Proceedings of PVP 2009 2009 ASME Pressure Vessel and Piping Division Conference July 26-30, 2009, Prague, Czech Republic

# PVP2009-77882

# **GAS LEAKAGE CORRELATION**

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# ABSTRACT

A project has been carried out on the correlation of leakage measurements of different gases in specific conditions in bolted flanged assembly.

The objectives were :

- to check if the relationship between the leakage measurements of Helium, CO<sub>2</sub>, refrigerant R134a and CH<sub>4</sub> would be similar to results of calculations that can be found when using correlation formulas,
- to compare emissions of a typical laboratory fluid (Helium) with process fluids like methane, used in petrochemical plants, as well as CO<sub>2</sub> and R134a, used in the refrigeration industry.

Referring to TA Luft and VDI 2440, which specify a unique test for the certification of gaskets, the configurations of the testing installation used consist of flanges assembled with either PTFE based, graphite or fibre based gaskets. Different gasket loads and internal pressure were applied in order to simulate molecular, intermediate or laminar leakage flow rates.

This article describes the test configurations and shows the leakage measurement results. It also compares the ratios between the leakage values and the results that would be obtained by predicting the leakage of another gas and/or in other pressure conditions, using Poiseuille or Knudsen laws.

# INTRODUCTION

It is mportant to know the leakage rate of a process gas in its service conditions. For example, refrigeration or airconditioning installations work with refrigerants whose emission to the atmosphere is harmful to the environment. Another example is to avoid emissions of volatile organic compounds (VOC) which are polluting the air.

In-lab leakage tests are generally performed with different parameters than to what is found in real service conditions. The in-lab measurement of leakage would be done with a tracer gas, preferably a safe one. Helium is for example the most used tracer gas for this purpose. Additionally, other conditions, like temperature and pressure, are sometimes adapted to the test rig capabilities.

The results of such in-lab tests are then expressed in terms of leakage rate for a particular gas in specific conditions. The interest is to know the equivalent leakage rate of the process gas (different to the tracer gas) in its service conditions. A way to find this equivalence is to perform a conversion using laws of physics. The corresponding equations, as simple as possible, would give a result, but is this result corresponding to the real one if it was measured?

This paper describes a small part of actions attempting to answer this question.

## **CORRELATION METHODS**

Leakage flow is commonly identified as being either laminar, molecular or intermediary. Laws of physics use certain parameters in the equations describing the flow types : molar mass or dynamic viscosity of gas, temperature, upstream and downstream pressures, diameter and length of capillary.

#### Determination of flow type with EN standard

Standard EN 1779 December 1999 [1] indicates the influence of flow conditions.

The usual laws governing gas flow shall be used to calculate variation in leakage rate, as a function of pressure, temperature and type of gas.

In quantitative leak detection two different flow regimes are normally considered. These are the regimes of viscous laminar or molecular flow.

The boundaries between these regimes are not precisely defined. Care shall be taken therefore in the selection of any of the formulas given below.

For practical purposes it is generally accepted that for helium leakage rates less than or equal  $10^{-7}$  Pa.m<sup>3</sup>/s, conditions for molecular flow apply. For helium leakage rates greater than  $10^{-5}$  Pa.m<sup>3</sup>/s, conditions for viscous laminar flow apply in the case of a single capillary leak.

For the different flow regimes the dependence of leakage rate on pressure, temperature and type of gas is different.

#### Determination of flow type with Kn

Kn is the Knudsen number. It is defined, for a straight cylindrical capillary, as being the ratio  $\lambda/D$ . However, it is generally written as follows so as not to have resulting numbers with decimals :

$$K_n = \frac{D}{\lambda} \tag{1}$$

The mean free path  $\lambda$  is the mean distance that a molecule can travel before meeting another one.

The Knudsen number can be calculated in different manners. For more practicality, we present it in the Chapman form[6] :

$$Kn = \frac{D}{\eta} \left( \frac{P_h + P_b}{2} \right) \sqrt{\frac{2M}{\pi RT}}$$
(2)

If the flow regime is the same over the entire length of the leak path, and

if Kn > 100, we could assume that the flow is laminar.

if Kn < 1, we could assume that the flow is molecular.

if 1 < Kn < 100, we could assume that the flow is intermediary.

Limits indicated in literature sometimes vary. They can be 1 and 80, 1 and 200, 0.1 and 1000.

On the molecular level, laminar flow corresponds to a free mean path smaller than the diameter of the leak path, or, in other words, to a number of shocks between molecules larger than the number of shocks of molecules on the walls of the leak path.

On the other hand, molecular flow corresponds to a free mean path larger than the diameter of the leak path, or to a number of shocks between molecules smaller than the number of shocks of molecules on the walls of the leak path.

## Determination of flow type with the critical diameter

This method consists of assuming that  $Q_{molecular} = Q_{la \min ar}$  for a value of D which is called *critical diameter*  $D_{C}$ .

Thus,

$$D_C = \frac{128}{3} \sqrt{\frac{2RT}{\pi M}} \frac{\eta}{(P_h + P_b)}$$
(3)

Assuming a certain length of capillary L, we can calculate the corresponding leakage rate :

$$Q_{C} = \frac{\pi D_{C}^{4}}{256\eta L} \left( P_{h}^{2} - P_{b}^{2} \right)$$
(4)

or

$$Q_C = \frac{D_C^3}{6L} \sqrt{\frac{2\pi RT}{M}} (P_h - P_b)$$
(5)

Then  $Q_C$  and Q are compared, Q being the measured leakage rate.

If  $Q >> Q_C$ , we could assume that the flow is laminar.

If  $Q \ll Q_C$ , we could assume that the flow is molecular.

If  $Q \approx Q_C$ , we could assume that the flow is intermediary.

#### **Correlation equations**

The equation that describes a molecular flow is the Knudsen equation [5]:

$$Q = \frac{D^3}{6L} \sqrt{\frac{2\pi RT}{M}} (P_h - P_b) \tag{6}$$

An example of conversion equation in a case of molecular flow is :

$$Q_2 = \sqrt{\frac{M_1 T_2}{M_2 T_1}} \times \frac{(P_{h2} - P_{b2})}{(P_{h1} - P_{b1})} Q_1$$
(7)

 $(Q_1 \text{ and } Q_2 \text{ having the same unit})$ 

The equation that describes a laminar flow is the Poiseuille equation [5]:

$$Q = \frac{\pi D^4}{256\eta L} (P_h^2 - P_b^2)$$
(8)

An example of conversion equation in a case of laminar flow is :

$$Q_{2} = \frac{\eta_{1}}{\eta_{2}} \times \frac{(P_{h2}^{2} - P_{b2}^{2})}{(P_{h1}^{2} - P_{b1}^{2})} Q_{1}$$
(9)

 $(Q_1 and Q_2 having the same unit)$ 

Correlation for intermediary regimes is a case that concerns leakages which are not taken into account in the EN standard. Indeed, the standard specifies the type of conversion to do in case of molecular or laminar flow, but not intermediary.

#### We suggest the following method.

For an intermediary flow, we admit the use of the Knudsen generalised equation.

$$Q_{C} = \frac{\pi D^{4}}{256\eta L} (P_{h}^{2} - P_{b}^{2}) + \frac{D^{3}}{6L} \sqrt{\frac{2\pi RT}{M}} (P_{h} - P_{b})$$
(10)

Using this equation does not lead to the elimination of the unknown quantities D and L.

For a test, during which the leakage rate  $Q_1$  of a particular gas in specific conditions is measured, we would have :

$$Q_{1} = \frac{\pi D^{4}}{256\eta_{1}L} \left( P_{h_{1}}^{2} - P_{b_{1}}^{2} \right) + \frac{D^{3}}{6L} \sqrt{\frac{2\pi RT_{1}}{M_{1}}} \left( P_{h_{1}} - P_{b_{1}} \right)$$
(11)

We then assume a value for the length of the capillary tube L and deduce a value for the diameter D while doing iterative calculations on a leakage rate Q until obtaining the desired leakage rate  $Q \approx Q_1$ .

Once this is done, the assumed value of L and the determined value of D are used in the Knudsen generalised equation for the calculation of the leakage rate for another gas, and possibly in other pressure and temperature conditions. Although it is best to be as close as possible to the real service conditions.

#### Precautions

The methods shown above, with which the type of flow regime of a leak can be determined, are based on the hypothesis that the gas flows through a unique capillary. This means that only one defect is assumed to be at the origin of the leak.

This assumption can be made when using the correlation formulas given earlier. However, it is necessary to be cautious not to conclude too quickly when obtaining a result with one of these formulas. Indeed, let's take an example from which the determination methods of the type of flow indicate that it is laminar (with a single defect). If we assume that in reality there is a large number of smaller leaks, each one of them in a much smaller capillary, in diameter, each flow could well be molecular. The result of a correlation in a regime or another can be very different.

Reality can also be very different : several leak paths, tortuous paths, of variable section and of all shapes, with separation(s) of paths, changes of flow regime, etc.

The ideal test would be that it is performed in conditions which are as close as possible to real service conditions. If some factors, like the use of a test gas which is different to the process gas, for safety reasons for example, it is recommended to do several correlation calculations, and the results are to be taken as an order of magnitude only.

## TESTS

TA Luft [2] and VDI 2440 [4] specify a unique test for certification of gaskets to be of High Grade Performance according to TA Luft. Evidence of high quality is provided, if a specific leakage rate of  $1.0.10^{-4}$  mbar·l/(s·m) at 0 bar internal pressure (overpressure) of Helium is maintained. At a given mean circumference of 0.215 m (DN40 PN40 gasket) the absolute leakage rate must be less than  $2.15 \cdot 10^{-5}$  mbar·l/s. Other verification procedures can be used.

Citation of VDI 2440 [4]: validated methods of testing, for example pressure decrease method (DIN 28090-2) or flushing gas method are permitted.

The aim of this topic was the determination of leakage rates for three types of gaskets at different gasket stresses, different media at different internal pressures. Pressure increase method was chosen as the testing method.

## Test rig

The test setup is described in Fig. 1. The test flange connection is welded with a standard 200 mm vacuum blind flange. The flanges are loaded by hollow drilled bolts with feeler pins welded in. The bolt force is determined with screwed dial gauges, which display the force as elongation of the calibrated bolts compared to the unloaded feeler pin.

A recipient is put over the test flange connection and sealed on the vacuum blind flange. The recipient is evacuated with the Helium mass spectrometer and an additional pump. For vacuum pressure detection a capacitive membrane pressure sensor with a digital interface is used. Measured data are recorded with a computer. For unexpected pressure increase, the sensor is pressure resistant until 2 bar overpressure.

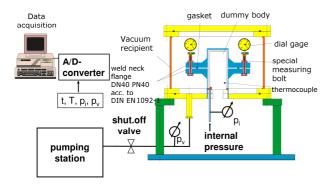


Figure 1 : Test setup for increase pressure method

### Test method

All measured leakage rates were determined by the pressure increase method. Thereby pressure increasing in the recipient, which is caused by gas particles passing through the gasket by diffusion, is established over time. Leakage rate can be evaluated by the formula in Fig. 2; V stands for the available chamber volume, that remains after subtracting test flange volume. As soon as pressure behaviour is becoming linear, total leakage rate can be determined. Therefore a numerical (method of least squares) and a manual adaptation, whereas a line of best fit is inserted into curve linearity, was conducted. Leakage data given below are derived from manual fitting.

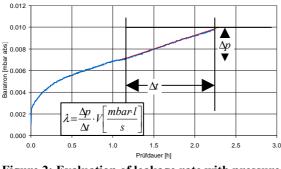


Figure 2: Evaluation of leakage rate with pressure increase method

The advantage of the pressure increase method lies in the determination of leakage rates for any media. Slow variances of pressure, which are caused for example by changes of the ambient temperature, have no effect on leakage rate, if the leakage rate is evaluated from pressure difference of small time slices. Only for low leakage rates, whereas pressure increasing can just be determined over a long period of time, the change of temperature has to be compensated arithmetically.

#### **Tested configurations**

All tests were carried out at ambient temperature. All gasket types were mounted at three different stress levels : 10, 20 and 40 MPa. For every stress level a new gasket was mounted. For each medium a new gasket was mounted. Every mounted gasket was pressurised with 3 or 4 internal pressure

levels (overpressure) : 0 bar (not for R134a), 10 bar, 20 bar and 40 bar.

The elements of configurations are :

- 3 gasket types : fibre, graphite and PTFE based
- 4 media : Helium, R134a, CH<sub>4</sub> and CO<sub>2</sub>
- 3 gasket stress levels : 10, 20 and 40 MPa
- 4 internal pressure levels : 0, 10, 20 and 40 bar.

A sum of 36 tests were performed: 3 gasket types x 3 gasket stress levels x 4 media.

# **TESTS RESULTS**

A few examples of results are shown as qualitative charts (Fig. 3 to 6) with gasket types and media on x- and y- axis and the leakage rate on the z-axis, for every combination of internal pressure and gasket stress.

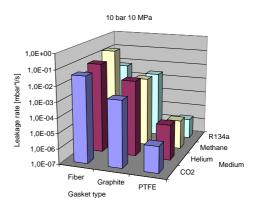


Figure 3: Leakage rate of 3 gasket types tested with 4 media at 10 bar, 10 MPa

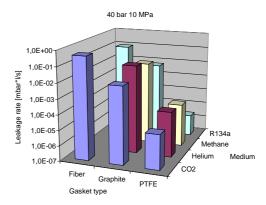


Figure 4: Leakage rate of 3 gasket types tested with 4 media at 40 bar, 10 MPa

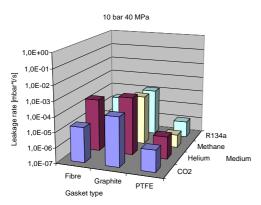


Figure 5: Leakage rate of 3 gasket types tested with 4 media at 10 bar, 40 MPa

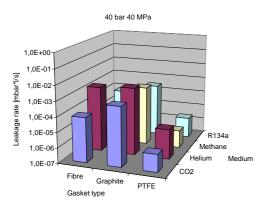


Figure 6: Leakage rate of 3 gasket types tested with 4 media at 40 bar, 40 MPa

#### Remarks

- a) At a stress level of 10 MPa the leakage rate was more than 1 mbar.l/s at internal pressure of 20 and 40 bar with helium and Methane for fibre and graphite based gaskets. So it was too high to be detected with pressure increase method. These measurements are not represented in the graphs.
- b) PTFE gaskets have the lowest leakage rates. Fibre based gaskets must have a gasket stress of more than 20 MPa to be equal or better than graphite gaskets. This behaviour is independent from the medium. It may depend only on this special product of fibre based gaskets.
- c) PTFE was observed to have the highest permeation to Helium.

Figures 7 through 9 show how leakage rate varies for each media type.

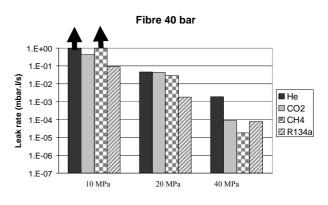


Figure 7: Leakage rate of fibre gasket tested with 4 media at 40 bar

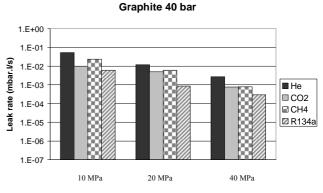


Figure 8: Leakage rate of graphite gasket tested with 4 media at 40 bar

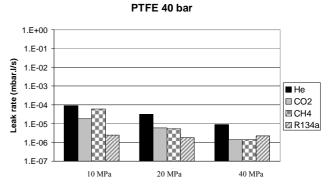


Figure 9: Leakage rate of PTFE gasket tested with 4 media at 40 bar

If we had put all the graphs, meaning the ones at 10 bar and 20 bar, we would notice that, for each type of material of gasket, the differences of leakage rate between the four media are identical in terms of shape from one pressure to another.

For the graphite gaskets, the tendency is that  $Q_{He} > Q_{CH4} > Q_{CO2} > Q_{R134a}$  with less than one decade of difference.

For fibre and PTFE gaskets, the same rough observation can be made but only for the higher leakages (10 and 20 MPa). For the lower leakages,  $Q_{R134a}$  jumps at a higher rank.

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#### CORRELATIONS

Below are shown some examples of charts (Fig. 10 to 13) presenting comparisons between leakage rate measurements of a particular process gas (R134a,  $CO_2$ ,  $CH_4$ ) (crosses on the graphs) and correlated leakage rate calculated from Helium leakage rate measurements using molecular, laminar and intermediary correlation equations (filled markers).

On the last graph (Fig. 13), white filled markers correspond to the values of correlated leakage rate of process gas at 40 bar calculated from Helium leakage rate measured at 10 bar. It is pointed out that correlations were plotted for the other configurations and the trends are similar. For the sake of brevity, only four plots are shown.

Correlations applied only on change of gas are marked with a "G" in the legends of the graphs. Correlations applied on changes of gas and pressure are marked with a "G" and "P" in the legends of the graphs.

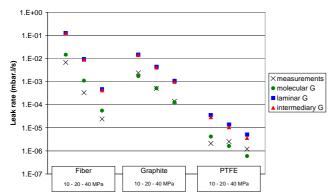
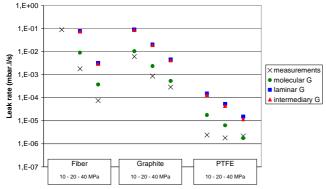
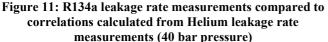


Figure 10: R134a leakage rate measurements compared to correlations calculated from Helium leakage rate measurements (10 bar pressure)





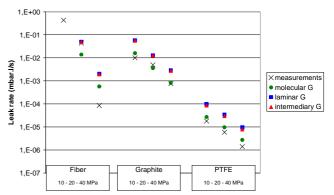
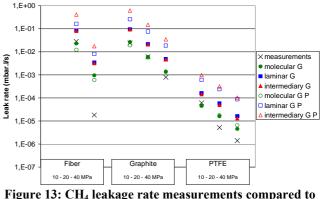


Figure 12: CO<sub>2</sub> leakage rate measurements compared to correlations calculated from Helium leakage rate measurements (40 bar pressure)



correlations calculated from Helium leakage rate measurements (40 bar pressure)

We compare measurements and correlations for each type of materiel separately.

1a) For fibre gaskets, there is no clear cut general observation. For the R134a correlation, the leakage rate is slightly overestimated compared to the measurement. The closest correlation is the one done with the molecular conversion equation (Eq. 7).

1b) For the  $CO_2$  correlation, the correlation in laminar regime (Eq. 9) equals the measurements at 20 MPa, but the correlation overestimates the measurement at 40 MPa.

1c) For the  $CH_4$  correlation, the correlation in molecular regime (Eq. 7) equals the measurements at 20 MPa, but the correlation underestimates the measurement at 10 MPa and overestimates at 40 MPa.

2) For graphite gaskets, the correlated values in molecular regime (Eq. 7) fit well to the leakage rate measurements in all cases (all gases and compression levels).

3) For PTFE gaskets, correlated values either fit the measurements or slightly overestimate them, in general.

In graphite cases, if we follow the EN standard, we would assume that the flow was laminar and therefore, we would use the conversion equation for a laminar flow condition. On the contrary, we notice on the graphs that it is the molecular conversion equation which fits to the measurements. In these particular cases, the guidance for the determination of the type of flow of the EN standard would not be applicable.

Correlated leakage rate for a process gas at a higher pressure, based on Helium leakage rate measurements at a lower pressure, overestimate to a higher extent that of the correlated leakage rate at identical pressure... in laminar or intermediary conditions.

However, correlated leakage rate, in these pressure changes conditions, but in molecular flow regime, is fitting nearly exactly.

Correlation does not only depend on the leakage rate level but also on materials and configurations (geometries, compression, pressure, etc).

## CONCLUSION

Leakage tests are carried out on different configurations which are a combination of gas (Helium, R134a,  $CO_2$  and  $CH_4$ ), type of gasket (fibre, graphite, PTFE), gasket stress level (10, 20, 40 MPa) and internal pressure levels (10, 20, 40 bar). Comparisons of leakage rates between gases are under way, particularly comparisons between measured process gas leakage rates and calculated leakage rate using correlation equations corresponding to different flow regimes.

The tests support the analytical assumptions, used in the EN standard, in certain cases, but not in others.

Some results show a very good correlation for certain configurations, especially with the conversion in molecular regime. This is particularly true for all the cases with graphite gaskets. The levels of measured leakage rate would have led to preferably use the laminar conversion equation according to the EN standard. Consequently, an excessive overestimation would have been done.

More tests would be needed to be performed in order to confirm certain tendencies and, if possible determine correction factors which could be added in the conversion equations.

Other materials, like different types of elastomers could be tested. Tests at different temperatures should be considered, as well as cycling (compression, pressure, temperature). Configurations leading to lower leakage rates should also be investigated.

#### NOMENCLATURE

- D Diameter of capillary[m]
- k Botlzmann's constant [1.381.10<sup>-23</sup> J/K]
- K<sub>n</sub> Knudsen number
- L Length of capillary [m]
- $\lambda$  Mean free path of molecules [m]

- M Molar mass [kg/mol]
- P<sub>h</sub> Upstream pressure [Pa]
- P<sub>b</sub> Downstream pressure [Pa]
- Q Leakage rate [Pa.m<sup>3</sup>/s]
- R Perfect gas constant [8.314 J/(K.mol)]
- T Temperature [K]
- η Dynamic viscosity [Pa.s]

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