

Prevention of fog in the condensation of vapour from mixtures with inert gas, by a regenerative thermal process

Stephan Kaufmann*, Karl Hilfiker**

FHZ (ZTL), Lucerne Engineering College, Technikumstrasse, CH-6048 Horw, Switzerland

(Received 14 April 1998, accepted 25 August 1998)

Abstract — In processing, vapours often have to be separated from mixtures mainly containing inert gas, as for example in the cleaning of exhaust air and the recovery of pollutant solvent vapours (volatile organic compounds, VOC). Whenever possible, condensation is applied for this task, since it is thermodynamically the most energy-efficient process. It can be carried out successfully, provided that the vapour can be liquefied and removed separately. However, fog often forms through *spontaneous condensation* in the gas flow. The very small aerosol drops are difficult to remove, and not all can be separated. Accordingly, there are limits to the separation of the gas–vapour mixture. The fact that mass and heat transfer occurs simultaneously at the cooling surface explains why the main flow of the gas–vapour mixture is supersaturated and fog can form from it. A method of preventing supersaturation and consequently avoiding fog has now been found. A relatively small amount of heat is applied to the mixture during condensation to prevent a spontaneous change of phase. This is accomplished by *regenerative* means. The vapour condenses only at the cooling surface, where the condensate drains off by itself. There is no need to filter off small condensed droplets of fog. Experiments have shown that far better separation is achieved by this *fog prevention* method; more condensate is recovered and there are fewer pollutants in the cleaned exhaust gas. Some particularly interesting applications are water-cooling and condensation of hot exhaust gases from a reaction, or low-temperature condensation in exhaust air cleaning and solvent recovery. © Elsevier, Paris.

fog / mist / regenerative / prevention / vapour / inert gas / mixture / condensation / heat and mass transfer

Résumé — **Empêchement thermique régénératif de la formation de brouillard lors de la condensation de vapeurs constituées de mélanges avec un gaz inerte.** Dans la technique des procédés, il faut souvent procéder à une séparation des vapeurs à partir de mélanges comportant une proportion prédominante de gaz inerte, par exemple lors de la décontamination des émissions gazeuses et la récupération des vapeurs de solvant, chargées en produits nocifs pour l'environnement (*volatile organic compounds*, VOC). Partout où cela est possible, on doit utiliser à cette fin la condensation ; d'après les lois de la thermodynamique, c'est le processus qui est énergiquement le plus avantageux. Ainsi, on peut effectuer la séparation dans la mesure où l'on peut liquéfier la vapeur et l'évacuer séparément. Souvent, il se constitue un brouillard par *condensation spontanée* dans le flux gazeux. La séparation de ces très petites gouttes d'aérosol est difficile et ne peut se faire qu'incomplètement. De manière correspondante, la séparation du mélange gaz–vapeur est limitée. Avec le transfert simultané de matière et de chaleur qui s'effectue sur la surface de refroidissement, on peut mettre en lumière le fait que l'écoulement principal du mélange gaz–vapeur est sursaturé et que, à partir de cela, il peut y avoir formation de brouillard. On a à présent élaboré une méthode permettant d'empêcher cette sursaturation et d'éviter de ce fait la formation de brouillard. Pendant la condensation, on ajoute au mélange une quantité de chaleur relativement petite, qui empêche tout changement de phase spontané. Ceci s'effectue de façon *régénérative*. La vapeur se condense seulement sur la surface de refroidissement, là où le condensat s'écoule de façon séparée. Il n'est pas nécessaire d'effectuer une séparation par filtration des petites gouttelettes de condensat du brouillard. Il a été prouvé lors d'essais que, avec cette méthode d'empêchement de la formation du brouillard, on atteignait des rendements de séparation nettement meilleurs ; la quantité de condensat récupérée était plus grande et il y avait moins de polluants dans les gaz d'échappement épurés. Pour ce qui concerne l'application, sont particulièrement intéressants par exemple le refroidissement et la condensation de gaz d'échappement de réaction très chauds, ceci étant fait avec de l'eau de refroidissement, ou bien la condensation à basse température lors de la décontamination des émissions gazeuses et la récupération de solvants. © Elsevier, Paris.

empêchement / régénératif / aérosol / brouillard / condensation / vapeurs / mélange / gaz inerte / séparation / transfert simultané de matière et de chaleur

* Present address: Bertrams Ltd., Chemical Engineering, Eptingerstrasse 41, CH-4132 Muttenz, Switzerland.
verkauf@bertrams.ch

** Correspondence and reprints.

Nomenclature

A	area	m^2
a	thermal diffusivity	$\text{m}^2 \cdot \text{s}^{-1}$
a_1, a_2, a_3	coefficients	
B	mass charge	$\text{kg} \cdot \text{kg}_{\text{gas}}^{-1}$
c_p	specific heat	$\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$
\tilde{c}_p	molar specific heat	$\text{J} \cdot \text{kmol}^{-1} \cdot \text{K}^{-1}$
D	diffusion coefficient	$\text{m}^2 \cdot \text{s}^{-1}$
\dot{H}	enthalpy flow rate	$\text{J} \cdot \text{s}^{-1}$
k	over-all heat-transfer coefficient	$\text{W} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$
L	characteristic length	m
Le	Lewis number, $\lambda/\rho c_p D$	
\dot{m}	mass flow rate	$\text{kg} \cdot \text{s}^{-1}$
Nu	Nusselt number $\alpha L/\lambda$	
\tilde{n}	molecular density	$\text{kmol} \cdot \text{m}^{-3}$
\dot{n}	molar flow rate	$\text{kmol} \cdot \text{s}^{-1}$
p	pressure	Pa
$p_s(T)$	vapor pressure at the temperature T	Pa
Pr	Prandtl number, $\eta c_p/\lambda$	
r	specific enthalpy of evaporation	$\text{J} \cdot \text{kg}^{-1}$
\tilde{r}	molar specific enthalpy of evaporation	$\text{J} \cdot \text{kmol}^{-1}$
\mathcal{R}	gas constant	$\text{J} \cdot \text{kmol}^{-1} \cdot \text{K}^{-1}$
Re	Reynolds number, $L v \rho/\eta$	
S	degree of saturation, $p_v/p_{v,e}$	
Sc	Schmidt number, $\eta/(\rho D)$	
Sh	Sherwood number, $\beta L/D$	
T	absolute temperature	K
y	molar fraction	$\text{kmol} \cdot \text{kmol}_{\text{mixture}}^{-1}$
Y	molar charge	$\text{kmol} \cdot \text{kmol}_{\text{gas}}^{-1}$
z	coordinate	m

Greek symbols

α	heat-transfer coefficient in the boundary layer	$\text{W} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$
β	mass-transfer coefficient in the film boundary layer	$\text{m} \cdot \text{s}^{-1}$
Δ	difference	
δ	boundary layer thickness	m
η	dynamic viscosity	$\text{Pa} \cdot \text{s}$
λ	thermal conductivity	$\text{W} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$
ρ	density	$\text{kg} \cdot \text{m}^{-3}$
ϑ	temperature on centigrade scale	$^{\circ}\text{C}$

Subscripts

0	total, sum
1	bulk of vapor-gas mixture
2	interface vapor-gas mixture to condensate
3	interface condensate film to wall
4	interface wall to coolant
5	bulk of the coolant
I	entry of the condensation process

α	heat transfer
ae	aerosol, fog particles
CH	complementary heat
d	diffusional mass transfer
de	demand, deficit
e	equilibrium
G	gas
M	vapor-gas mixture
V	vapor

1. SEPARATION OF VAPOUR FROM MIXTURES WITH INERT GAS BY CONDENSATION

Mixtures of vapour and a high proportion of non-condensable components—that is, the inert gas—often have to be separated in the processing industry. For example, the recovery of vapours from volatile organic solvents (VOC) has been practised for several decades for economic reasons. As awareness of environmental pollution caused by these substances has grown, statutory limits for cleaning exhaust air have been set.

In industry the inert gas is frequently nitrogen, which is employed to fill tanks, reactor vessels and the like above the liquid in order to prevent explosive mixtures forming with the oxygen from the ambient air. The nitrogen becomes saturated with vapour from volatile organic substances, which then have to be separated.

Condensation permits the recovery of solvent vapours in the proximity of the exhaust air source and prior to their mixing with other flows or auxiliary substances, thereby promoting their re-use. It is the most energy-efficient process according to thermodynamic principles.

Condensers may also be used for explosive mixtures, which they can clean to below the lower ignition limit. In drying circuits too, vapour is condensed out of the moist gas-vapour mixture and the gas can again be used for drying. As condensation only calls for simple apparatus and large amounts of vapour can be recovered efficiently, the method is widely employed, frequently in combination with other cleaning processes.

The actual condensation process separates only the vapour that liquefies on a cold surface and drains off by itself. However, spontaneous condensation often takes place in the main flow, and fog forms. The small drops of condensate are suspended in the gas-vapour mixture, which is why they are commonly described as condensation aerosols. In this case the gas-vapour mixture can be separated to the extent only that the fog drops can be extracted. Drop separators consisting e.g. of wire mesh located in the flow path of the condensed gas bring about a pressure drop, a risk of obstruction and complicated maintenance. Drops of only a few mm

or less cannot be completely removed. An attempt will now be made to condense all the vapour exclusively on the cooling surface, where the condensate drains off by itself.

2. FOG FORMATION IN CONVENTIONAL CONDENSERS

Great importance attaches to condensation aerosols in the Earth's atmosphere because mist and cloud arise in this form. These processes have been studied widely and comprehensively in respect of water vapour/air mixtures. In contrast, the condensation processes to be found in technical applications involve totally different mixtures of substances. The pressure and temperature ranges are also very different and there is a wider spectrum of foreign particles initiating fog formation. In this research project, fog formation in condensation has therefore been studied experimentally on a pilot scale.

What interested us in our first experiments was: under what processing conditions is fog formed? For this purpose a heat exchanger with three-tiered helical cooling tubes was employed (*figure 1*). The cooling tubes were enclosed in a glass shell through which the gas-vapour mixture was conducted in an axial direction and perpendicular to the cooling coils. This condenser was run in a test plant. The process flow sheet of this plant, with another type of condenser, is discussed and illustrated in § 5.

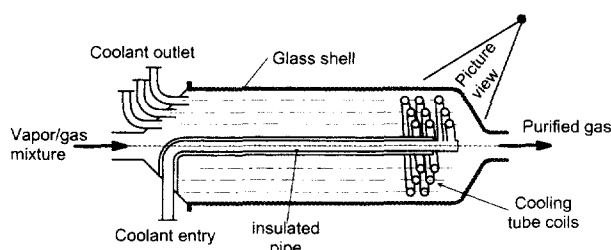


Figure 1. Sketch of the test condenser for inquiries into the main influences on fog formation. It was used in the installation according to *figure 9*, instead of the condenser shown there. Dimensions: outside diameter of cooling tube 12 mm; coil diameters 65 mm, 97 mm and 128 mm; inside diameter of the glass shell 151 mm; cooling area 1.5 m².

Pure nitrogen was charged with a solvent and heated to the desired entry temperature. The composition could be set so that the mixture entered the condenser either in a saturated or only partly saturated, i.e. overheated, state. The coolant entry temperature was also varied and controlled by the coolant flow rate, so that the temperature of the exit coolant could also be set. Thus it was possible to set the temperature difference between the gas-vapour mixture and the countercurrent coolant.

The two photographs in *figure 2* were taken at the end of the cooling area, i.e. at the outlet of the purified gas: the left one in a case with only slight fog formation, while the right-hand side shows the same section with dense fog. Tests were also carried out at different overall pressures, which showed minor influence on fog intensity.

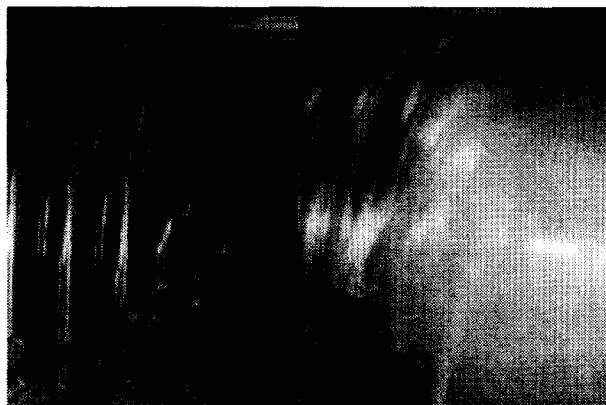


Figure 2. Fog formation during condensation of a mixture of ethanol and nitrogen, photographed through the glass shell (view marked in *figure 1*): left with very little fog, right with dense fog.

In order to determine the quantity and size of the condensation aerosols, a measurement was made by laser at right angles through the glass jacket, behind the cooling coils in the free flow of the mixture. The employed laser three-wave-length-extinction measurement and its application are described by Schaber et al. [1]. A flow sample was taken continuously and *isokinetically* from the emerging mixture and the total solvent charge was determined. This is plotted in *figure 3* against the temperature difference ΔT between the gas-vapour mixture entering the heat exchanger (from left in *figure 1*) and the coolant leaving it (at left-hand side in *figure 1*). Nitrogen had been charged with ethanol-water of azeotropic composition (7 % by weight water, 93 % ethanol) for these tests. With a moderate temperature difference ΔT (left-hand side of *figure 3*), the mixture without any foreign particles could be purified almost to equilibrium charge, and virtually no fog was measured or could be perceived. When cooling was intensified, i.e. when the temperature difference ΔT was increased with a higher coolant flow rate (right-hand side of *figure 3*), thick fog formed. The effluent mixture contained a solvent quantity many times higher than the amount corresponding to the equilibrium value at the temperature of the emerging mixture measured.

In order to simulate mixtures with foreign particles, 3 % by volume of cigarette smoke was added continuously upstream of the condenser. Even with a slight temperature difference quite a thick fog formed and the charge in the exhaust gas flow was correspondingly large. It increased further with an increased tempera-

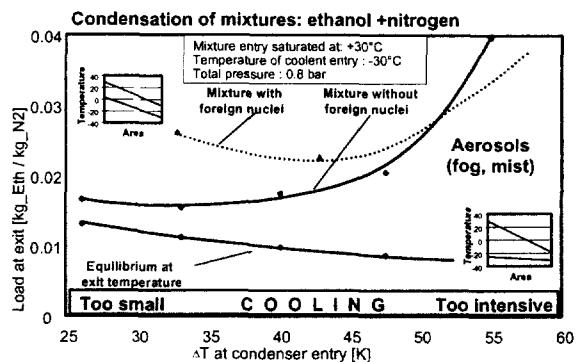


Figure 3. Charge measured at exit of countercurrent condenser, plotted against temperature difference $\Delta T = T_{\text{mixture entry}} - T_{\text{coolant exit}}$, which was altered by various flowrates of the coolant. Only if the mixture contains no foreign particles and with a moderate ΔT (left-hand side of graph) does the charge measured approximately follow the equilibrium change. When ΔT is large (right-hand side) the charge including aerosols is considerably higher, as in mixtures with foreign particles such as the process gas at a chemical plant.

ture difference. It was found that the smoke by itself was not detected by the measuring instruments.

With other solvents tested too, the *temperature difference* exerted the greatest effect on fog formation. The other process parameters that we studied were less significant in terms of fog formation. For example, even superheating the mixture upstream of the condenser by 15 K above dewpoint only brought about slightly reduced fog formation.

We tried to find parameter settings at which larger aerosol drops would form, since this would facilitate subsequent separation. The mean drop size measured for the fog in the same series of tests mentioned so far is entered on the right-hand ordinate in *figure 4*. In the mixture, without any foreign particles, it ranged between 1 and 2.5 μm ; in that with foreign particles, it ranged between 2.6 and 2.7 μm . With the vapours studied (methanol, azeotropic mixture of ethanol and water, toluene, dichloromethane), the drop size was about 2.5 μm or less in most cases. It was not possible to produce drops larger than 3 μm .

In the same test condenser we were able to investigate fog formation in *process gas* from a chemical plant. The gas was taken from the top of a rectification column in which mother liquor from a preceding crystallisation process was destined. Under the same conditions as for mixtures with and without artificial foreign nuclei, fog formation in the process gas was partly stochastic, though, in the majority of cases, it was the same as in the mixture with foreign particles, with fog appearing even with a small temperature difference.

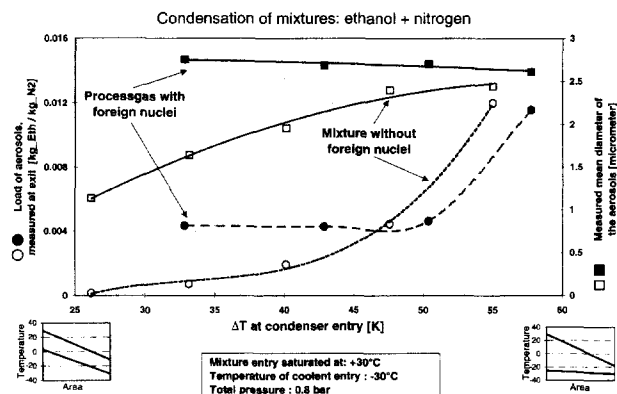


Figure 4. Aerosol charge in lean gas (left ordinate) and mean diameter of condensation aerosols (right ordinate), measured in condenser by laser optics on the three-wavelength extinction principle [1]. The drop diameters were less than 3 μm in all the tests, even with widely varying temperature, pressure and relative saturation at the entry and with different solvents. It was not possible to produce larger drops, which could have been more readily separated mechanically.

3. FOG FORMATION AS A RESULT OF SUPERSATURATION AND NUCLEATION

3.1. Fog-formation phenomena

The prerequisites for fog formation, notably supersaturation and the presence of condensation nuclei, will first be briefly presented. Following this the phenomena leading to the condensation process and the resultant supersaturation will be discussed, and finally its mathematics will be summarised.

A survey of aerosol formation in industrial processes was carried out by Schaber [2]. Fog does not form until the vapour content of the mixture is higher than in the saturated state, that is, until the equilibrium concentration has been exceeded. As a criterion of this supersaturated state, the degree of saturation is defined as follows:

$$S = \frac{p_v}{p_s(T)} \quad (1)$$

The actual partial vapour pressure p_v is compared with that at equilibrium p_e . Consequently, for a gas-vapour mixture it is compared with the boiling pressure $p_s(T)$ of the condensable component at the temperature T of the mixture. The term *supersaturation* is commonly employed for the relationship S according to equation (1).

Supersaturation, however, is not the only prerequisite for the formation of condensation aerosols. Only when nuclei, too, are present do liquid aerosol drops grow on them by the condensation of excess vapour. Two types of nucleation are known: homogeneous and heterogeneous.

For instance, nuclei may be present in the gas-vapour mixture as foreign particles and trigger *heterogeneous nucleation* even if the degree of saturation is only slightly over one, as established by Hinds [3]. This also applies to mist and cloud formation in the atmosphere, where the substances are predominantly salt crystals produced by drops and splashes of sea water evaporating and particles of desert sand carried by the wind across continents. Water vapour condenses on to the solid particles in the atmospheric air. The small drops of moisture thus created may be suspended in the air as a mist for a long time or may grow through condensation and coagulation of a number of several small drops into larger ones and descend as rain drops. In industrial processes, growth by coagulation is limited to drops of only a few μm in size because the dwell times in the compact apparatus are generally short.

The size and number of condensation aerosols also depend on the degree of supersaturation and the number of foreign particles acting as nuclei. Only rarely can the number of nuclei be influenced in practice. The addition of foreign particles is sometimes not allowed, while, in many other cases, the mixture already contains a large number of foreign nuclei.

In cases where there are no foreign particles in the mixture, condensation nuclei may be created spontaneously through *homogeneous nucleation*, providing sufficient vapour molecules combine to form moisture particles capable of growth. This occurs only in the event of a higher degree of supersaturation, e.g. $S > 2 \dots 10$ for various organic solvents, as in the experiments reported by Steinmeyer [4]. In mixtures without any foreign particles fog formation can be prevented by maintaining supersaturation below these levels. In the condensation tests this was accomplished by limiting the temperature difference between the gas-vapour mixture at entry and the coolant. As a rough guideline 20 to 25 K should not be exceeded.

However, should the mixture already contain foreign particles, which is to be expected in many areas of application, these measures will not be sufficient to prevent fog formation. We shall therefore investigate more closely the other precondition, that of *supersaturation*. For the vapour to be separated from a gas-vapour mixture, it must reach the cooling surface, condense on it and drain off by itself either as a film or in a trickle (*figure 5*). The gaseous mixture loses vapour in the region of the liquid interface, but is enriched with the non-condensable component (inert gas).

There is a *concentration gradient* which forms from the bulk flow to the condensation surface, and as a consequence vapour is diffused from the bulk flow onto the cold surface. This *mass transfer* is impeded by the boundary layer enriched with inert gas, which the

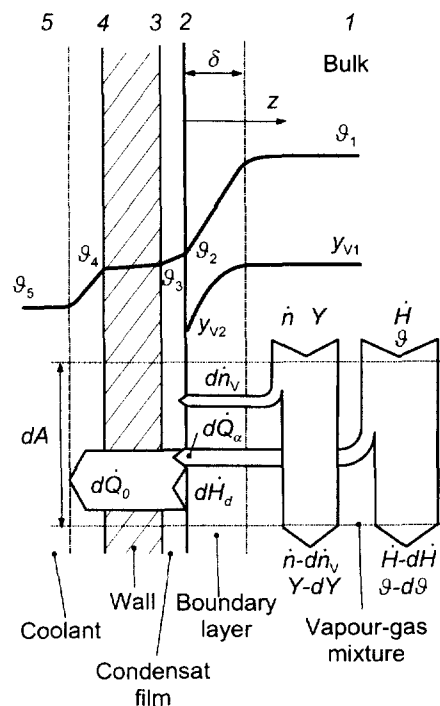


Figure 5. In condensation of a gas-vapour mixture a mass flow rate $d\dot{n}_v$ and a heat flow rate $d\dot{Q}_\alpha$ are simultaneously transferred from the bulk (1) to the phase boundary (2). This results in reductions in temperature $d\vartheta$ and vapour concentration dY in the bulk of the mixture.

vapour has to penetrate before condensing on the cold surface.

As the face temperature is lower than that of the mixture, *heat transfer* occurs simultaneously, thereby cooling the gas-vapour mixture in the bulk flow. That the course of the bulk flow between entry and exit is not isotherm is therefore rather a consequence of the heat transfer and not a precondition for the condensation process. The mixture in the bulk flow need not be cooled to dew point to condense on the cold face. So long as the face temperature is below dew point, vapour will condense, even when the bulk flow is maintained at a constant temperature.

Mass and heat are transferred simultaneously in the same flow field with both processes having a permanent interrelationship with each other. In many cases the gas-vapour mixture cools during condensation to such an extent that the state of the main flow approaches and exceeds saturation, thereby giving rise to supersaturation. A large mass flow with a simultaneous, not too large heat flow would be required in order to prevent it. However, only by coolant temperature optimisation can these combined processes be adjusted slightly in favour of transfer of substance, but supersaturation cannot be completely avoided.

In order to quantify the degree of supersaturation, the changing state of the gas–vapour mixture must be calculated and compared with the saturation state. The combined heat and mass transfer is described for an infinite volume element of the condenser (*figure 5*), following which a balance is drawn for the bulk flow of the mixture to determine its change of state. A steady state process is assumed with the loss of pressure being neglected. The condensate film is taken to flow as a layer and to have a smooth surface. (Investigations on the condensation in presence of a noncondensable gas and a wavy interface were done by Kim and Kang [5]). The enthalpy from cooling of the diffusing vapour and that from subcooling of the condensate are not taken into consideration. Heat and mass transfer are assumed to occur exclusively at right angles to the bulk flow, perpendicular to the cooling surface. For the bulk flow only the temperature-sensitive enthalpy is shown in *figure 5*. An alternative model was presented by West and Hansen [6], but the experimental correlation was obtained with humid air.

3.2. Heat transfer

The following heat flow from the gas–vapour mixture results from the temperature difference between the bulk flow and the phase boundary:

$$d\dot{Q}_\alpha = \alpha_{12} (\vartheta_1 - \vartheta_2) dA \quad (2)$$

The condensation heat

$$d\dot{Q}_d = d\dot{m}_{v,d} r_v = d\dot{m}_{v,d} \tilde{r}_v \quad (3)$$

is released on the surface of the condensation film only and has no direct influence on the temperature in the bulk flow. The sum of these heat flows is conveyed to the coolant:

$$d\dot{Q}_\alpha + d\dot{Q}_d = k_{25} (\vartheta_2 - \vartheta_5) dA \quad (4)$$

The heat transfer coefficient k_{25} covers the resistance of thermal conductivity through the condensate film and cooling wall and heat transfer in the coolant.

3.3. Mass transfer

The diffusing mass flow in the vapour $d\dot{m}_{v,d}$ now has to be determined. As for heat transfer, mass transfer will also be dealt with here in one-dimensionality. According to *Fick's basic law* the mass flow by diffusion for unit area is

$$\frac{d\dot{m}_{v,d}}{dA} = -D_{VG} \frac{d\tilde{n}_v}{dz} \quad (5)$$

where D_{VG} is the diffusion coefficient of the vapour in the gas and $d\tilde{n}_v/dz$ is the concentration gradient. Allowance must be made for the fact that the inert

gas *stagnates* in front of the condensate film and is diffused back counter to the vapour flow, as described by Stefan [7]. With integration through the thickness δ of the diffusion boundary layer we arrive at the following equation for the mass transfer of the vapour in a mixture with a stagnating gas component by *diffusion and convection*:

$$\frac{d\dot{m}_v}{dA} = \frac{D_{VG}}{\delta} \tilde{n}_M \ln \frac{\tilde{n}_M - \tilde{n}_{v2}}{\tilde{n}_M - \tilde{n}_{v1}} = \frac{D_{VG}}{\delta} \frac{p_M}{\Re T_M} \ln \frac{1 - y_{v2}}{1 - y_{v1}} \quad (6)$$

A detailed derivation can be found e.g. in Stefan [7].

As the thickness of the diffusion boundary layer δ is unknown, the *coefficient β for the mass transfer* is introduced in analogy with heat transfer:

$$\beta_{12} = \frac{D_{VG}}{\delta} \quad (7)$$

β_{12} is discussed in the next section employing *Lewis's* analogy. If, in addition, we now apply the molar charge $Y = \dot{m}_v/\dot{m}_G$ as a proportion of concentration and insert it in (6), we obtain:

$$\frac{d\dot{m}_v}{dA} = \beta_{12} \frac{p_M}{\Re T_M} \ln \frac{1 + Y_{v1}}{1 + Y_{v2}} \quad (8)$$

3.4. The analogy of combined heat and mass transfer

We now need the coefficient α_{12} of heat transfer for equation (2) and the coefficient β_{12} of mass transfer for equation (8). Both are dependent upon a number of substance properties and flow conditions. To reduce the number of variables affecting forced convective transfer of heat and substance, dimensionless coefficients for the transfer of heat and substance are employed, containing α and β respectively:

$$Nu = \frac{\alpha L}{\lambda} \quad \text{and} \quad Sh = \frac{\beta L}{D} \quad (9),(10)$$

The Nusselt number Nu and the Sherwood number Sh are only dependent on two variables only: Nu on the Reynolds number Re and the Prandtl number Pr , and Sh similarly on Re and also on the Schmidt number Sc . For a turbulent flow of constant cross-section, simple correlations may be used as in equations (11) and (12) with Sc and Pr representing fluid properties only.

$$Nu = a_1 Re^{a_2} Pr^{a_3} \quad (11)$$

$$Sh = a_1 Re^{a_2} Sc^{a_3} \quad (12)$$

Finally, the ratio of Sc and Pr is introduced and we arrive at the Lewis number Le :

$$Le = \frac{Sc}{Pr} = \frac{a_M}{D_{VG}} = \frac{\lambda_M}{D_{VG} \rho_M c_{pM}} \quad (13)$$

Le is also a dimensionless coefficient and a substance quantity. It is employed to find a relationship between the coefficient β for mass transfer and the coefficient α for heat transfer. By dividing the equations (11) and (12) we obtain the ratio:

$$\frac{Nu}{Sh} = \frac{1}{Le^{a_3}} \quad (14)$$

By employing the definitions of the Nusselt, Sherwood and Lewis numbers (see nomenclature) and equations (13) and (14) we arrive at the following equation for the coefficient β for the mass transfer:

$$\beta_{12} = \frac{\alpha_{12}}{\rho_M c_{pM}} \frac{1}{Le^{1-a_3}} \quad (15)$$

Equation (15) shows that the coefficient for the mass transfer can be calculated from the heat transfer coefficient, and that the two are *proportional* to each other. Consequently, if the mass transfer is intensified through a higher flow velocity and turbulence, heat transfer will also increase. With a turbulent flow, however, the coefficient $a_3 = 0.4$ or more and the ratio β_{12}/α_{12} is therefore higher than with a laminar flow, where $a_3 = 0.33$. This applies to mixtures with $Le > 1$, where the fog risk is greater in any case.

If the density $\rho_M = p/(R_M T)$ is replaced in equation (15) and equation (15) is inserted in (6), we obtain the molar vapour flow discharged per unit area as:

$$\frac{d\dot{n}_V}{dA} = \frac{\alpha_{12}}{\tilde{c}_{pM}} \frac{1}{Le^{1-a_3}} \ln \frac{1+Y_{V1}}{1+Y_{V2}} \quad (16)$$

whereby $\tilde{c}_{pM} = M_M c_{pM}$ is the molar specific heat capacity of the mixture at constant pressure.

With the substance and heat flows withdrawn from the bulk flow, equations (16) and (2), we calculate the change in the state of the gas-vapour mixture for the infinite volume element. From the energy balance:

$$d\dot{Q}_\alpha = -d\dot{H}$$

it follows that:

$$\alpha_{12}(\vartheta_1 - \vartheta_2)dA = \dot{n}_M \tilde{c}_{pM} (-d\vartheta) \quad (17)$$

The enthalpy flow $d\dot{n}_{V,d} \tilde{r}_V$ carried to the phase boundary with the diffusing vapour flow is not considered in this balance because it does not influence the temperature of the bulk flow.

From equation (17) we obtain the temperature drop in the volume element in the direction of the bulk flow:

$$d\vartheta = \frac{\alpha_{12}(\vartheta_1 - \vartheta_2)}{\dot{n}_M \tilde{c}_{pM}} dA \quad (18)$$

By analogy with the mass balance, we bring about the reduction in the molar charge $dY = d\dot{n}_V/\dot{n}_G$:

$$dY = \frac{\alpha_{12}}{\dot{n}_G \tilde{c}_{pM}} \frac{1}{Le^{1-a_3}} \ln \left(\frac{1+Y_{V1}}{1+Y_{V2}} \right) dA \quad (19)$$

Replacing $\dot{n}_M/\dot{n}_G = 1+Y_V$, we divide equations (19) and (18) by each other to produce an important interpretation with this quotient:

$$\frac{dY}{d\vartheta} = \frac{1+Y_{V1}}{\vartheta_1 - \vartheta_2} \frac{1}{Le^{1-a_3}} \ln \left(\frac{1+Y_{V1}}{1+Y_{V2}(\vartheta_2)} \right) \quad (20)$$

The expression for the vapour concentration at the phase boundary may also be described by employing the boiling pressure $p_V(\vartheta_2)$ of the vapour component at the corresponding temperature and the total pressure p :

$$1+Y_{V2} = \frac{1}{1 - \frac{p_V(\vartheta_2)}{p}} \quad (21)$$

Equation (20) represents the *gradient of the process line* in a Y, ϑ graph (figure 6). Integration to obtain the process line is generally performed numerically. If constant material properties in a simplified form for a mean composition, and if a constant phase boundary temperature ϑ_2 is assumed, algebraic integration is possible.

However, the most important question in respect of supersaturation is: how great is the process gradient $dY/d\vartheta$ compared with the gradient of the saturation line $dY_s/d\vartheta$? For many mixtures the process gradient proves to be smaller than that of the saturation line.

Consequently, the state of a gas-vapour mixture approaches the saturation line, crosses it, and is then in the supersaturated region. This was already established by Brouwers and Chesters [8]. Further, they pointed out

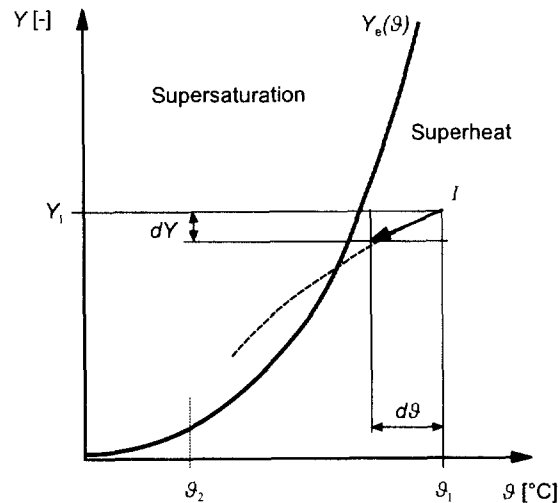


Figure 6. In the charge versus temperature diagram the process line (broken line) shows the states through which the gas-vapour mixture passes between the entry I and exit II of the condenser. If they are above the saturation line, fog may form. The process gradient $dY/d\vartheta$ depends mainly on the condensation temperature and on particle sizes.

that under certain conditions, supersaturation and thus fog formation can take place in the boundary layer, even if the bulk flow is superheated.

If the temperature at the phase boundary is raised in equation (20) so that the temperature difference $\vartheta_1 - \vartheta_2$ becomes smaller, a greater process gradient will be formed. In this way, the risk and extent of supersaturation may be reduced. This finding tallies with the observation made in the tests and described above. In the case of a saturated mixture, however, the process gradient only approaches the equilibrium gradient as the temperature difference disappears. If a non-saturated (superheated) gas-vapour mixture is condensed, there is an optimum phase boundary temperature; this is described in more detail by Kaufmann [9].

The previous finding is that the state of the mixture in the condenser approaches saturation and exceeds it in many instances. Generally, this phenomenon cannot be prevented by adjusting the mass transfer independently of heat transfer.

Detailed consideration of this finding is given in figures 6a-c, which represent the temperature-sensitive energy flows at a infinite volume element for the transfer area dA . At this element, the heat flow $d\dot{Q}_\alpha$ is removed from the bulk flow. Starting from a saturated entry state and with adjustment at the element according to the simultaneous heat and mass transfer, the resultant state at the exit is generally $d\dot{H}_{de}$ below saturation. Three consequences are possible.

- *Supersaturation without fog formation (figure 7a):* the enthalpy deficit $d\dot{H}_{de}$ in the bulk flow is maintained, resulting in a temperature below saturation temperature in the bulk flow.

- *Fog formation (figure 7b):* this happens when sufficient nuclei are present in the bulk flow, either through heterogeneous or for homogeneous nucleation. Drops of liquid (aerosols) are formed and the enthalpy deficit is compensated for by the condensation heat $d\dot{n}_{ae}\tilde{r}_D$.

- *Compensating for the enthalpy deficit $d\dot{H}_{de}$ by supplying heat (figure 7c):* this last possibility will be discussed further in the following section.

4. PREVENTION OF FOG BY REGENERATIVE HEAT SUPPLY

4.1. The principle: the supply of a small amount of heat prevents supersaturation in the condenser

Calculated examples show that supersaturation is generally inevitable in the condensation of vapours from

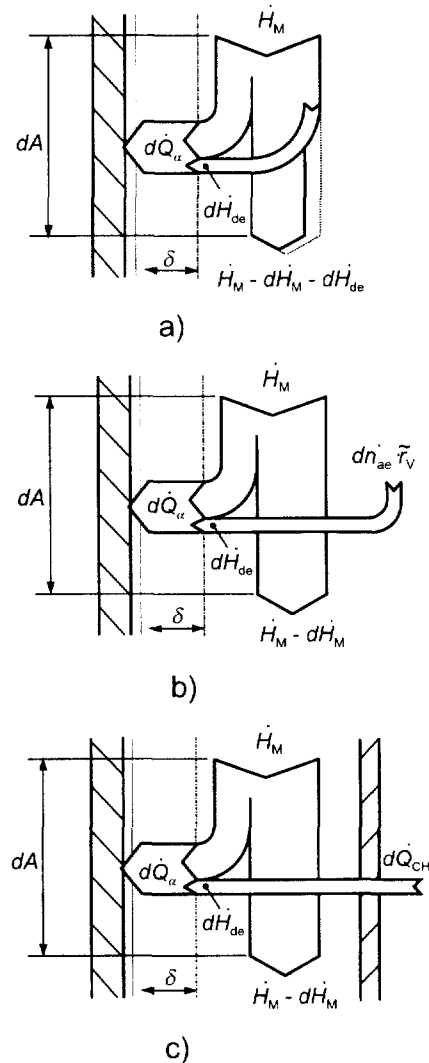


Figure 7. Assuming a saturated state at the entry, the change in temperature and vapour content at the exit, produced by the simultaneous heat and mass transfer, results in a state generally $d\dot{H}_{de}$ below saturation. There are three possible consequences: a) *supersaturation without fog formation:* the enthalpy deficit is maintained and the temperature in the bulk flow is lower than saturation temperature; b) *fog formation:* if sufficient nuclei are present in the bulk flow, drops of liquid (aerosols) are formed and the enthalpy deficit is compensated for by the condensation heat $d\dot{n}_{ae}\tilde{r}_D$ released; c) *enthalpy deficit $d\dot{H}_{de}$ compensated for by supplying heat:* thus no supersaturation (no super-cooling) occurs and fog is prevented.

mixtures with inert gas. The combination of simultaneous heat and mass transfer cannot be overruled to avoid supersaturated states, however the energy balance may be suitably adjusted by supplying a complementary heat flow.

Figure 7c is a flow diagram showing the temperature-sensitive enthalpy for an infinite volume element with a transfer area dA . If the bulk flow is assumed to be saturated at both entry and exit, the result of drawing up a mass and energy balance is an enthalpy deficit:

$$d\dot{H}_{de} = \dot{m}_G dh_{de} \quad (22)$$

To prevent the bulk flow from dropping below the saturation line, a relatively small, *complementary heat flow*

$$d\dot{Q}_{CH} = d\dot{H}_{de} \quad (23)$$

must be supplied to the infinite volume element. It has to compensate for the excess of energy in terms of the combined heat and mass transfer taken from the bulk flow. It could also be said that the mixture is re-heated from a supercooled state to saturation temperature.

The effectiveness of this complementary heat supply for preventing fog formation during condensation has already been demonstrated by Colburn and Edison [10]. It applies regardless of whether any foreign particles are present in the mixture, because condensation aerosols cannot form without supersaturation in either case. A suitable method of supplying the complementary heat in industrial applications must now be developed.

4.2. The concept of internal regenerative heat transfer

Condensers for industrial applications involving fog prevention should not call for the use of an external heat source. The energy required should be recovered internally by precooling the relatively warm mixture entering the condenser. Figure 8 outlines the principle of such energy recovery with corresponding temperature profiles. The mixture entering at I is precooled to a lower temperature at II, while the heat flow \dot{Q}_{CH} is transferred to the bulk of the condensation channel from III to IV. Following precooling, the mixture is passed from II to the condensation entry at III. The cooling heat flow \dot{Q}_{1tot} causes the vapour to condense and cools the mixture down further from III to IV. The relatively small (compared to \dot{Q}_{1tot}), complementary heat flow \dot{Q}_{CH} prevents supersaturation and thus fog formation in this zone. Some of the vapour may also condense in the precooling zone.

4.3. The concept compared to conventional condensation with fog separation

The prevention of fog formation causes the vapour to condense exclusively on the cooling surface, where the condensate drains off by itself. Thus more condensate is recovered and the inert gas is cleaned more effectively without the necessity for any drop separators.

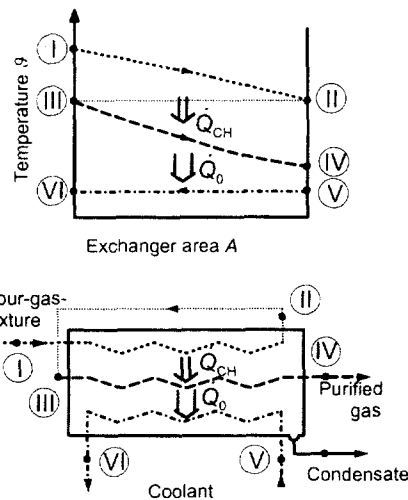


Figure 8. New concept for thermal prevention of fog during condensation: while the heat flow rate \dot{Q}_0 is extracted from the condensation zone from III to IV and passed to the coolant (VI to V), vapour condenses and the gas-vapour mixture cools down from III to IV. In order to prevent supersaturation and fog formation a small heat flow rate \dot{Q}_{CH} is supplied. This is accomplished regeneratively by precooling and pre-condensing the gas-vapour mixture from I to II.

Cleaning of exhaust gases by condensation reduces emissions of substances that pollute the environment. If condensation can be accomplished without fog formation in a multi-stage process, minute moisture droplets will not be transferred to later stages, thereby preventing resultant damage such as corrosion or deposits in pipes and plant. The additional expenditure in condensers with fog prevention is the heat-exchange area, which is required between precooling and the condensation zone. It can be a very simple structure, since it has only to withstand the slight pressure difference caused by pressure drop at the precooling stage.

In contrast to that, in conventional condensation fog droplets of only a few μm in size may form, and very fine-mesh filter structures are required to separate these. These cause a loss of pressure and may become dirty and clogged, involving additional servicing and cleaning. This is not necessary if fog is prevented, and the flow cross-section for the mixture remains unobstructed, thereby offering advantages particularly for multiple-product plant such as is used in biotechnology, pharmaceuticals or the food industry.

Fog prevention with regenerative cooling is self-adjusting over wide ranges of throughput. Higher flowrate creates an increasing need for complementary heat in the condensation zone. In addition, the heat flowrate increases automatically because this higher mass flowrate passes through the precooling. The fog is still prevented even with a partial load. In conventional condensers, however, particularly dense fog forms at low

throughput and is not satisfactorily removed because normal drop separators are less efficient at a lower flowrate.

5. TEST RESULTS FOR THE CONDENSER WITH FOG PREVENTION

A simple geometry was selected for the test condenser (*figure 9*). Four concentric tubes form the three annular flow channels, namely for precooling (outermost), condensation (middle) and coolant (inner). A V-shaped duct is inserted at the bottom of the condensation channel, in which the condensate can drain off without running onto the heated outer surface. The gas-vapour mixture enters the outermost annular gap (the precooling zone) at the right-hand side and passes through it to the left. From there it is returned externally and passed into the condensation zone on the right-hand side. In that zone vapour condenses on the inner tube, while heat from precooling comes in from the outer tube and prevents fog formation. On the left-hand side the cleaned gas is withdrawn through holes around the circumference, and the condensate flows along the duct to the left end and into the condensate pipe. The coolant flows through the plant in the opposite direction, from left to right. For comparison with conventional condensation a "no precooling" mode is also possible, with the mixture being passed directly into the condensation channel.

The temperature and composition of the mixture were measured both at the entry and exit of each channel and at five positions along the flow path. The tests that gave the results reported below were carried out under the following conditions. A mixture of $14.8 \text{ kg}\cdot\text{h}^{-1}$ nitrogen and $0.94 \text{ kg}\cdot\text{h}^{-1}$ isopropanol vapour was introduced into the condenser at 24.7°C . Condensation nuclei were mixed with 3 % by volume of cigarette smoke immediately before the condenser. The total pressure in the gas-vapour mixture was 0.899 bar. $1\,058 \text{ kg}\cdot\text{h}^{-1}$ liquid methanol flowed into the condenser at minus 8.4°C as the coolant.

Results for the overall condensation process with regenerative precooling for fog prevention are given in *figure 10* and compared with those from the corresponding test without precooling, i.e. with conventional condensation (values in brackets). 8.5 % more condensate was obtained with precooling, namely $0.293 \text{ kg}\cdot\text{h}^{-1}$ ($0.270 \text{ kg}\cdot\text{h}^{-1}$). $0.023 \text{ kg}\cdot\text{h}^{-1}$ was condensed in the actual precooling channel. Given the small cooling surface in the test apparatus the mixture could only be cooled to 7.0°C (5.1°C), so the charge of the cleaned gas that could be obtained was not very deep; it was $0.0439 \text{ kg}\cdot\text{kg}_{\text{N}_2}^{-1}$ ($0.0457 \text{ kg}\cdot\text{kg}_{\text{N}_2}^{-1}$). The 4 % reduction in charging therefore seems relatively small. With a separation rate of

$$A = (B_{\text{entry}} - B_{\text{out,eff}})/(B_{\text{entry}} - B_e(T_{\text{out}})) \quad (24)$$

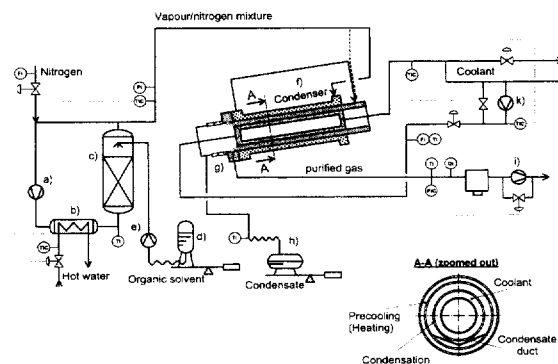


Figure 9. Diagram of installation for tests on fog prevention during condensation of vapour from mixtures with inert gas: a) circulating fan, b) heater, c) evaporation column, d) solvent tank, e) dosing pump, f) test condenser, g) aerosol laser measuring unit, h) Condensate tank, i) vacuum pump, k) cooling circuit with circulating pump. Regenerative fog prevention with four concentric tubes in the test condenser: the gas-vapour mixture flows first through the outermost annular gap (the precooling zone) then through the middle one (the condensation zone). The inner annular gap contains the coolant. The condensate is drained out of the condensation channel in the V-shaped duct, so that it does not run onto the heated outer tube.

the charge reduction obtained is compared with the reduction which would have been obtained given an equilibrium charge at the temperature measured on exit. The separation rate rose by a third with fog prevention and by 12 to 25 % in other tests.

The decisive factor in preventing fog is that the degree of saturation at exit (of $S = 1.23$) can be lowered to $S = 1.04$. In *figure 11* this is plotted against the flow distance in the various channels. In the "no precooling" mode the area where fog forms is marked. With precooling, its actual state and also that in the actual condensation channel are virtually always below saturation ($S < 1$), so no fog forms in the main flow. However, vapour simultaneously condenses on the cooling surface and is thus effectively separated from the gas.

6. FINAL REMARKS

Fog often forms during conventional condensation of vapour from mixtures containing a major proportion of inert gas, sometimes considerably reducing the amount of condensate recovered and the purity of the gas. In conventional condensers fog formation can only be reduced by maintaining a small temperature difference between the gas-vapour mixture and the coolant, and can only be avoided altogether, providing the mixture is free of foreign condensation nuclei. The latter restriction is not fulfilled in the majority of industrial applications,

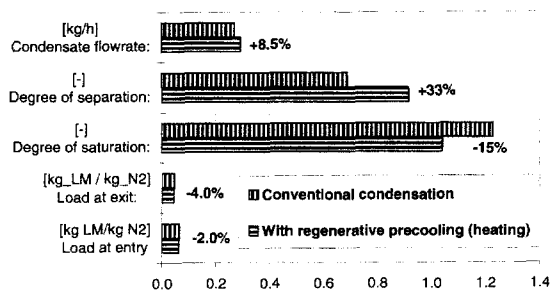


Figure 10. Comparison between condensation with regenerative fog prevention and conventional condensation: mixture of $0.94 \text{ kg}\cdot\text{h}^{-1}$ isopropanol and $14.8 \text{ kg}\cdot\text{h}^{-1}$ nitrogen enters the condenser at 24.7°C and 0.899 bar . Coolant temperature: -8.2°C , cooling area 0.38 m^2 . Temperature at gas exit: with fog prevention 7.0°C , without precooling 5.1°C .

because minute foreign particles are present in the mixture and trigger fog formation even with a slight temperature difference. Incidentally, mist formation in the earth's atmosphere is also triggered by smallest solid particles.

The novel method described permits the total prevention of fog in condensation processes, regardless of any foreign particles in the mixture. A relatively small heat flow is fed into the gas-vapour mixture during condensation, thereby preventing supersaturation of the mixture and consequently fog formation. The heat is supplied regeneratively through precooling of the incoming, warm gas-vapour mixture. This condensation method with fog prevention produced larger condensate flows and cleaner exhaust gases in the test than conventional condensation under the same conditions. The results are also confirmed by mathematical modelling of the process.

Obvious applications for fog-free condensation are processes where there is a large temperature difference between the gas-vapour mixture and the coolant, e.g. low-temperature condensation in exhaust air cleaning and solvent recovery or condensation of vapour from hot reaction gases using cooling water. A more detailed discussion of fog formation, the method of its prevention and draft plant designs may be found in the book (Kaufmann 1998).

Acknowledgement

This paper has been written in connection with the research project on "Efficient Recovery of Solvents by Condensation" (EROS) at the Fachhochschule Zentralschweiz FHZ (former ZTL, Ingenierschule Luzern). The authors would like to thank Hoffmann-La Roche of Basel, Kühni of Allschwil, Prof. K. Schaber of the Institut für Technische Thermodynamik und Kältetechnik der Universität Karlsruhe and the Swiss Kommission für Technologie und Innovation (KTI) for their support on the project and their active collaboration.

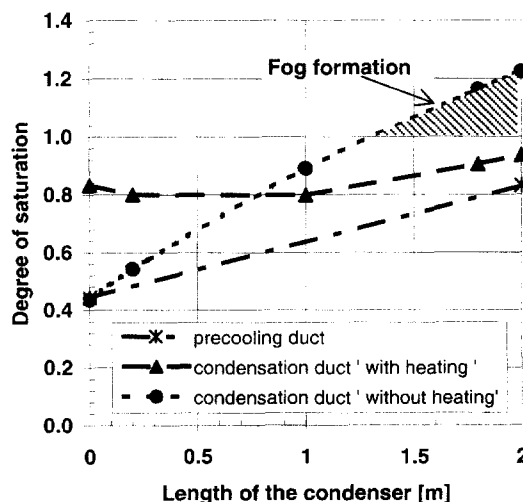


Figure 11. Degree of saturation $S = p_v/p_s(T)$ of gas-vapour mixture, calculated on the basis of concentrations and temperatures measured locally and plotted against the flow distance in the condenser: in condensation with regenerative precooling S is always < 1 , whereas in conventional condensation fog forms within the $S > 1$ range.

REFERENCES

- [1] Schaber K., Schenkel A., Zahoransky R., Drei-Wellenlängen-Extinktionsverfahren zur Charakterisierung von Aerosolen unter industriellen Bedingungen, *TM-Technisches Messen* 61 (7-8) (1994) 295-300.
- [2] Schaber K., Aerosolbildung durch spontane Phasenübergänge bei Absorptions- und Kondensationsprozessen, *Chem Ing. Tech.* 67 (11) (1995) 1443-1452.
- [3] Hinds W.C., *Aerosol Technology*, John Wiley and Sons, New York, 1982.
- [4] Steinmeyer D., Fog formation in partial condensers, *Chem. Eng. Prog.* 68 (7) (1972) 64-68.
- [5] Kim M.H., Kang H.C., Condensation phenomena with wavy interface in the presence of a noncondensable gas, in: Taborak J., Rose J., Tanasawa I. (Eds.), *Condensation and condenser design*, ASME, St. Augustine, Florida, 1993, pp. 219-230.
- [6] West M.K., Hansen E.C., Boundary layer model of condensation in the presence of noncondensable gas, in: Taborak J., Rose J., Tanasawa I. (Eds.), *Condensation and condenser design*, ASME, St. Augustine, Florida, 1993, pp. 93-104.
- [7] Stefan J., *Wien. Ber.* 68 (19??) 385-425 to be completed.
- [8] Brouwers H.J.H., Chesters A.K. Film models for transport phenomena with fog formation: the classical film model, *Int. J. Heat Mass Tran.* 35 (1992) 13-28.
- [9] Kaufmann S., *Kondensation von Dampf aus Gemischen mit Inertgas: Effiziente Trennung statt Nebelbildung* (available at: FHZ-HTA, Fachhochschule Zentralschweiz-Hochschule für Technik + Architektur, Prof. Dr. K. Hilfiker, Technikumstrasse, CH-6048 Horw/Luzern), 1998.
- [10] Colburn A.P., Edison A.G., Prevention of fog in cooler-condensers, *Ind. Eng. Chem. Res.* 33 (4) (1941) 457-458.