer to the experimental results than these calculated with the simplified methods.

The tabulation of the calculated diameters for the runaway emulsion polymerization of vinyl acetate reveals that in this case generally larger diameters are also calculated (Tab. 3). The overprediction is here less than in the case of the esterification reaction. This can be attributed to the actually occurring homogeneous level swell of the reactor contents due to the highly viscous liquid phase in the reaction mixture in the experiments. Again, with the code SAFIRE/VENT only

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insignificantly larger relief diameters are predicted than experimentally estimated. On comparing only the experimental results, where a small temporary pressure overshoot occurred, a systematic high overestimation of the relief diameters can be observed (Tab. 2 and 3). On the other hand, for the esterification reaction, diameters are predicted with the model by H. K. Fauske (1984), which are only 1.6 times larger than the actual diameter, although incorrect assumptions about the mixture mass discharge and the phase separation are to some extent included here. An underprediction of the relief area is also possible, e.g., with the method by H. K. Fauske (1989) on the basis of the assumption of a final vapor fraction of 50 % in the reactor, and with the method by J. Fründt in the case of introducing a characteristic bubble swarm rise velocity of 0.05 m/s. This is the consequence of the arithmetically too large temporary pressure overshoot and, hence, the overpredicted mass flux as well as the too small heat release ascribed to the reaction in this phase of the relief. The other simplifying suppositions obviously cancel each other to a large extent, so that a relatively correct or a too small of a diameter is calculated. J. Fründt [19] therefore recommends using the largest heat release of the reaction during the relief phase for a large interim pressure overshoot, a procedure similar to that proposed by J. C. Leung. In total, in view of the degree of accuracy obtainable, the preference of the transient method compared to that of the simplified method is obvious, although the necessary effort involved for the calculation of the relief diameters is clearly higher and more costly.

With regard to the integrated design of the vent line and particularly of the containment, the amount and the transport concentration of the vented mass flow rate are depicted in Tabs. 4 and 5. These are by definition constant during the complete relief time. The results according to SAFIRE/VENT are not given, because in the transient simulation the mass flow rate is calculated for every time step and therefore varies. A comparison, at least, between the calculated and the measured integral results is also not possible here, since the total mass discharge has not been measured. The predicted results, however, would be only physically meaningful if calculated with SAFIRE/VENT, because the mass flow rates predicted with the simplified methods are only equivalent, fictitious quantities, leading only mathematically to the same pressure relief. But it is not known, whether larger mass flow rates than these computed with the simplified methods can, nevertheless, occur because of other conservative suppositions.

In general, depending on the choice of the design method, the relief areas calculated for each of the two reaction systems, will be larger than it is actually required for the postulated specific blow down scenario. In connection with this overprediction inevitably an at least temporarily larger mass flux must be handled in the containment, thus, also making an oversized disposal system necessary, which in the actual case would work with an unexpected low efficiency.

With respect to the predictions of the experiments with the transient method, for reasons of simplicity, a discharge coefficient between 0.5 and unity and constant during the whole pressure relief process has been used to take into account the throughput reduction due to the flow resistance in the vent line according to the respective experimental arrangement. Against this, for the calculations with the simplified methods also for simplicity, a constant discharge coefficient of unity during venting was introduced. This allowed for a sound intercomparison of the simplified method predictions, because it was not possible to take into consideration a throughput reduction due to friction in the long vent lines with all methods. Indeed, the statement referring to the magnitude of the overpredictions and the differences in relation to each other remain valid, because in the case of application of those discharge coefficients used in the transient calculation to the results according to the simplified methods will lead to a further and proportional enlargement of the vent area.

4 Conclusion

The relief diameters calculated with selected, simplified methods are as a rule considerably larger than the experi-

Table 4. Predicted two-phase flow vapor mass fraction and mass flow rate for a non-catalyzed methanol/acetic anhydride esterification runaway reaction.

Reactor volume [l]/ Initial liquid level [%]	Relief condition Pressure [bar]/ Pressure overshoot [%]	Relief diameter [mm]	Mass flow quality [%] Homogeneous level swell	Churn-turbulent flow	Mass flow rate [kg/s] Fauske (1984)	Fauske (1989) α_D=1/0.5	Leung	Huff	Fründt u_{0c}= 0/0.05 m/s	Ratio largest/smallest Mass flow rate
1.7/60	11 / 9	2.5	0.36	100.0	2.1	1.1 / 0.2	0.3	0.2	0.5/ 0.2	11
14 /70	11 / 5	10.0	0.23	3.9	26.9	13.4 / 3.9	3.3	2.7	4.2/ 2.9	10
14 /70	11 / 17	7.1	0.23	7.7	12.4	6.2 / 1.8	3.0	1.8	3.8/ 2.5	7
105 /62	7.5/ 17	27.1	0.33	6.9	46.1	23.0 / 4.4	20.1	11.3	14.6/ 8.6	10
105 /62	6.1/ 2	27.1	0.33	5.0	206.3	103.1 / 20.0	64.1	66.3	36.3/ 28.7	10
105 /62	8 /115	20.0	0.33	22.9	6.9	3.4 / 0.7	9.1	13.4	11.0/ 6.5	20
280 /90	2.1/100	15.0	0.008	12.9	0.09	0.05/ 0.02	0.36	0.49	0.8/ 0.3	25
280 /90	2.0/140	13.0	0.007	42.3	0.16	0.08/ 0.04	0.28	0.35	0.6/ 0.2	9
10^4 /80	7.4/ 30	150.0	0.02	0.9	641.5	320.8 /120.0	362.6	352.4	405.0/300.6	5

Table 5. Predicted two-phase flow vapor mass fraction and mass flow rate for a radical initiated vinyl acetate runaway emulsion polymerization.

Initial liquid level [%]	Relief condition Pressure [bar]/ Pressure overshoot [%]	Relief diameter [mm]	Mass flow quality [%] Homogeneous level swell	Mass flow rate [kg/s] Fauske (1984)	Fauske (1989) α_D=1/0.5	Leung	Huff	Ratio largest/smallest mass flow rate
80	5.9/ 5	8	0.06	4.2	2.1/1.1	0.6	0.4	11
40	3.1/19	6	0.18	0.2	0.1/0.04	0.1	0.1	5
60	4 /23	6	0.11	0.3	0.2/0.03	0.1	0.2	10

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mental results. However, the predictive accuracy of these design methods is higher if in the experiments the assumptions included in the methods are met and the validity range of these methods are kept. Due to the overprediction of the relief cross-sections in the case of a relief through a rupture disc a higher mass flow rate could also occur for the assumed worst case scenario than calculated. For this mass flow rate, the disposal system would possibly then not be adequately designed. Also, by using the transient method regularly, an overprediction of the relief diameters occurs. Indeed, the calculated values are closer to the experimental results. Therefore, with these methods a more balanced prediction of the safety device and the containment system is achievable. Nevertheless, in the available codes simplified assumptions are also still included, which can only be replaced by submodels validated by pressure relief experiments with runaway reaction systems.

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Symbols used

c_p	[J/kg K]	specific heat at constant pressure
c_v	[J/kg K]	specific heat at constant volume
G	[kg/m^{2}s]	mass flux
h_{fg}	[J/kg]	specific heat of evaporation
m	[kg]	mass
$\dot{Q}$	[W]	heat flow
t	[s]	time
T	[K]	temperature
U_∞	[m/s]	characteristic bubble swarm rise velocity
v_{fg}	[m^3/kg]	difference in specific volume of vapor and liquid phase
V	[m^3]	Volume
α	[–]	initial reactor void fraction

Subscripts

0	initial
D	phase separation
f	liquid
crit	fluiddynamic critical condition
max	maximum
R	reactor
s	relief set condition

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