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THE GAS-INTERFEROMETER - AN IMPORTANT AID

IN SAFETY TECHNIQUES AND ANAESTHESIA

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75 Jahre Optische Messinstrumente. Oberkochen/Württ.,  
Carl Zeiss, 1968, p. 135-140

Translated at CERN by B. Hodge

(Original: German)

Revised by H. MacCabe

(CERN Trans. Int. 73-6)

Geneva

January 1973

## 1. INTRODUCTION

For more than 50 years gas interferometers of various designs have been manufactured by Carl Zeiss (Haber and Loewe /1/, Loewe /2/, /3/, Hansen /4/, Kinder /5/). Although at an earlier date the preferred interferometric principle was that based on diffraction in a double slit as described by Arago /6/, we today exclusively use gas interferometers based on physical beam splitting, in which the characteristic element is a splitter plate, on account of their greater brightness. We shall first review the basic principles of gas refractometry, describing the most important considerations for determining the concentration of the gases in the mixture. We shall then give two examples of possible applications, one in the field of safety techniques (prevention of explosions), the other in the field of medicine (testing of narcosis equipment).

## 2. BASIC PRINCIPLES OF GAS REFRACTOMETRY

### 2.1 Principle of Interferometric Gas Analysis

Our current programme of equipment for gas analysis offers the Zeiss fire-damp interferometer and the Zeiss interference refractometer. Both devices have already been described at length (Kinder /7/, /8/, Torge /9/), so that it is not necessary here to go into their design in detail. Figures 1 and 2 show the two interferometers, and Figures 3 and 4 are general layout diagrams of the optical system. Both devices are 2-ray interferometers, and both have a double beam path through the gas chambers. The two light beams interfering with each other pass each through a chamber, one of which contains the gas- or vapour-mixture to be examined, and the other a comparison gas. The displacement of the interference fringes (which in the case of the interference refractometer, is measured by means of a compensator, and, in the case of the fire-damp interferometer, can be read off on a scale) is directly proportional to the difference of refractive index of the two chamber fillings. Interferometric gas analysis

is based on the relationship between the concentration and the refractive index of a mixture. For practical purposes, the resulting refractive power  $n-1$  of a mixture of  $k$  components is obtained by summation in proportion to the concentration  $V$  (percentage volume of the individual contributions to the overall mixture) (Bittel /10/), i.e.

$$n-1 = \sum_{i=0}^k \frac{V_i}{100} \cdot (n_i-1) \quad (1)$$

$V_i$  = concentration of the  $i$ -th component,  
 $n_i$  = refraction index of the  $i$ -th component.

If a gas having a refractive power  $(n_0 - 1)$  contains an admixture of  $V_1$  percent by volume of a second gas with a refractive power  $(n_1 - 1)$ , the refractive power obtained for the mixture is therefore:

$$n-1 = (n_1-1) \frac{V_1}{100} + \frac{100-V_1}{100} (n_0-1) \quad (2)$$

The relationship between the fringe number  $h$ , the chamber length  $L$ , the difference of refractive index of the two chamber fillings and the vacuum wavelength  $\lambda_0$  will then be as follows, taking the main component of the mixture, which has the refractive power  $n_0 - 1$ , as the comparison gas

$$h = \frac{2L}{\lambda_0} \left[ (n-1) - (n_0-1) \right] = \frac{2L}{\lambda_0} \cdot \frac{V}{100} \cdot (n_1 - n_0) \quad (3)$$

$$V = h \cdot \frac{\lambda_0}{2 \cdot L \cdot (n_1 - n_0)} \cdot 100 \quad (4)$$

These are the fundamental interferometric equations for gas analyses. The refractive indices for the substances involved are taken from the known tables (e.g. Landolt-Börnstein, 6th edition 1962, Vol. 2 Part 8). As the individual interference fringes cannot, in practice, be distinguished when monochromatic light is used, the measurement is made with white light, and the darkest region (black fringe), which corresponds to path difference zero, is used as a fiducial mark.

The scale of the interference refractometer is divided into interference fringes. The dimensions of the compensator (Torge /9/) are chosen in such a way that a linear scale can be used, for which a measurement range of 0-200 fringes has been selected. The result of the measurement is thus read out directly in the form of the fringe number  $h$  and the volume concentration  $V$  is calculated by means of Formula (4).

If the difference  $n_1 - n_0$  in the refractive indices of the two gases has to be found, the measurement chamber must be filled completely with the gas with the refractive power  $n_1 - 1$ , and  $V_1 = 100$  must be inserted in Formula (3). We then have:

$$n_1 - n_0 = h \cdot \frac{\lambda_0}{2L} \quad (3a)$$

For a wavelength  $\lambda_0 = 0.5460/\mu\text{m}$ , the scale of the fire-damp interferometer is calibrated for a mixture of methane, which has the refractive power  $n_1 - 1 = 443.3 \times 10^{-6}$  (Tausz and Hornung /11/), and dry  $\text{CO}_2$  - free air, for which  $n_0 - 1 = 293.24 \times 10^{-6}$  (Engelhard /12/); Formula (3) then gives:

$$h_{\text{methane}} = \frac{2L}{\lambda_0} \cdot \frac{V_{\text{methane}}}{100} \cdot 150.1 \cdot 10^{-6} \quad (5)$$

This relationship is valid for  $0^\circ\text{C}$  and 760 Torr (see Chapter 2.2 for conversion to other temperature and pressure conditions).

If we want to use the fire-damp interferometer for measuring any given gas mixture containing a component 1, with a volume concentration  $V_1$  and a refractive power  $n_1 - 1$ , in a basic gas 0 with a refractive power  $n_0 - 1$ , and if we take this basic gas as the comparison gas, we obtain

$$h_1 = \frac{2L}{\lambda_0} \cdot \frac{V_1}{100} \cdot (n_1 - n_0) \quad (6)$$

The same fringe number  $h_1$  would also be obtained by a methane concentration of  $V_M$  percent by volume in air, i.e.

$$h_1 = \frac{2L}{\lambda_0} \cdot \frac{V_M}{100} \cdot 150.1 \cdot 10^{-6} \quad (7)$$

Formulas (6) and (7) yield the following equation:

$$V_1 = V_M \cdot \frac{150.1 \cdot 10^{-6}}{n_1 - n_0} \quad (8)$$

The measurement result for the gas mixture 1 - 0 is thus obtained by multiplying the reading on the fire-damp interferometer scale by the calibration factor

$$G = \frac{150.1}{n_1 - n_0} \times 10^{-6} *$$

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\* Earlier devices have a slightly different calibration factor of  $G = \frac{148.26}{n_1 - n_0} \times 10^{-6}$ .

## 2.2 Influence of Temperature and Pressure

As the refractive index of gases and vapours depends on their temperature and pressure, these values must be taken into account during measurements.

### 2.2.1 Measurement of gases and vapours with the interference refractometer

Let us assume that, at a temperature  $T$  (degrees Kelvin) and pressure  $p$  (Torr), both chambers of the interference refractometer are filled with substances which have the refraction powers  $n_1 - 1$  and  $n_0 - 1$ . The fringe is then:

$$h = \frac{2L}{\lambda_0} \cdot (n_1 - n_0)_{p, T} \quad (9)$$

In order to effect the transition to another temperature  $T_0$  (degrees Kelvin) and another pressure  $p_0$  (Torr), we use the Lorenz-Lorentz equation according to which  $\frac{n^2 - 1}{n^2 + 2}$  is proportional

to the density  $\varrho$ . In the case of gases, we can assume that  $n - 1 = \text{const.} \cdot \varrho$  on account of the small value of  $n - 1$ . Furthermore, if the ideal gas equation is valid,  $\varrho = \frac{p}{T} \cdot \frac{1}{R}$ . For two refractive powers  $(n - 1)_{p, T}$  and  $(n - 1)_{p_0, T_0}$  under the conditions  $p, T$  and  $p_0, T_0$  we have thus:

$$(n - 1)_{p, T} = (n - 1)_{p_0, T_0} \cdot \frac{p \cdot T_0}{p_0 \cdot T} \quad (10)$$

Consequently, when measuring two substances for which the refractive powers for the temperature  $T_0$  and pressure  $p_0$  have been obtained from tables, the measurement result at temperature  $T$  and pressure  $p$  will be as follows, using equations (9) and (10):

$$h = \frac{2L}{\lambda_0} \cdot (n_1 - n_0) \rho_0 \cdot T_0 \cdot \frac{p \cdot T_0}{\rho_0 \cdot T} \quad (11)$$

For practical purposes, it may be assumed that  $\frac{T}{T_0} = 1 + a \cdot t$ , where  $a = 1 : 273.15$  and  $t$  is expressed in degrees centigrade. For very precise measurements, it is recommended to refer to the original literature for the type of reduction to  $T_0$  and  $p_0$ . Furthermore, when making precision measurements of the highest accuracy, it may be necessary, in certain cases, to use a better approximation for the density dependence and the divergence from the ideal gas law (Edlén /13/).

### 2.2.2 Measurement of gases and vapours with the fire-damp interferometer

The setting of the interference fringes on the fire-damp interferometer has been based on a pressure of 790 Torr and a temperature of 20° centigrade in accordance with the conditions prevailing underground. If the measurement and comparison chambers have a different temperature  $t$  and pressure  $p$ , the result must be multiplied by the factor  $F = 2.69 \times \frac{273 + t}{p}$ . This formula has been derived using the ideal gas law and the linear relationship between the refractive power and the gas density.

### 2.3 Discontinuities

To avoid troublesome irregularities in the white-light interferences, it is sufficient, according to Hansen /4/, to make the relative reciprocal effective dispersion  $v^* = \frac{G}{\frac{\delta G}{\delta n} \cdot \Delta n}$  equal to the dispersion  $\bar{v} = \frac{(n_1 - n_0)e}{\Delta n_1 - \Delta n_0}$  of the chamber filling. ( $G =$  compensator path difference;

$$\Delta n_1 = n_{1F} - n_{1C}; \Delta n_0 = n_{0F} - n_{0C}.$$

For the achromatic fringe used for the read-out, the path difference for two wavelengths is then simultaneously compensated. If, for the achromatic fringe, the fringe number  $h$  is to be independent of wavelength, i.e. if  $\left. \frac{\delta h}{\delta \lambda} \right|_{\lambda = \lambda_0} = 0$  (in this connection see, for example, Väisälä /14/), and if we introduce the group refraction index  $n_g = n - \lambda \frac{\delta n}{\delta \lambda}$  for the substance and the corresponding expression

$G_g = G - \lambda \frac{\delta G}{\delta \lambda}$  for the compensator, we obtain for the balance condition

$$G_g = L (n_{lg} - n_{og}) \quad (12)$$

The achromatism condition is found to be

$$\frac{G}{\delta G} = \frac{n_1 - n_0}{\frac{\delta}{\delta \lambda} (n_1 - n_0)} \quad (13)$$

which, of course, is also obtained if the two wavelengths coincide in Hansen's achromatism condition (see above).

As a mean value of  $v^* = 55$  provides good achromatism for many gas-air mixtures, the compensator of the interference refractometer has been designed accordingly.

With the fire-damp interferometer, too, the lack of colour may, as the concentration increases, move from the true zero fringe to a neighbouring fringe if the refraction difference  $n_1 - n_0$  in a gas mixture is so strongly dependent on the wavelength that the dispersion  $\bar{v}$  of the chamber fillings has a small finite value. In the case of the fire-damp interferometers, with the measurement ranges 0-5% and 0-10%, the first discontinuity occurs in the vicinity of the scale value  $s = \bar{v}/4$  (Kinder /15/). In the case of equipment having a measurement range of 0-100%, the first discontinuity would occur at a scale value ten times greater, on account of the shorter length of the measurement chambers.



### 3. CONCENTRATION MEASUREMENTS IN HYDROCARBON-AIR MIXTURES

#### 3.1 Measurement Method

For the purpose of safety techniques, notably in order to verify that there is no danger of explosion, it is particularly important to know the calibration factors for hydrocarbons. In this case, it is expedient to use a relative concentration measure K for the vapour content and to relate the volume concentration to the lower explosion limit (LEL). K is defined as follows:

$$K = \frac{\text{concentration (percent by volume)}}{(\text{LEL})} \frac{(\text{percent by volume})}{(\text{percent by volume})}$$

As such safety measurements very frequently call for a portable device, the following paragraphs refer only to the portable fire-damp interferometer. If G is the calibration factor of the substance concerned for the fire-damp gas interferometer (see 2.1), the lower explosion limit of the mixture under review lies at the scale value  $X = \frac{\text{LEL}}{G}$ . Table 1 lists, for a number of materials which are important in the field of safety, the boiling point, the volume percentage of the lower explosion limit (Nabert, Schön /16/) and the position of the lower explosion limit on the scale of the fire-damp interferometer. Figure 5 shows graphically the relationship between readings of the fire-damp interferometer and the relative concentration K. The most significant result is that, for materials having a boiling point of over 30°C, the interferometric readings corresponding to the lower explosion limit are concentrated in a relatively narrow range.

#### 3.2 Influence of Temperature and Pressure

Using the factor given in 2.2, Figure 5 also shows, for substances whose boiling point is over 30°C, the limiting lines for the distribution of the true measurement values when variations of pressure and temperature occur.

### 3.3 Influence of Water Vapour

If, in addition to hydrocarbon vapour ( $V_x$ ), the air contains also water vapour ( $V_{H_2O}$ ), (this is, for example, the case after vessels have been cleaned with steam), the refractive power of the mixture is, according to Formula (1):

$$n-1 = \frac{V_x}{100} \cdot (n_x-1) + \frac{V_{H_2O}}{100} \cdot (n_{H_2O}-1) + \frac{100-V_x-V_{H_2O}}{100} (n_{Luft}-1) \quad (15)$$

and the fringe shift is:

$$h = \frac{2 \cdot L}{\lambda_0} \cdot \left[ \frac{V_x}{100} (n_x - n_{Luft}) + \frac{V_{H_2O}}{100} (n_{H_2O} - n_{Luft}) \right] \quad (16)$$

Compared with a dry mixture, the error introduced by the water vapour amounts, therefore, to S graduations of the methane scale, where S is given by

$$S = \frac{n_{H_2O} - n_{Luft}}{150,1 \cdot 10^{-6}} \cdot V_{H_2O} = -0,27 \cdot V_{H_2O} \quad (17)$$

Hence, with a water vapour concentration of about 4 percent by volume, which corresponds approximately to the very high saturation concentration at 30°C and 760 Torr, the water vapour error is approximately equivalent to 1 graduation interval on the interferometer scale, an amount by which the measurement value is lowered.

### 3.4 Consequences for Safety Techniques

Figure 5 shows that the lower explosion limits of fuel vapours mixed with air lie in a relatively narrow range on the fire-

damp interferometer scale. It is therefore possible to set a mark valid for all these fuels below which no danger of explosion occurs. If a number of additional requirements are taken into account, such as, for example, the use of almost absorption-free tubes, etc., the danger-mark can be made to coincide with the graduation line 1 of the methane scale. These questions of safety technique have been investigated in detail by the Federal Physico-Technical Office in Braunschweig. An easy-to-use device for safety measurements is therefore available. Because it is based on the optical interferometric measurement principle, it can moreover, be used not only for fuel vapour but also for non-flammable and other vapour-air mixtures.

#### 4. CONCENTRATION MEASUREMENTS OF NARCOTIC SUBSTANCES

When calibrating and checking evaporators of narcotic substances, it is necessary to determine the concentrations of vapour-air mixtures. If high accuracy is required, the interference refractometer is used for the measurement. With a standard chamber-length of 250 mm, traversed twice, we have, taking fluothane with a mean refractive power  $n - 1 = 1600 \times 10^{-6}$ , as an example, the following relationship between the fringe number  $h$  and the volume concentration  $V$  (percent per volume):  $h = 12 \cdot V$  (fringe at  $0^\circ\text{C}$ , 760 Torr). For  $20^\circ\text{C}$  and 760 Torr, we obtain, for a chamber length of 223.5 mm the relation  $h = 10 V$ , so that the interferometer scale which is divided linearly into fringes, gives directly the percentage of fluothane (similar solutions can be found for other substances). The dispersion of the compensator on the interference refractometer is so dimensioned that no discontinuities occur in the range of concentrations encountered in practice (0 - 5% fluothane).

The fire-damp interferometer can also prove very useful for routine control of the evaporators (Hohmann /17/). The calibration factors for a number of substances which are of importance in anaesthesia are given in table 2.

We are grateful to Professor G. Schön and the staff of the Federal Physico-Technical Office, Braunschweig, for valuable discussions on questions concerning safety techniques, and to Mr. A. Dehner of the firm Rhein-Pharma in Heidelberg and to Dr. V. Kapfhammer of the Kaiserslautern City Hospital for making it possible to carry out measurements with fluothane evaporators.

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T a b l e 1

Substance	Boiling point at 760 Torr (°C)	Lower explosion limit (percent per volume)	Lower explosion limit (number on fire-damp inter- ferometer scale)
Methane	-161	5.0	5
Ethane	- 89	3.0	10
n-Pentane	36	1.4	13 partly
n-Hexane	69	1.2	14 calculated
n-Heptane	98	1.1	15 from the
n-Octane	126	0.8	12 molar
n-Nonane	151	0.7	12 refraction
n-Decane	174	0.7	14 of the liquid

T a b l e 2

Calibration factors for various vapours  
in relation to air

Substance	Calibration factor G
Nitrous oxide $N_2O$ (Laughing gas)	0.692
Diethylether $(C_2H_5)_2O$	0.119
Chloroform $CHCl_3$	0.130
Fluothane $C_2HF_3BrCl$ (Trifluorbromochlorethane).	0.115
Cyclopropane $C_3H_6$	0.225

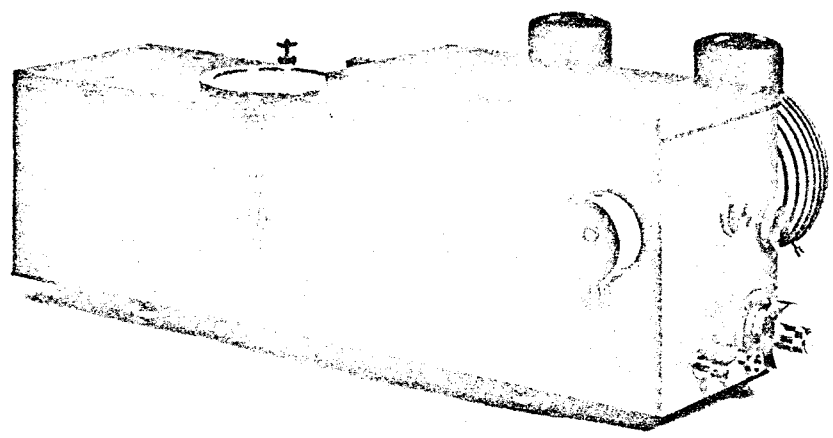


Fig. 1. Interference refractometer.



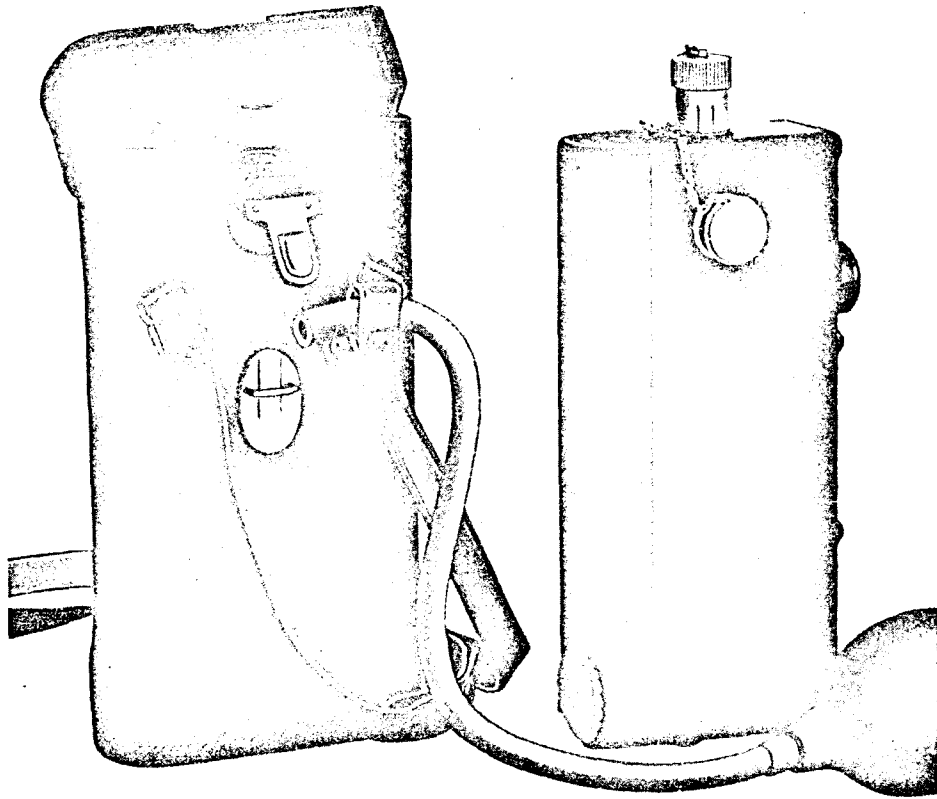


Fig. 2. Fire-damp interferometer.

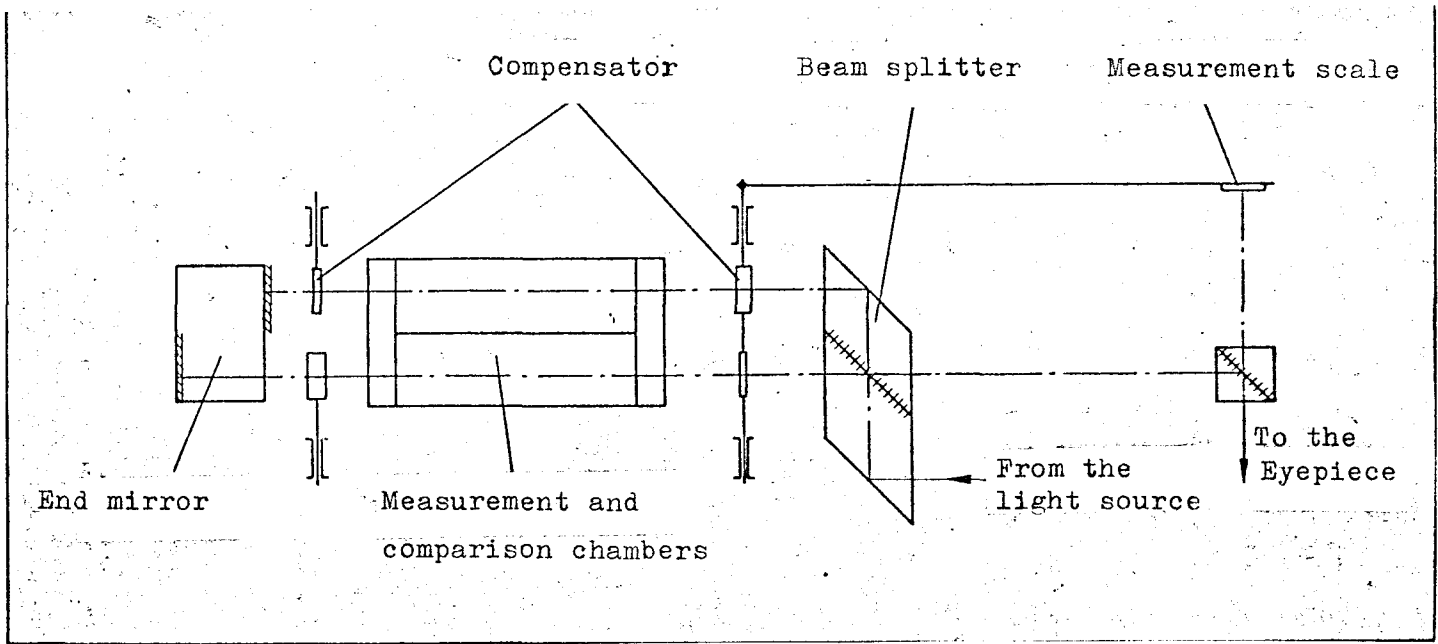


Fig. 3. Optical layout of the interference refractometer.

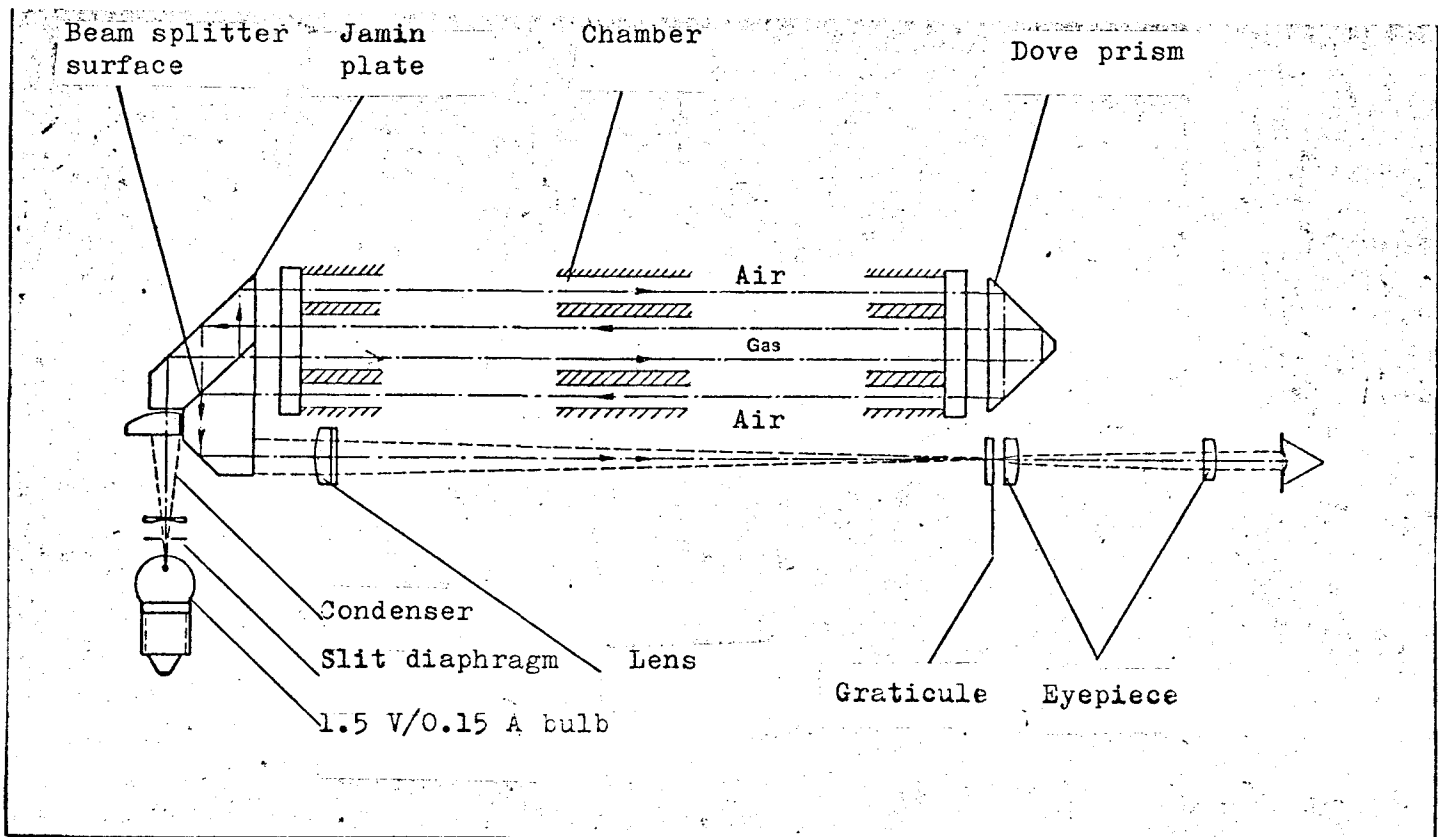
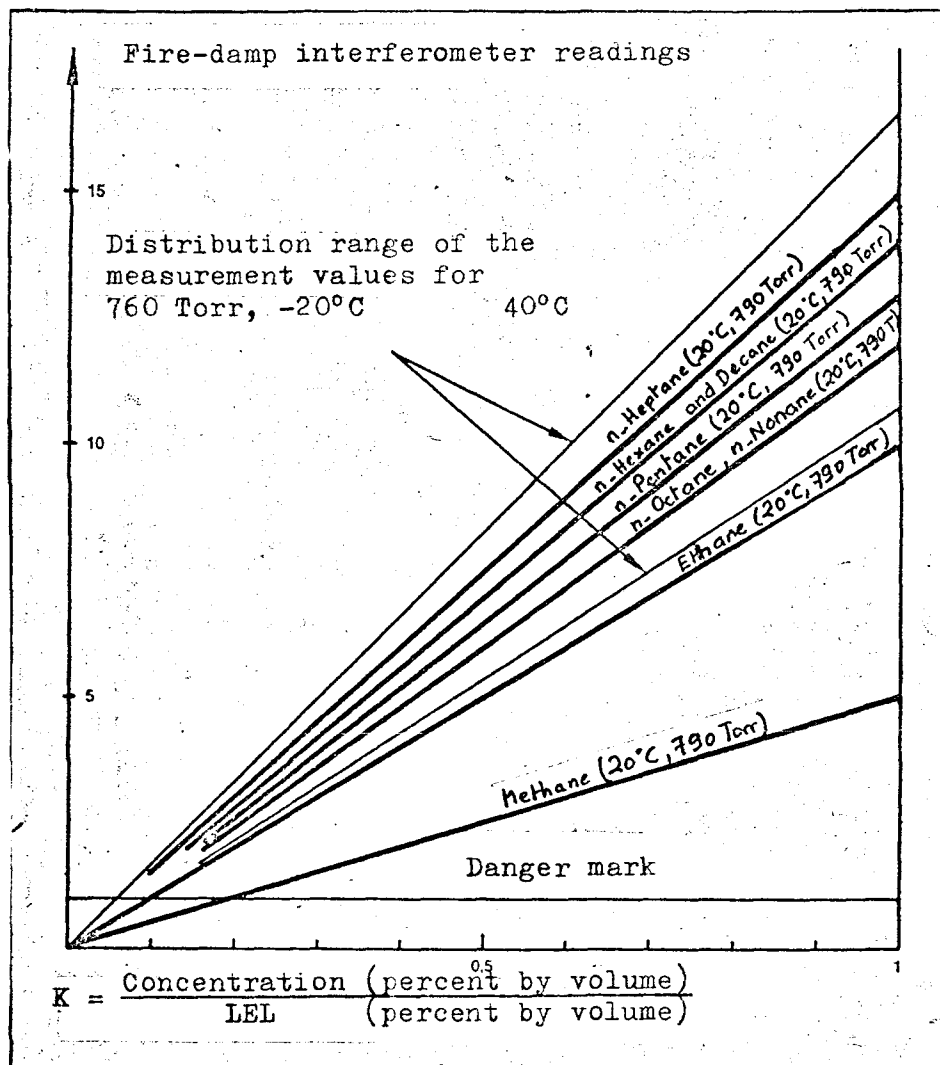


Fig. 4. Optical layout of the fire-damp interferometer.



**Fig. 5.** Dependence of the measurement value on the relative concentration. Distribution of the true value during variation of temperature and pressure.