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USE OF CHROMATOGRAPHS IN COMMERCIAL AUTOMATED MONITORING OF TRACE IMPURITIES AND CONTROL SYSTEMS FOR HELIUM PRODUCTION

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SUMMARY

An automated monitoring and control system using various chromatographs has been designed for the solution of a number of problems associated with the measurement of analytical parameters and close control of production processes. The chromatographs employed enable the production of pure gases of a required composition.

INTRODUCTION

The efficient monitoring and control of modern processes involves the use of a large body of information on the production flow composition. Of special importance is the case of pure gases, particularly helium.

Helium is separated from natural gas by a low-temperature condensation followed by evaporation and purification of the helium concentrate. The production capacity in terms of natural gas may be as high as $3 \cdot 10^9$ m^3 per year.

In order to obtain highly pure products, the chromatographs should be capable of monitoring impurities down to at least $1 \cdot 10^{-5}$ %, (v/v) the need for technological optimization and minimization of losses means that the desired products should be monitored down to at least $1 \cdot 10^{-4}$% (v/v)[1].

EXPERIMENTAL

An automated information system was designed as an integral part of an high-capacity helium production facility to solve the following problems (Fig. 1): monitoring of the product composition; classification of the products; indication and warning of faults; presentation of technological documents and certificates for the products; diagnosis, indication and warning of the chromatograph and measuring channel conditions; testing of the channel calibration stability and determination of the calibration factors; individual channel adjustment in the start-up and service modes.

Chromatographic information is useful in controlling the production flow as

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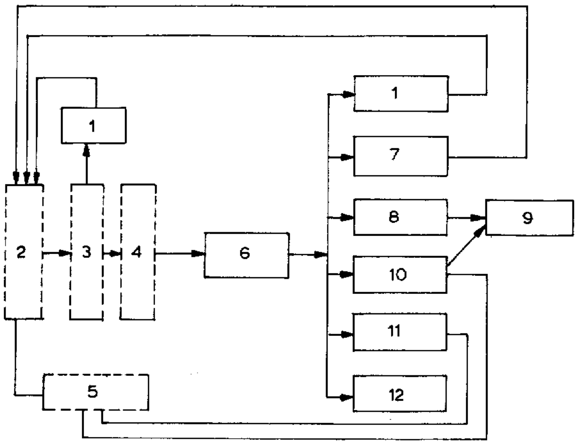


Fig. 1. Functional tasks of the automated monitoring and control system and the corresponding information flows: 1 = control of production parameters; 2 = process equipment; 3 = chromatograph; 4 = voltage-to-frequency converter; 5 = operator; 6 = monitoring of product composition; 7 = switching of the equipment; 8 = helium classification; 9 = document output; 10 = warning of deviations; 11 = parameter optimization; 12 = monitoring of the system's state, independent check-out of the channels.

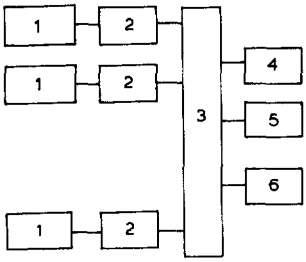
well as in the regulation and optimization of the processes. In this case the information output is fed both to video terminals, dispatcher's printers and directly to the control circuits for local production parameters.

The multiple-channel information-control system comprises chromatographs, voltage-to-frequency converters and a central computing complex (Fig. 2). It is made up of as many as 150 measuring channels. The chromatographs can be operated either in an in-flow (continuous) mode or periodic mode, *i.e.*, when they are connected to the containers to be analyzed. The detector output voltage (0–200 mV) is converted in to a proportional frequency signal (10^3–10^7 pulses per second) and fed into the digital pulse signal input modules forming part of the computing complex which incorporates a central processor. The processor makes it possible to perform a space-saving solution of identical problems associated with the treatment of chromatographic information and close control functions. The operating converter frequency is determined by the broad dynamic measurement range of the chromatographs.

The system operates in the on-line mode. The structure of the algorithmic software is in accordance with the features of the production process and the method of chromatographic signal processing. The system meets rigid requirements for enquiry intervals (0.5 s), a large number of computations and printed documents (up to 4000 per day).

The concentrations of the components are determined from the integrated chromatographic peak areas, taking into account baseline drift and individual calibration factors obtained with the aid of absolute calibration. The algorithm contemplates determination of the characteristics of the peaks due to the components to be analyzed, such as their origin, time taken to attain a maximum and termination.

An integrated value stored in the modules during the period between two enquiries serves as a measure of the instantaneous chromatographic signal. This allows



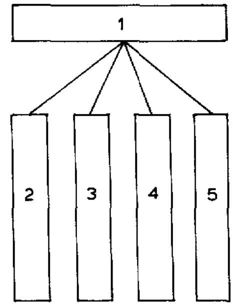


Fig. 2. Block-diagram of the automated analytical system: 1 = chromatograph; 2 = voltage-to-frequency converter; 3 = computing complex; 4 = printers; 5 = panels of the operators and attendant personnel; 6 = displays.

Fig. 3. Programs (1) for system control and independent check-out: 2 = channel check-out; 3 = channel-interference testing; 4 = analysis of chromatographic information; 5 = calibration.

the high-frequency noise to be filtered out and information loss between the two successive enquiries to be eliminated thereby avoiding relatively cumbersome smoothing procedures. This calculation of the chromatographic peak areas is of a fairly good accuracy, so that the mean-square deviation of the integrated values within the peak does not exceed 1%.

To enhance measurement reliability, the characteristic peak points are determined by two last successive measurements. The measuring channels are timed by generation of a zero signal frequency at the onset of an analytical cycle in each of the channels.

The chromatographs provide the a major proportion (over 75%) of the analytical information in the automated helium production facility. The principal problems are the determination of the helium concentration in the intermediate products and natural gas ($1 \cdot 10^{-4}$–$1 \cdot 10^{-2}$%, v/v the hydrogen concentration in helium in the process of its purification (at the same levels) and the concentrations of a number of impurities ($1 \cdot 10^{-5}$–$1 \cdot 10^{-1}$%, v/v) in commercial helium.

Taking into account the requirement for high reliability, coupled with some limitations on the chromatograph maintenance in industrial situations, the above problems call for the use of the most popular and already practically tested detectors that sense variations in heat conduction and in the heat of combustion. To reduce the lower measurement limit, the chromatographs provide for a preliminary enrichment of the sample in the components to be analyzed.

The helium content in the products at the botton of the evaporation columns is determined by a frontal-adsorption concentration of the less sorbable impurities on evacuated columns. This gives a 100–200-fold increase in the content of helium for the final analysis. Approximately the same concentration effect can be attained in a chromatograph for hydrogen analysis in helium[2] by a two-stage process with a discharge of the major components. In the first stage, hydrogen, together with the residual amount of helium, is fed from the outlet of the separation column to its inlet. The second stage provides for an efficient separation of the hydrogen from the residual helium. The analytical cycle does not exceed 20 min.

For the analysis of impurities in helium whose sorption capacity is higher than that of the major components such as hydrogen, neon, oxygen and argon, methane, carbon monoxide and dioxide, a method of thermodynamic concentration was used. The automated commercial chromatograph developed for this purpose with a two-channel storage permits accumulation of impurities on two successive columns packed with different sorbents at liquid nitrogen temperature. Desorption is effected into two independent gas streams each of which is equipped with a heat conduction detector. One of them is additionally fitted with a heat of combustion detector for determining hydrogen as an impurity. The computing complex is connected to the chromatograph through three independent frequency channels.

Despite the fact that helium chromatographs are rather sophisticated in design, they have proved to be commercially efficient. The first types of such chromatographs have been in operation since 1978.

To calibrate the chromatographs by theoretical and experimental techniques, a method was proposed ensuring optimum values in terms of relative concentration measurement errors[3]. A program for determining and correcting the channel calibration factors forms part of the mathematical software of the present system.

To ensure correct operation of the chromatographs regularly connected to the cyclic processes, the system incorporates special panels designed as an interface between the computer and the attendant personnel, and for indication of the system's modes and input of command signals into the computing complex.

Important among the functional tasks of the system are the indication and warning of faults. Based on the data stored in the computing complex, the system gives warnings of deviations of the process parameters, evaluates the state of the chromatographic equipment and provides signals on the absence of peaks or their mismatch with the preset limits. The fault signals are discarded.

The efficiency and methodological characteristics of the measuring channels are evaluated by use of control and independent check-out programs, thereby accomplishing a set of tasks ensuring the operating availability of the channels (Fig. 3). Calibration and testing of the channels are performed on all the channels, which reduces the risk of obtaining unreliable results and the time taken to perform start-up and routine operations.

RESULTS AND CONCLUSIONS

The operational reliability of the chromatographs in the system favours an automated monitoring of helium and preparation of certificates for the finished product.

The satisfactory results obtained upon introduction of the described information and control system into large-scale helium production facilities, its efficient operation and resolution of a broad spectrum of problems offer the possibility of applying such systems to situations where the use of chromatographs for monitoring trace impurities is essential.

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