

## Gas Permeability Apparatus for Films and Sheets

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### Synopsis

An apparatus for measuring the gas permeability of films and sheet material has been developed at the Central Laboratory TNO, Delft (The Netherlands). The apparatus consists of a high vacuum unit, connected to six film holders of a new design and construction. The pressure of the gas which permeates through the film in a constant volume, is measured with the aid of McLeod manometers. The apparatus was developed for rapid, accurate measurements of materials of very low permeability, but it is also suitable for films of high permeability. The range of permeabilities to be measured is between  $10^{-7}$  and  $10^{-13}$  cm.<sup>3</sup> (S.T.P.)-film thickness/cm.<sup>2</sup>-sec.-cm. Hg. The measuring time for the  $10^{-13}$  range is about 6 hr. (for plastic laminated with aluminum) and for the  $10^{-7}$  range is about 15 min. (for polyethylene). Measurements can be executed simultaneously on six heads and easily at various temperatures and relative humidities. The apparatus is suitable both for research purposes and for routine measurements in industry.

### INTRODUCTION

The permeability apparatus described in this paper is especially developed for quick, accurate measurements of materials of very low permeability such as saran, aluminum laminates, and of extremely thick specimens (up to 10 mm. thickness). It is also suitable for films of high permeability.

Special attention has been paid to the following points: (1) easy handling of the film holder (Installation of six film specimens can be performed within 10 min. without the use of special tools or vacuum grease.); (2) easy method of operation (Six measurements can be made simultaneously.); (3) simple temperature control without the use of gas or liquid thermostats; (4) influence of relative humidity on the permeability of hydrophilic films (e.g., plain regenerated cellulose), with both sides of these films under constant relative humidity; (5) easy control of the leak rate under test conditions (A dummy test and the CO<sub>2</sub> permeability can be done in the same run.).

The range of permeabilities to be measured is between  $10^{-7}$  and  $10^{-13}$ /cm.<sup>3</sup> (S.T.P.)-film thickness/cm.<sup>2</sup>-sec.-cm. Hg, i.e., a range of six decades. The measuring time for the  $10^{-13}$  range is about 6 hr. (for aluminum laminate) and for the  $10^{-7}$  range about 15 min. (for polyethylene).

The starting point for the development of the TNO gas permeability apparatus was the high vacuum technique introduced by Barrer<sup>1,2</sup> and

developed further by Rogers, Meyer, Stannett, and Szwarc.<sup>3,4</sup> The measuring principle of the high vacuum technique is the pressure-volume measurement of the permeated gas through the test film at very low pressure and, accordingly, large volume.

The apparatus of Brown and Sauber<sup>5</sup> (ASTM D1434-58), on the other hand, works with a high pressure and a small volume. Brown and Sauber consider the high vacuum technique developed by Stannett et al. expensive and time-consuming and therefore not suitable for industrial purposes.

TNO, on the other hand, took the view that, provided an apparatus of appropriate design is used, the high vacuum technique should yield by far the quickest and most accurate method of measurement. This view is based on the fact that for measurement of absolute amounts of gas with a predetermined accuracy, the high vacuum technique requires far smaller amounts of gas than the low vacuum technique.

Use is made of a McLeod manometer as the measuring instrument, the permeated gas being compressed into a small capillary (0.8 mm. diameter). Thus the apparatus is very sensitive.

## EXPERIMENTAL

### Description of Apparatus

Figure 1 shows a photograph of the apparatus with all six units in operation. Figure 2 shows a diagram of the TNO gas permeability apparatus.

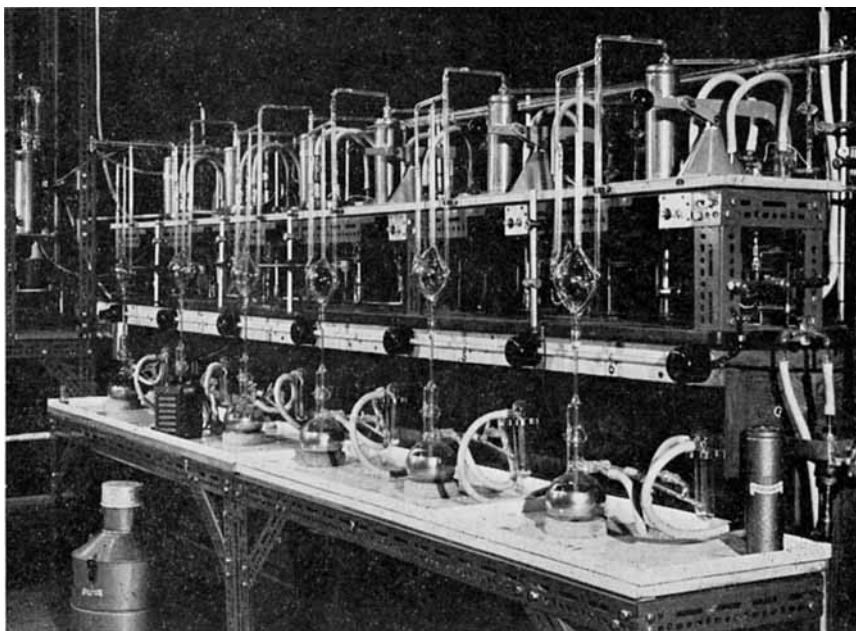


Fig. 1. Gas permeability apparatus. Survey of the six measuring systems.

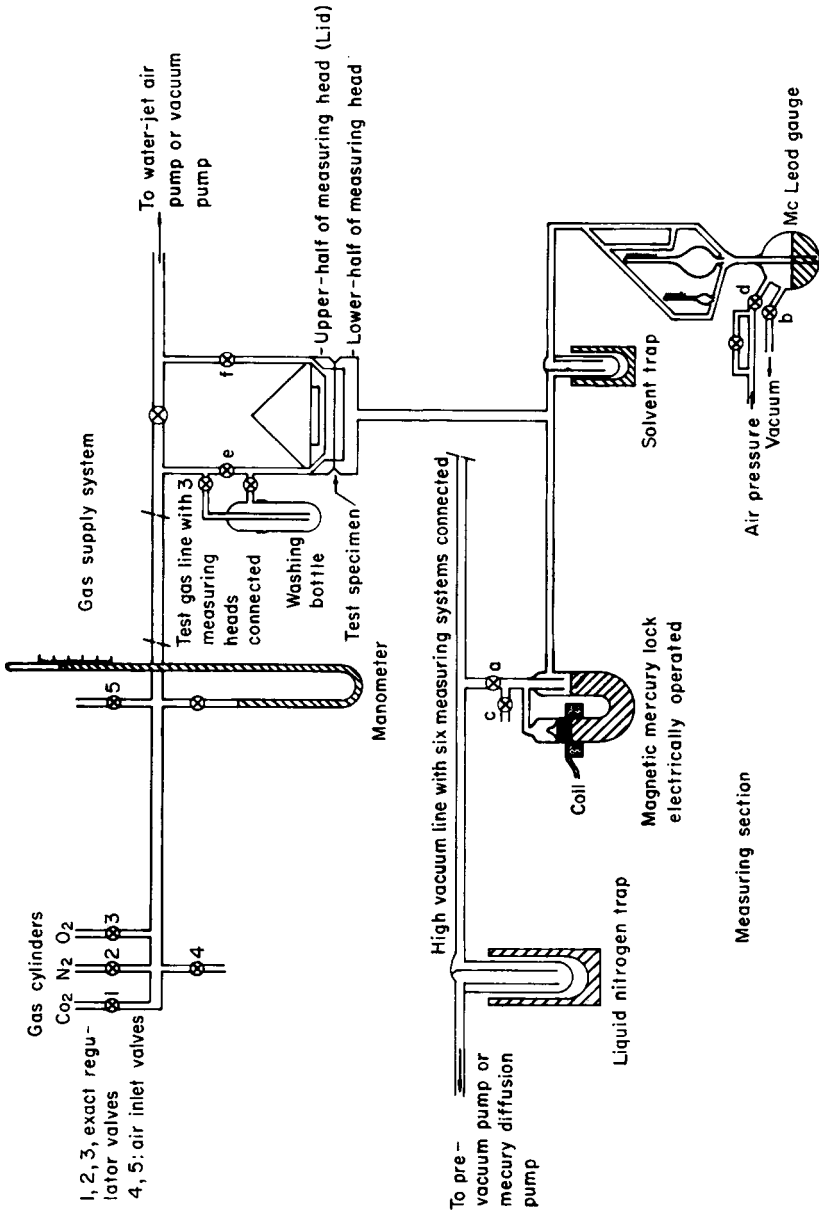


Fig. 2. Schematic view of gas permeability apparatus.

Its film holders, high vacuum system, measuring chambers, gas supply system, and sealing mercury supply system are described below.

### Film Holder

Much importance has been attached to the correct design and construction of the film holder. A detailed drawing of it is shown in Figure 3. The holder consists of two parts: a glass bottom part with a sintered glass filter is in one piece with the measuring chamber; a metal top part or lid is connected to the test gas supply system. The place of the film is between both parts. By maintaining a particular subpressure in the gas chamber over the film, the two halves are pressed firmly together, and the film is secured in place.

Minor unevennesses in the film are offset with a rubber O-ring fitting in an edge of the metal lid.

The film is supported by a sintered glass filter of about 10 cm. diameter. The upper surface of the glass plate with filter is ground and polished. The area of the glass filter determines the size of the diffusing area of the film.

The advantages of this glass film holder over the designs known at present, are: (1) glass, in contrast with metal, has an extremely low gas absorption; (2) the metal-glass seal is eliminated; (3) a sintered glass filter is applied as the film support.

Filter paper often used for this purpose has the unpleasant properties of adsorption and desorption and has no homogeneous structure and density.

The lid of the film holder is made of metal. This metal lid, combined with the low heat-conducting glass bottom, gives an attractive solution for problems of temperature control. The lid is blackened on the inside surface. In the upper part of the lid a heating element is mounted; the temperature at which the test is to be conducted is controlled with the aid of a variable transformer. The temperature is measured by using a thermocouple placed as close as possible to the film surface.

For sealing uneven films, thick plates, etc. (without using grease), there is a channel round the film holder into which mercury is poured automatically after the film is secured (see Fig. 4c).

### High Vacuum System

The high vacuum unit consists of a rotary oil pump (for the prevacuum of  $10^{-2}$  mm. Hg) and a mercury diffusion pump. The vacuum in the unit is automatically retained if the current supply fails, or if the prevacuum or water-cooling of the mercury diffusion pump is not adequate.

The mercury diffusion pump is connected to a collector line, to which are connected six measuring systems. Between the vacuum pump and the collector line there is a condensation flask in a Dewar with liquid nitrogen, in order to increase the efficiency of the vacuum unit.

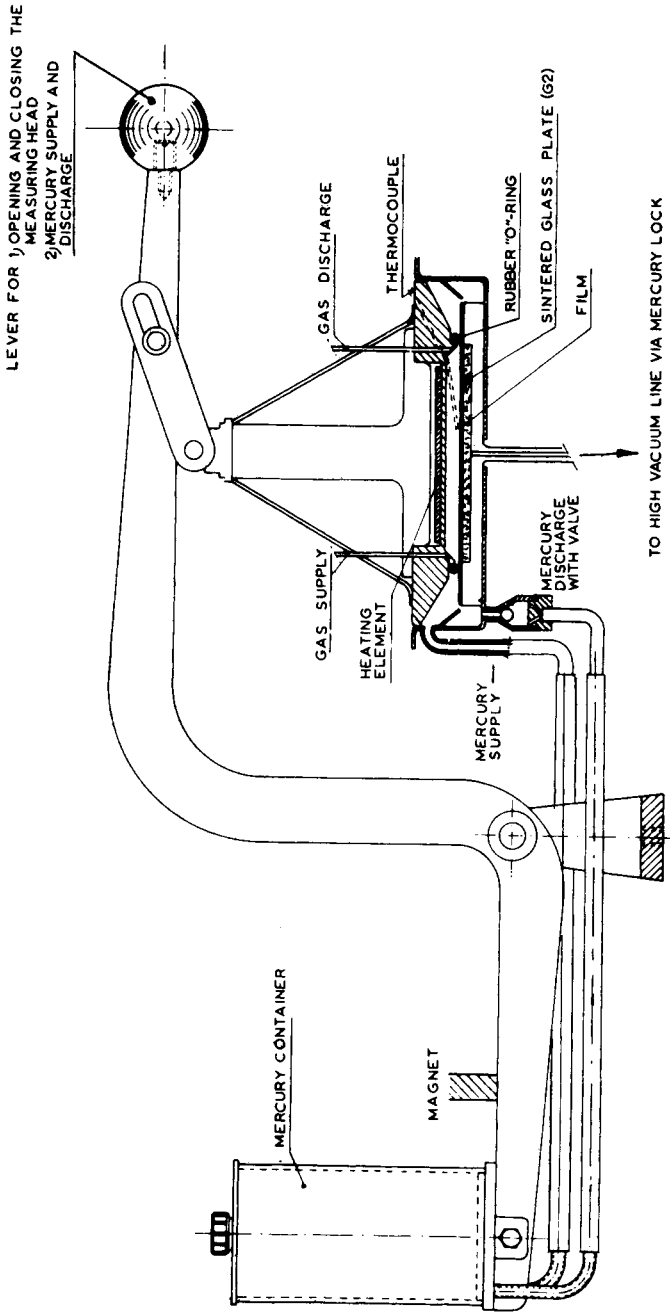


Fig. 3. Measuring head.

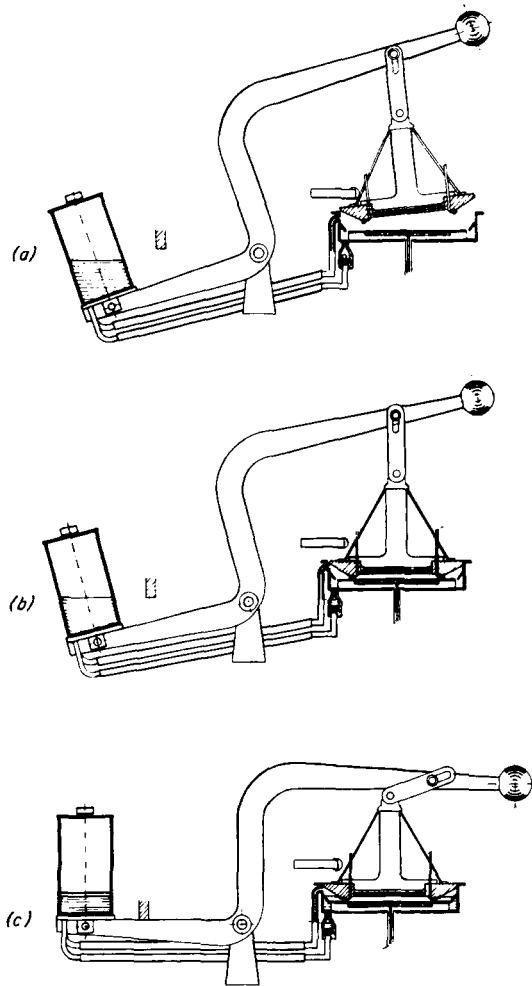


Fig. 4. Mercury supply system: (a) lid in lifted position; (b) lid in closed position; (c) mercury is supplied for sealing uneven films and thick plates.

### Measuring Chamber

All dangers of leakage in the measuring chamber are fundamentally eliminated *inter alia* because no stopcocks are used. The measuring chamber is shut off from the high vacuum collector line by a magnetic mercury lock.

A miniature condensation flask with Dewar removes condensable gas and vapor.

The vacuum is measured with a McLeod gage; this gage requires further description. In the tube between mercury reservoir and compression bulb through which the mercury will rise there is no stopcock; this avoids risks of leakage which influences the test results and prevents the mercury from being contaminated with grease. The absence of a stopcock

has been made possible by connecting the mercury movement system to the vacuum of another rotary oil pump. The mercury in the manometer can be raised at two speeds which allows quicker work and accurate setting of the reading (For details on a pneumatically controlled, self-setting compression-type McLeod vacuum gage unaffected by atmospheric pressure see Peche<sup>6</sup>). There are two compression sections in the manometer for measurements in two connecting vacuum ranges. This makes the unit suitable for rapid measuring of films, varying greatly in permeability.

An advantage of the McLeod gage as a measuring instrument is that only clean mercury from the bottom rises in the capillaries while impurities keep floating on the surface of the mercury in the reservoir. After four years of continuous service it has not been necessary to renew the mercury, nor to clean the measuring system of the gages.

### Gas Supply System

With the TNO system, gas can continuously be blown over the film. The gas flows from a cylinder via a needle valve to the lid and hence over the film. The subpressure of the gas is obtained by means of a water-jet air pump or a vacuum pump. This reduced pressure is adjusted with the needle valve between 0 and 76 cm. Hg, and is measured with a closed mercury manometer. By running the gas through a gas washing bottle mounted parallel to the gas supply line and filled with water or plasticizer, etc., the gas can be conditioned if required. By bubbling the gas through saturated salt solutions it is possible to conduct measurements at constant relative humidities.

One gas supply system normally serves a maximum of three measuring heads.

### Mercury Supply System

In order to seal uneven specimens, thick sheets, etc., without the use of grease, mercury is poured into the channel round the film holder. If the lever is pressed down (see Fig. 4c) the mercury flows from the mercury container through a tube ending over the specimen, and fills the channel.

A non-return valve in the lower backflow tube forces the mercury to flow through the upper tube. A metal ring is placed at such a height that mercury stays on top of the specimen during the filling of the channel. In this way there is no lifting of the edge of the specimen.

### General Method of Operation

The film under test is cut to size (the outside diameter of the glass film holder is taken as a standard), and placed on the sintered glass filter. No vacuum grease is used for sealing; vacuum grease can influence the properties of the test film.

By manipulating stopcocks a and b (see Fig. 2) a vacuum is created on both sides of the mercury in the reservoir of the McLeod manometer. The film is sucked firmly to the glass bottom part of the film holder during

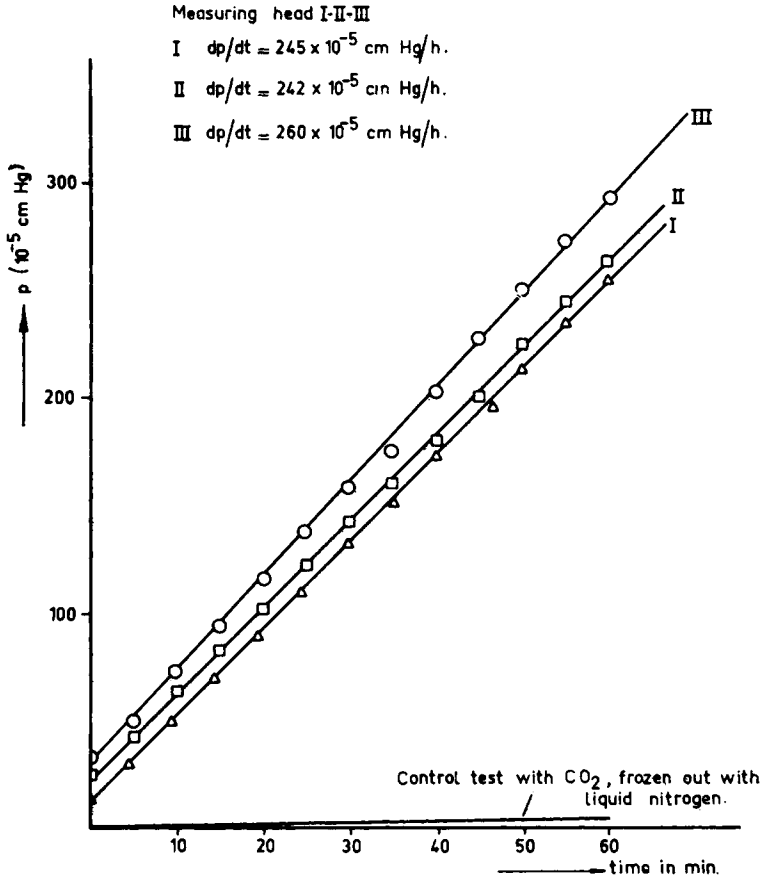


Fig. 5. Oxygen permeability of polyester film.

formation of the vacuum. Any imperfections in the film can be detected immediately: creases show up, and pinholes hinder the building up of a vacuum. For correction air is supplied by means of stopcocks *c* and *d*, and a new film is inserted.

Now the lid is placed on the film (see Fig. 4*b*). The flow of the test gas is switched on with the aid of stopcocks *e* and *f*. The pressure of the test gas is adjusted normally at 30 cm. Hg. It is advisable to run mercury into the channel when testing nonflat film material (blow-extruded) or thick plates (see Fig. 4*c*).

Next, the temperature is controlled. Thus three films are prepared in succession. The mounting of the film works much more quickly with this mechanism and is far easier than with the conventional system using bolts and nuts.

After about 30 min. the mercury diffusion pump is switched on. When the pressure in the measuring chamber has become constant ( $10^{-2}$ – $10^{-6}$  mm. Hg), which takes about 1 hr. for films with a high permeability (poly-



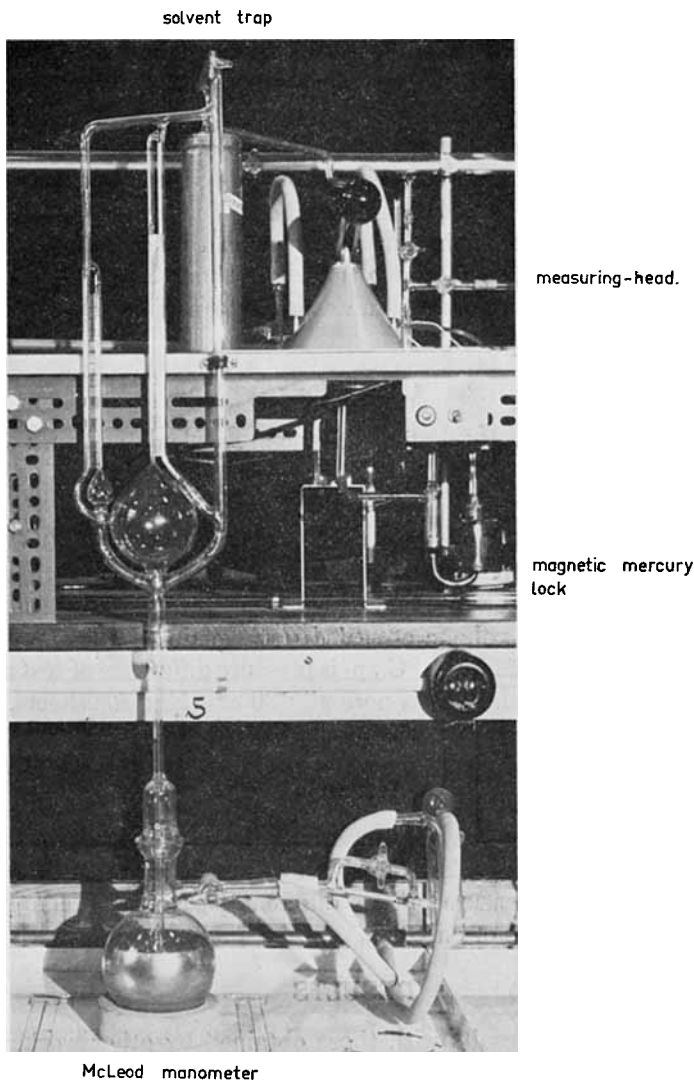


Fig. 6. Gas permeability apparatus.

ethylene) and about 4 hr. for films with a low permeability (saran), the magnetic mercury valve is closed and measuring starts. Owing to concentration equilibrium existing over the film while the vacuum is being formed, the time lag—which is sometimes substantial with very impermeable films—is eliminated. The pressure in the measuring chamber rises proportionately to time, and one pressure value is determined every 5–10 min. for about 1 hr. (see Fig. 5).

When measuring is ended the mercury is allowed to run out of the channel round the film holder, the magnetic mercury valve is opened, and

air is admitted first in the test gas line, and then in the measuring chamber. For measuring films with a low gas permeability, the miniature condensation flasks are cooled with liquefied nitrogen, in order to remove water vapor from the inner glass surface.

For measuring the permeability of the same film to another gas, only the magnetic mercury lock need be opened, the measuring chamber evacuated, and the flow of the other test gas switched on.

On the TNO apparatus six film specimens can be conditioned and the permeability measured at the same time. In about 5 hr. it is possible to measure the permeability of six saran samples to a gas. Figure 6 shows one of the six measuring units.

### Calculations

The gas permeability  $P$  (per film thickness) is calculated from:

$$P = (dp/dt) \{ 273V / [76(273 + t)p_1](O) \}$$

where  $P$  is permeability per film thickness in  $\text{cm.}^3$  (1 atm.,  $0^\circ\text{C.}$ )/ $\text{cm.}^2$ -sec.-cm. Hg,  $dp/dt$  is the measured pressure rise per unit of time in cm. Hg/sec. (if necessary corrected by subtracting the value measured in a control test),  $V$  is the volume of sealed chamber in  $\text{cm.}^3$  = approx. 370  $\text{cm.}^3$ ,  $t$  is temperature of film in  $^\circ\text{C.}$ ,  $p_1$  is pressure difference of test gas over film in cm. Hg. For films,  $p_1$  is normally 30 cm. Hg; for sheets, 50 cm. Hg.  $O$  is the area of film in  $\text{cm.}^2$ , approx. 100  $\text{cm.}^2$ .

### Accuracy

We estimated the accuracy by measuring the same sample in the same measuring head. The standard deviation was not more than 1%.

The same sample measured in six different measuring heads gives an overall scatter of results of only 3%.

### RESULTS

Comparison of our results with those obtained by other workers (e.g., Rogers et al.<sup>3</sup>) has shown very good agreement. Figure 5 gives the measurements of the oxygen permeability of a polyester film (25  $\mu$  thickness). The conditioning time was 4 hr. We took a reading every 5 min. The total duration of the estimation in threefold (measurement and preparation) was  $5\frac{1}{2}$  hr.

From the slope of the graph (Fig. 5) one finds the values for the gas permeability ( $P$ ) given in Table I (temperature  $20^\circ\text{C.}$ ).

When measuring the gas permeability in three concurrent runs, we often find considerable differences among the three samples. Changes in thickness, especially play a part in the scatter of results. Important information about these thickness variations of a film is obtained by measuring with a micrometer eyepiece the thickness of sections made with a freezing microtome (see Fig. 7).

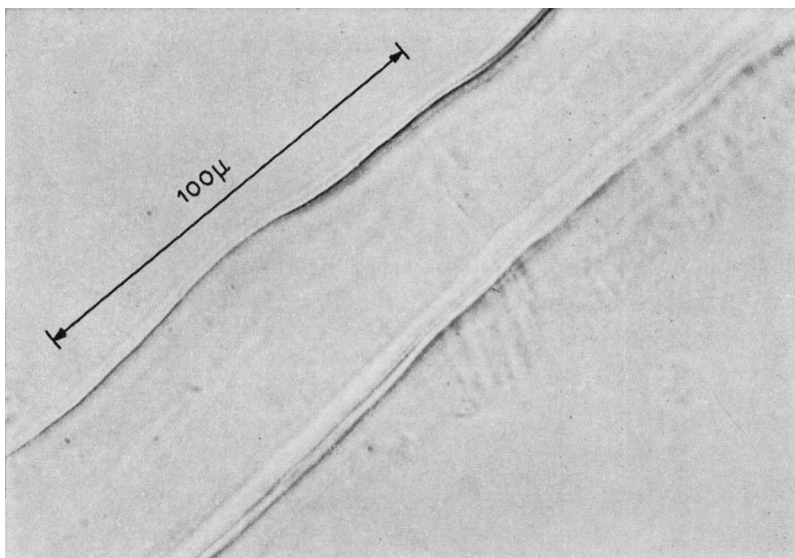


Fig. 7. Section of a polyethylene film made with a freezing microtome.

For the determination of the thickness of the material, i.e., nonlaminates, the specimens are weighed with the aid of an analytical balance prior to measuring of the gas permeability. After this measuring, the density of the specimen is measured by the density-gradient technique, ASTM Designation D1505-57T. The thickness is calculated from weight, density, and area of the specimen.

### Control Test

To determine any air leakage and the presence of foreign gases originating from the measuring chamber during measuring, carbon dioxide is taken as the test gas. The permeated carbon dioxide is frozen out in the miniature condensation flask with Dewar filled with liquid nitrogen. The permeation now measured shows the presence of foreign gases (see Figs. 5 and 8).

TABLE I  
Oxygen Permeability of a Polyester Film (25  $\mu$  Thickness)

Measuring head	Pressure difference over the film	$dp/dt$ , cm. Hg/hr.	$P$	$P$
	$p_1$ , cm. Hg		(per film thickness) $\text{cm}^3 \text{ (S.T.P.)} / \text{cm}^2 \text{-sec.-cm. Hg}$	$\text{cm}^3 \text{ (S.T.P.)-mm.} / \text{cm}^2 \text{-sec.-cm. Hg}$
I	30	$245 \times 10^{-5}$	$11.9 \times 10^{-10}$	$0.30 \times 10^{-10}$
II	30	$242 \times 10^{-5}$	$11.5 \times 10^{-10}$	$0.29 \times 10^{-10}$
III	30	$260 \times 10^{-5}$	$12.5 \times 10^{-10}$	$0.31 \times 10^{-10}$

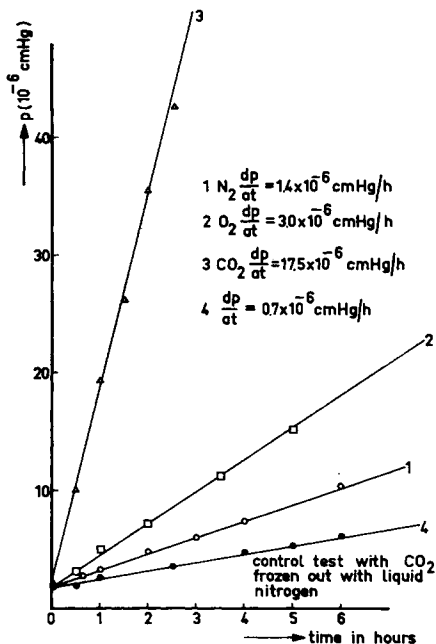


Fig. 8. Gas permeability of a laminate consisting of cellophane-aluminum-saran.

Thus for the control test the same specimen, under the same conditions as those in the test procedure, is used.

This measuring can be done simultaneously with the measuring of the permeation of carbon dioxide by determining a pressure value, alternately with and without the use of the Dewar filled with liquid nitrogen.

The measured pressure rise per unit of time ( $dp/dt$ ) can now be corrected by subtracting the value measured in the control test (see Fig. 8).

### “Gas-Tight” Films

Most “gas-tight” films which are presently available include combinations of aluminum and/or paper with cellophane and/or saran.

The highest influence on the gas permeability of a laminate with paper is obtained in the case of: (1) a good adhesion between plastic film and paper; (2) “tight” smooth papers (e.g., glassine).

For instance a Rilsan-glassine combination (Rilsan = nylon 11) gave a gas permeability 300 times lower than the plain Rilsan film. A Videne ( $50 \mu$ )-glassine laminate (Videne = polyester) had the extremely low oxygen permeability per film thickness at  $20^\circ C$ . of  $P_{O_2} = 2 \times 10^{-13} \text{ cm.}^3$  (S.T.P.)/ $\text{cm.}^2 \text{ sec. cm. Hg}$ .

The effect is explained by the big decrease of the “working” surface of the plastic film by the paper. Actually, the paper fibers themselves show very low gas permeabilities.<sup>7</sup>

Aluminum foil less than 30  $\mu$  in thickness is not an absolute gas barrier, because the foil usually contains a number of perforations, the number depending on the thickness of the foil. These perforations (pinholes) are usually small, but they greatly influence the gas permeability. In combination with a plastic film, the aluminum has the same action as the paper.

We measured the gas permeability ( $O_2$ ,  $N_2$ , and  $CO_2$ ) of a laminate consisting of cellophane MSAT, 30 g./m.<sup>2</sup>; aluminum, 56 g./m.<sup>2</sup> (thickness about 21  $\mu$ ); and saran, 30 g./m.<sup>2</sup>. The measurements were carried out at a temperature of 20°C. and a relative humidity of the cellophane of about 50%. The conditioning time was 12 hr. The differential pressure of the test gas over the film was 30 cm. Hg. The results are shown in Figure 8 and Table II.

TABLE II  
Gas Permeability of a Laminate of Cellophane-Aluminum-Saran

Gas	$dp/dt$ , cm. Hg/hr.	$dp/dt$ , corrected for the blank	$P$ (per film thickness), cm. <sup>3</sup> (S.T.P.)/ cm. <sup>2</sup> -sec.-cm. Hg
$O_2$	$3.0 \times 10^{-6}$	$2.3 \times 10^{-6}$	$11 \times 10^{-13}$
$N_2$	$1.4 \times 10^{-6}$	$0.7 \times 10^{-6}$	$3.5 \times 10^{-13}$
$CO_2$	$19.5 \times 10^{-6}$	$16.8 \times 10^{-6}$	$80 \times 10^{-13}$

The results show that the inequality of the permeability of plastic films is still valid, i.e.,  $P_{CO_2} > P_{O_2} > P_{N_2}$ .

### Relative Humidity

For the testing of material with relative humidity as a parameter, the test gas flows through a washing bottle filled with a suitable salt solution, mounted in the gas supply line of each film holder. A 100% relative humidity is realized by using water in the washing bottle, or a layer of water on top of the specimen.

The influence of relative humidity on the permeability of hydrophilic films (e.g., plain regenerated cellulose) is large, as the degree of swelling is influenced by the percentage of moisture. This degree of swelling is also dependent on the structure of the material, which in its turn is defined by the way of preparing, the pretreatment, the solvent in the case of a coating, the adhesion in the case of a laminate, etc.. Such films had to be conditioned at the same relative humidity on both sides of the film (entry and exit side).

In contrast with the assertions of Buchner,<sup>8,9</sup> Pilar,<sup>10</sup> and Fricke<sup>11</sup> that such measurements are impossible with the high vacuum technique in that the exit side is at zero relative humidity, the problem can be solved by placing a "sieve" film under the hydrophilic specimen to be tested. This is a film with a high gas permeability but a low water vapor permeability (polyethylene is found suitable for this purpose).

In this case, the relative humidity in the interface can be calculated as follows (see Fig. 9).

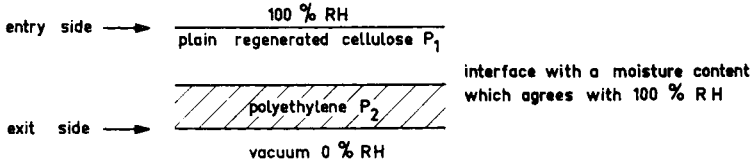


Fig. 9. Relative humidity of a plain regenerated cellulose film determined by using a "sieve" film.

The rate of permeation  $q$ , through each layer must be constant under steady-state conditions, and if we assume that the permeability constants ( $P_1, P_2$ ) are independent of pressure:

$$q = P_1(p_1 - p_2)/x_1 = P_2(p_2 - p_3)/x_2$$

where  $p_1, p_2, p_3$  are pressures of the gas on the surface in equilibrium with the film and  $x_1, x_2$  are the thicknesses of the film layers.

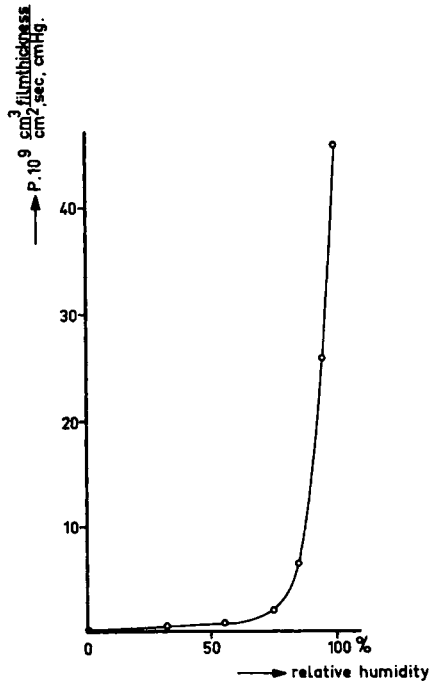


Fig. 10. Influence of the relative humidity on the oxygen permeability of a plain regenerated cellulose film.

In the case of two layers where subscripts 1 = cellulose, 2 = polyethylene with the same thickness ( $x_1 = x_2$ ) and where the relative humidity is 100% at the entry side of layer 1, and zero at the exit of layer 2, the relative humidity at the interface ( $RH_2$ ) can be calculated as follows:

$$RH_2 = 100\% (1/P_2)/(1/P_1) + (1/P_2) \simeq 100\%$$

where  $P_1$  is the water vapor permeability of plain regenerated cellulose =  $5000 \times 10^{-9}$  cm.<sup>3</sup>/cm.<sup>2</sup>/sec./mm./cm. Hg;  $P_2$  = water vapor permeability of polyethylene =  $90 \times 10^{-9}$  cm.<sup>3</sup>/cm.<sup>2</sup>/sec./mm./cm. Hg.

Thus we can determine the gas permeability of a hydrophilic film, while the whole film is conditioned at a constant relative humidity. Although this theory employs "ideal" laws of permeability, for water vapor it should give a fair approximation to the prevailing system.

The influence of the relative humidity on the oxygen permeability of a plain regenerated cellulose film is illustrated in Figure 10.

We can also use the "sieve" film in the case of a laminate, but mostly the hydrophilic film is laminated with a hydrophobic material in order to decrease the water vapor permeability. In this case the film is measured with the polyethylene or saran side immediately at the high vacuum (influence of the relative humidity on the gas permeability of polyethylene or saran has never been found).

When the hydrophilic material is coated on both sides with a hydrophobic film it is necessary to precondition the film in the atmosphere of interest, because it is too time-consuming to do this on the gas permeability apparatus.

## CONCLUSION

Thousands of permeability measurements have already been made with the apparatus during the last four years, both for research and industrial purposes, and in our opinion the apparatus is universally applicable to all permeation problems and fulfills the requirements of a rapid, accurate, and versatile procedure.

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### Résumé

Le Laboratoire Central TNO-Delft (Pays-Bas) a conçu et mis au point un appareil de mesure de la perméabilité aux gaz des films et feuilles plastiques. Cet appareil se compose d'un élément à vide relié à six cellules à mesure de conception et de construction nouvelles. La pression du gaz qui entre dans un volume constant à travers le film est mesurée à l'aide de manomètres McLeod. L'appareil fut développé pour permettre la mesure précise et rapide de matériaux de basse perméabilité, mais il peut également servir pour des films de haute perméabilité. La gamme des perméabilités à mesurer est comprise entre  $10^{-7}$  et  $10^{-13}$  cm.<sup>3</sup> (S.T.P)-épaisseur de film/cm.<sup>2</sup>-sec.-cm. Hg. La durée des opérations de mesure pour la gamme des  $10^{-13}$  est de 6 heures environ (en cas des laminés avec de l'aluminium), et pour la gamme des  $10^{-7}$  d'environ 15 minutes (en cas de polyéthylène). Les opérations de mesure peuvent être effectuées simultanément dans six cellules à mesure et aisément par températures et humidités relatives variées. L'appareil convient à la fois aux fins de recherche scientifique et aux opérations de mesure courantes dans l'industrie.

### Zusammenfassung

In dem Zentrallaboratorium TNO, Delft, Niederlande, wurde ein Gerät zur Messung der Gasdurchlässigkeit von Folien und Platten entwickelt. Das Gerät besteht aus einer Hochvakuumeinheit, welche mit sechs gänzlich neu entworfenen und neu konstruierten Folienhaltern verbunden ist. Der Druck des Gases, das durch die Folie hindurchdringt in einem konstanten Volumen, wird mit Hilfe McLeod-Manometern gemessen. Das Gerät wurde entwickelt um schnellere und zuverlässigere Messungen von Materialien mit sehr niedriger Durchlässigkeit zu ermöglichen; es kann jedoch ebenfalls für Folien mit einem hohen Durchlässigkeitsgrad gebraucht werden. Der Bereich der zu messenden Durchlässigkeiten liegt zwischen  $10^{-7}$  und  $10^{-13}$  cm.<sup>3</sup> (S.T.P.)-Dicke der Folie/cm.<sup>2</sup>-Sek.-cm. Hg. Die Messzeit für den  $10^{-13}$ -Bereich beträgt ungefähr 6 Stunden (z.B. bei Verbundfolie mit Aluminium). Die Messzeit für den  $10^{-7}$  Bereich beträgt ungefähr 15 Minuten (z.B. bei Polyäthylen). Es können sechs Messungen gleichzeitig ohne Schwierigkeiten durchgeführt werden bei verschiedenen Temperaturen und relativen Feuchtigkeitsgraden. Das Gerät ist sowohl für Forschungszwecke wie auch für routinemässige Messungen in der Industrie zu verwenden.

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