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# DIFFUSIVE GAS EXCHANGE IN RECIRCULATIVE RECOVERY OF THE AIR RATIO IN CLOSED APPARATUS AND ROOMS

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## Introduction

The present level of industrial technologies strictly requires that the environment should be protected against the discharge of microparticles and of biological objects, and that a certain microclimate should be maintained in the production area of the cleanrooms. First of all, it concerns the precision-sterile conditions of an air (gas) atmosphere. Gas supply to various enclosed spaces and rooms depends on the specific features of the production procedure and the industrial branch where it is applied. Some modern industries (electronic industry, instrument engineering, biotechnology, medical industry, etc), many research activities (gene treatment, work with chemically and biologically dangerous agents) are impossible without air and gas media thoroughly purified of dust and micro-organisms.

Air purification is most important for the production of large-scale integration circuits and in biotechnology where sterile cultivation conditions are required. In fact, even a minimal amount of alien microflora that has penetrated into the nutrient of a fermenter with a flow of air can inhibit the cultivation, and just several microparticles 0.10 μm in size in a cubic foot of air in the IC production area considerably increase the number of spoiled items (e.g. in production of 1 Mbyte memory units). So, in the air supplied to sterile biotechnological industries and emitted afterwards (ecological requirement) there must be no micro-organisms, and in microelectronics strict limits are imposed on the size and number of microparticles in the air of production areas (e.g. in production of 4 Mbyte MOS memory units there must be no particles of size larger than 0.06 μm, and the density of defects at the 10% output of serviceable chips must not exceed 0.05 defect (cm )/layer [1]).

At present the problems of air (gas atmosphere) purification are being solved in a traditional way by direct filtration through sterilizing filters and fine filters. In this case the atmospheric air undergoes passing purification and sterilization, is supplied to an enclosed space or room, partially re-circulated and then emitted into the air through filters, if necessary (when there are harmful microparticles or micro-organisms). To prevent contamination of enclosed spaces with micro-objects, an excessive air pressure is used.

However, even with partial re-circulation of the purified air the energy consumption for maintaining the required quality of the air is mainly determined by the hydro-dynamic characteristics of the filter elements, whose service life is limited because of constant dust attack.

In this paper we propose a new approach to the supply of clean (sterile) air to enclosed spaces to prevent microparticles (micro-organisms) from penetrating both into the enclosed space and back into the atmosphere. It is the air recirculative recovery based on diffusive gas exchange through a polymeric nuclear track membrane.

## 1. Principle of operation of the gas supply system

The principle of operation of a gas exchange device is shown schematically in Fig.1.

The air of the enclosed space 1 with the technological equipment (with a culture medium in the case of a fermenter) passes through gas exchange device 2 and returns back. The air is blown through the channels of the gas exchange device along the surface of the nuclear track membrane by using forced-draught fan 3. Along the other membrane surface the atmospheric air is blown through by fan 4 in the mutually perpendicular direction. The



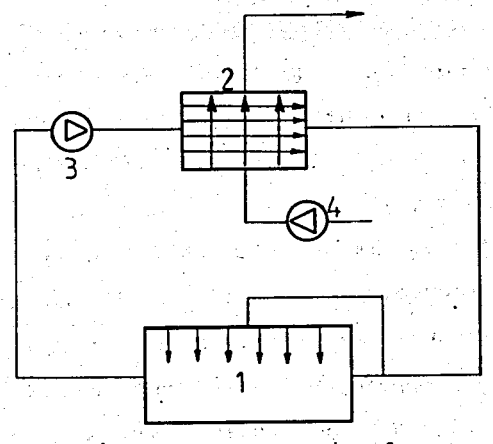


Fig.1. Gas exchange arrangement: 1 - enclosed space with technological equipment; 2 - gas exchange device; 3 - force-draught fan; 4 - atmospheric air fan.

composition, humidity and temperature of the blown-through air depend on the required microclimate conditions of space 1. There is practically no pressure difference on the membrane (there are no reasons for the membrane being clogged), and the gas exchange through the pores occurs owing to a difference in the partial pressures of the air components. This is how oxygen and nitrogen are supplied to the enclosed space and excess carbon dioxide and other gaseous compounds are released. Since nuclear track membranes are made of polymeric films 5-10 μm thick with pores of equal size and regular geometry [2,3], they have minimal diffusive resistance that is limited mainly by the near-wall air layers. Penetration of microparticles and micro-organisms less than 0.5 μm in size through a membrane with pores over 0.50 μm in size is possible only due to diffusion in the air. The experiments show, however, that microparticles of this size are either repelled by the electrostatic field of the membrane (the quantitative estimation of the charge was done only for polycarbonate nuclear track membranes in Ref.[4]) or captured by

the front or intrapore surfaces of the membrane, depending on the kind of the particle. Indeed, for the ratio $l/D \geq 50$ (l, D are the length and diameter of the pores) each microparticle collides more than once with the surface of the pore, which makes the retaining ability membrane of the practically as high as 100%.

To avoid the contamination of the enclosed space with alien microflora and throw-out of cultivated cells and their fragments into the atmosphere (when a pressure difference or favourable growing conditions appear on the membrane surface), the diameter of the membrane pores is chosen so to ensure a 100% retaining ability against cells [5].

Fig.2 shows a lay-out of a gas-exchange device with a pack of perforated polymeric plates with nuclear track membranes of

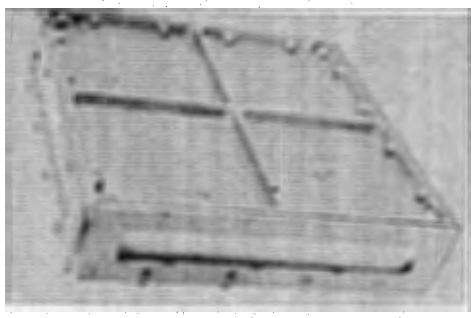

Fig.2. General view of the gas exchange device.

polyethelene terephthalate film 10 μm thick (a similar device was tested by the authors in direct filtration of air in clean zones [6,7]). The working surface of the membrane was 1.6 $m^2$ . The pore diameter was chosen to be 0.1 μm, which is the minimal diameter for the the Knudsen number to satisfy the flow regime described by the Poiseuille expression. The fans of this device provided the air flow rate 60 $m^3$ /h, the input-output pressure difference being ~22 Pa.



## 2. Test results and discussion

### 2.1. Biotechnological processes

This gas supply system was tested at a biochemical plant producing glutamic acid (Luzhany). The gas-exchange device was a single-membrane flat unit with the working surface of the membrane 0.06 $m^2$ . The unit was connected to the Ankum fermentation apparatus 10 l in volume (see Fig.1). The experiments included cultivation of Corynebacterium glutamicum, which produces glutamic acid, and Saccharomyces cerevisiae yeast. The dimensions of the cells are 0.5-1.0 μm (diameter), 0.8-2.1 μm (length) and 2.2-3.5 μm, 4.0-5.5 μm, respectively.

The critical pore dimension, i.e. the largest of those preventing the culture cells from penetrating or growing through the membrane, was chosen on the basis of experimental calibration curves. For this purpose the culture cells were put onto the nuclear track membrane placed in Petri dishes with meat-peptone agar (since the membrane surface is hydrophilic, the cells had a free access to the nutrient). The membrane pore dimensions in the interval 0.10-2 μm were investigated. The results are shown in Fig.3 as the dependence of the retaining efficiency of a membrane on the dimensionless parameter d/D (d, D are the minimal diameter of the cells and the diameter of the pores). The gas-dynamic pore

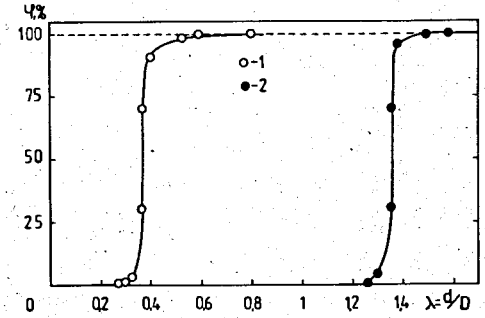


Fig.3. Retaining efficiency of the membrane as a function of the parameter d/D; 1 - Corynebacterium glutamicum; 2 - Saccharomyces cerevisiae.

diameter was used for calibration (the technique and the facility are described in detail in Ref.[8]). The attempts to plot similar curves for the electron-microscopic pore diameter and the one obtained by the bubble method were ineffective because of large variance dispersion in values. Electron microscopy yields information on the surface dimension of the pores [9] while the bubble method [10,11] allows one to obtain information on the narrowest part of the channel. Owing to the specific features of both techniques one cannot obtain complete information, e.g. because of double and more complex overlapping of pores which are most preferable for growing through. On the other hand, the gas-dynamic method, which yields the average value of the pore diameter, is the best to take into account the double and more complex overlapping of pores (the air flow through a pore is proportional to its diameter to the 4th power).

The calibration curves shown in Fig.3 allow the conclusion that the critical gas-dynamic size of pores for Corynebacterium glutamicum and Saccharomyces cerevisiae is 0.8 and 1.5 μm. It should be emphasized that the calibration curves are important only for the choice of the membrane size for a gas-exchange device if there are favourable conditions for their life on the membrane surface, or if there is an accidental pressure difference on the membrane.

The Ankum fermenter gas supply system was tested using a gas exchange device with pores 0.10 and 0.50 μm in diameter. The oxygen-based results obtained for the degree of gas exchange for both cultures are shown in the following Table.

Oxygen concentrations were measured with a GCP-100 chemical gas analyser. The throw-out of cells into the atmosphere and contamination of the biomass with alien microflora were not observed.

The test results show that the oxygen- and CO2-based degree of gas exchange in micro-organism cultivation is satisfactory



Table

Oxygen-based results on gas exchange in the gas exchange device connected to the Ankum fermenter (the results are averaged over the measurements in two cultures)*

| D, μm | $O_2$ concentration at Ankum outlet, % | $O_2$ concentration at Ankum inlet, % | $O_2$ concentration in atmosphere, % | $O_2$ concentration as emitted into atm., % |
|---|---|---|---|---|
| 0.10 | $16.4\pm0.9$ | $20.5\pm0.4$ | $21.87\pm0.09$ | $20.7\pm0.4$ |
| 0.50 | $18.9\pm0.5$ | $20.8\pm0.6$ | $21.80\pm0.16$ | $20.90\pm0.08$ |

* Test conditions were identical for both cultures.

both with 0.10 and 0.50 μm pores. The absence of the throw-out of cultivated cells and of contamination with alien microflora proves that the sterile conditions are maintained with this gas supply method.

2.2. Microelectronics

Applied to optimization of gas supply for cleanrooms (CR) in microelectronics (supply of oxygen, removal of carbon dioxide, water vapour and other volatile compounds), this method implies the use of much less effective fine filters (e.g. FPP and FPA produced in the USSR) against internal microimpurities than those used now (HEPA and ULPA). The gas supply system was experimentally tested in a clean chamber 3 $m^3$ in volume with a working team of 3 men. Two gas exchange devices with pores 0.27 and 0.10 μm in diameter and average porosity of 7% were used. Oxygen and carbon dioxide concentrations were measured with a GCP-100 gas analyser.

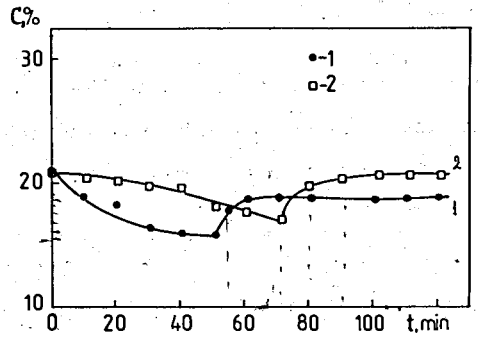


Fig.4. Oxygen concentration variation in time for the 3 $m^3$ chamber: 1 - the team did an easy physical work; 2 - the team had a rest; the point of inflection corresponds to the moment of switching on the gas exchange device.

The oxygen- and carbon-dioxide-based results of measurements of the degree of gas exchange in the chamber are shown in Figs.4 and 5, respectively. The moment of switching on the gas supply system coincides with a considerable excess of the maximum permissible concentration (MPC) of carbon dioxide for the working team. When the gas exchange device with 0.27 μm pores was tested, the team did an easy physical work. An increase in the carbon

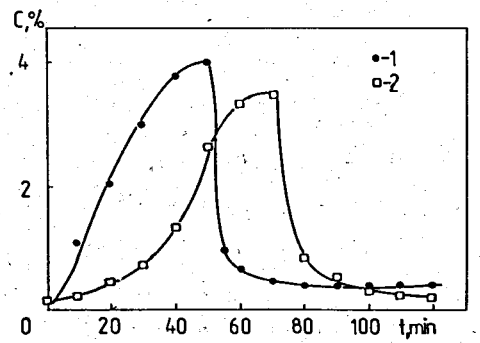


Fig.5. Carbon dioxide concentration variation in time for the 3 $m^3$ chamber: 1 - the team did an easy physical work; 2 - the team had a rest; the point of inflection corresponds to the moment of switching on the gas exchange device.



dioxide concentration has shown it. When the membrane with 0.1 μm pores was used, the team had a rest. (In both cases the air in the CR was cleaned with FPP filters, class 100 for particles of 0.5 μm in size of US Federal Standard 209 D being maintained.)

The experimental results obtained were approximated by the exponential relation $C = C_o \exp(-t/\tau^*)$, where $C_o$ is the initial concentration of the carbon dioxide at the moment of switching on the gas supply system; $\tau^*$ is the relaxation time for the concentration of the given component. The least-squares method (LSM) was used to estimate the relaxation time values in each case. They turned out to be almost equal; the average value for the given gas exchange device was $(12.5 \pm 2.0)$ min for a 3 $m^3$ chamber. These results prove to a large extent that the degree of gas exchange is practically independent of the nuclear track membrane pore size. Similar measurements were carried out with a gas exchange device connected to a box 0.4 $m^3$ for chemical and biological investigations. The nuclear track membrane had pores 0.1 μm in size. A schematic view is shown in Fig.2.

In the vacuum atmosphere of the box there were acetone vapours of concentration C = 600 $mg/m^3$. After switching on the

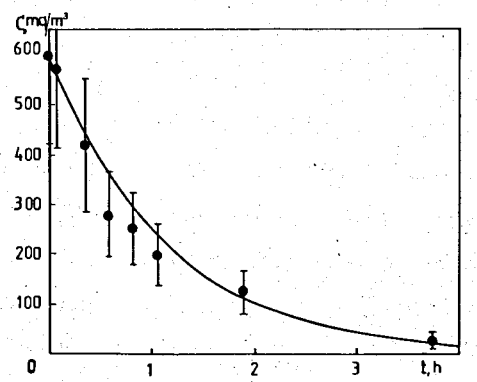


Fig.6. Variation of the acetone vapour concentration in the 0.4 $m^3$ box for chemical and biological investigations.

fans of the gas exchange device the acetone vapour contents began to decrease according to the exponential law (Fig.6). The concentration was measured with an UG-2 universal portable gas analyser, the measurement error being $\pm$ 30%. As in the case of measurements in the CR 3 $m^3$ in volume, the results were analysed by the LSM on the basis of the exponential relation. The value $\tau^* =$ 1.15 h was obtained. The MPC, which is 200 mg/$m^3$ for acetone vapours, was achieved in an hour. It can be explained by a relatively low coefficient of acetone diffusion in the air (as compared with the carbon dioxide) and by an insufficient air exchange ratio in the box. Nevertheless, the results clearly show the possibility of gas supply for small enclosed spaces where different investigations are carried out.

## Conclusion

The results of the experimental tests of the gas supply system to different enclosed spaces in biotechnology, microelectronics and other possible fields have shown that even nuclear track membranes of small area ensure the degree of gas exchange, e.g. for oxygen and carbon dioxide, which is sufficient for survival of the staff and cultivated cells. For instance, to provide sufficient supply of oxygen and removal of carbon dioxide required for one person, it is enough to have 0.5 $m^2$ of nuclear track membranes per 1 $m^3$ of the space at the air flow rate 60 $m^3$/h. A decrease in the degree of gas exchange is not observed for the membranes with pores down to 0.10 $\mu$m in size (a smaller diameter can worsen gas exchange owing to the transition of the gas molecule from to the Knudsen pattern of gas molecule flow in the channels).

Contamination of the internal air with microparticles and alien microflora and penetration of cultivated cells through the nuclear track membrane were not observed.



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