POSSIBILITIES OF AUTOMATIZATION IN RADIOCHEMISTRY

G. TÖRÖK, L. GY. NAGY, J. RUIP, M. VODICSKA,
N. VAJDA and J. SOLYMOSI
Department of Applied Chemistry,
Technical University, H-1521 Budapest

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Summary

In radiochemistry chemical processing operations of high work and time demand form today the bottleneck of these systems. Therefore, in our days the automatization of chemical operations becomes more and more a necessity. Operational units (microvalves, debubblers, columns suitable for the construction of variable chemical units of modul system) were developed for this purpose. The automatic control of the processing unit was solved by an electropneumatic control unit and program control by means of a personal computer. This automatized processing system can be widely used in radiochemistry and in other fields.

Introduction

In radiochemistry, at a high automation level of nuclear measurement and data processing there is a remarkable shortfall in the automatization of (radio)chemical processing operations. Therefore, in our days chemical processing operations form the bottleneck of radiochemical systems. Thus, their automatization became a topical task. A precondition of the realization — besides a well defined processing technology — consists in electrically or pneumatically controlable operational units and elements (microvalves, columns, debubblers), further the availability and expert application of a computer suitable for program control.

By the automation of radiochemical processing subjective error, arising from the mistake of the operator, can be completely eliminated, and a further advantage is that personnel radiation dose substantially decreases, as compared to manual handling. Processing series of a large number of samples these factors emphasize all the more the necessity of automation.

In this paper a few operational elements of univerSal applicability, developed at the Department of Applied Chemistry of the Technical University, will be described, further a suitable form of realization of the radiochemical processing unit, controlled by a desk computer will be shown.

Precedents

Among the radiochemical processes reactor neutron activation analysis has been often used at our Department from the early seventies to solve element-analytical tasks, concerning mainly biological and environmental samples. A common characteristic of samples of this kind is that suitable sensitivity during analysis can be provided only, if the isotope to be measured and the isotopes responsible for the matrix activity are separated from one another after activation. Therefore, the processing of samples was carried out from the beginning by radiochemical (destructive) method. Moreover, the scintillation measuring technique of the initial period often requested the individual and/or group separation of the isotopes to be measured.

It became evident within a short time that the digestion of the samples and separation operations are very work- and time-consuming, so that using conventional equipment and technique, and manual operation satisfactory productivity cannot be achieved during the series analysis of samples. Therefore a chemical processing system was designed and constructed to realize operations of this character in a remote controlled and programmed way.

Chemical processing system

The first mechanical variant of the equipment, making possible manual remote control with the aid of an electropneumatic control unit, was constructed in 1978. This was followed by further modernized prototypes. The programmed, remote controlled unit of the most recent equipment was realized by means of a desk computer ZX Spectrum, which controls the chemical processing unit according to a time program via the interface and the electro-pneumatic converter.

The flow chart of the chemical processing unit is shown in Fig. 1, the block diagram of the equipment in Fig. 2.

Characteristics of the whole equipment and its main units can be readily seen in Fig. 1.

The functional connection of the chemical processing part and the programmer unit can be followed in Fig. 2. Elements of the programmer unit are: personal desk computer ZX Spectrum, further a tape recorder as magnetic background program file, television as display, and the interface, transmitting the signal of the programmer to the electro-pneumatic control unit. The control unit generates the electric and pneumatic signals, needed for the control of the operational elements of the chemical processing unit. The order and the duration of the operations to be performed indicated as an example in the

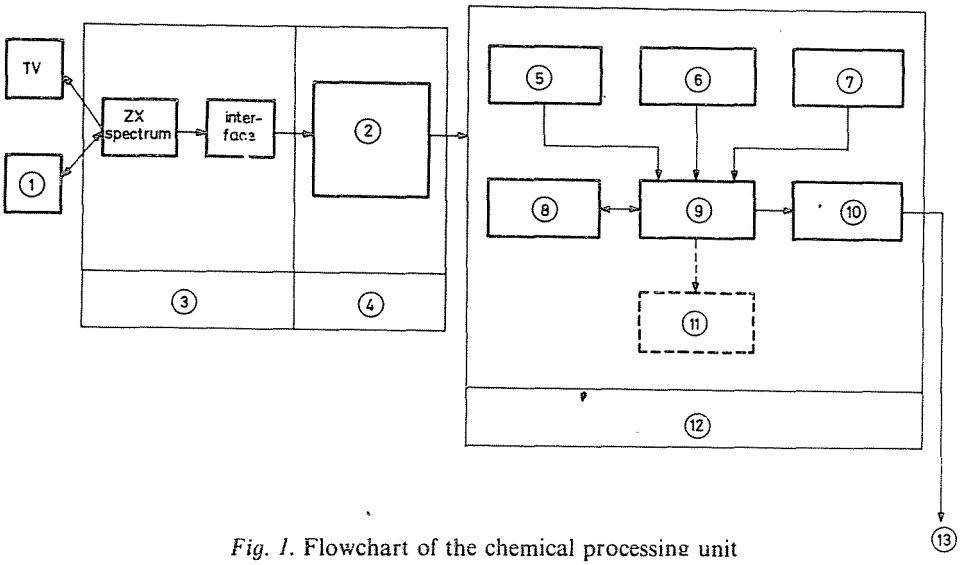

Fig. 1. Flowchart of the chemical processing unit
1. Magnetic tape recorder; 2. Electric electropneumatic elements; 3. Programmer unit; 4. Control unit; 5. sampler/feeder; 6. Reagent storage/feeder; 7. elution headpiece; 8. decomposition/distillation unit; 9. liquid moving/transport; 10. liquid chromatographic unit; 11. optional extension; 12. chemical processing unit; 13. Measuring instrument or fraction receiver

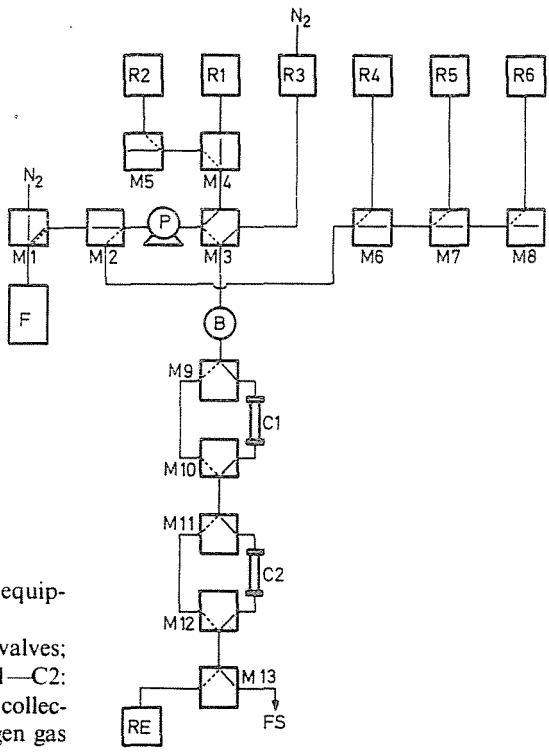

Fig. 2. Block diagram of the processing equipment.
R1—R6: reservoirs; M1—M13: microvalves; P: pump; F: flask for destruction; C1—C2: chromatographic columns; FS: fraction collector; RE: waste; B: debubbler; N_2: nitrogen gas

flowchart of the chemical processing unit, are determined by the position of the microvalves operated according to the time program, thus opening the pathway of the fluids flowing in the closed system into a given direction for a given time interval. The liquids are transferred by overpressure and/or with a peristaltic pump. At the output of the processing unit a sample suitably prepared for nuclear measurement appears.

Chemical processing unit

To meet practical requirements, low and high-pressure liquid chromatographic systems [1], continuous extraction and distillation equipments have been developed and applied in a wide field. These systems are suitable to solve special tasks, but they are not able to meet the requirement often arising in practice that the single operations and their order should be variable. Moreover, processing radiochemical samples of high activity, problems in conjunction with radiation protection and decontamination arise generally.

To solve the problem a processing unit meeting the requirements arising in radiochemistry was developed partly from commercially available liquid chromatographic elements and partly from elements developed at our Department [2, 3]. Owing to the modul system, a unit fitted to the given special task can be assembled using the elements needed. However, not only optimal instruments for special purposes can be constructed from the elements, but simple and easily variable, so called development system can be assembled, too.

At present the chemical processing unit makes possible the realization of the optional combination of the following operations:

— wet destruction of solid samples;
— distillation, refluxing;
— low-pressure liquid chromatographic separations;
— feeding of reagents, transporting, fractionation of liquids.

Moreover, the system can be complemented by a continuous extraction unit.

Elements developed at the Department

Microvalves

In the first specimens of the equipment modified variants of the three- and four-way sliding valves of the "Cheminert" Company were used. However, their relatively complicated construction and certain packing difficulties induced us to develop new valves, based on rotation principle. Their operation is similar to that of glass taps. In the stator the tapered cylindrical rotor with

bores can be set in several positions, thus connecting the inlet bores of the stator in different ways. So far, two-position, maximum six-way microvalves have been made, at present multi-position (16-way) units, which can be used as reagent feeding or fraction-collecting valves are under development.

Parts in contact with flowing liquid are made of PVC or teflon, but operations using organic solvents can be carried out using KEL–F as construction material. Thus, microvalves which are resistant to chemicals and corrosion, can be easily cleaned or decontamined. The dead volume of the valves is less than 0.05 cm^3, and switching time is less than 1 second.

Testing the life of the rotary valves no changes were observed up to 20 000 switchings. Thus, rotary valves — according to the most important parameters — can be compared with international products.

Debubblers

In chromatographic technique the removal of gas bubbles from the liquid generally needs attention, because bubbles getting into the column lead to so called aerifying. Debubblers are intended to prevent this phenomenon.

In our case the debubbler is a through-flow cell of a volume close to 2 cm^3, from which the release of accumulated gases is controlled by the decrease of the liquid level in the cell. The change in liquid level (in the case of any electrically conductive liquid) is detected by three electrodes arranged at different heights, connected according to Wien-bridge principle. If the quantity of gas above the liquid in the closed dome of the cell increases due to bubbles liquid level decreases. When the level sinks to the given minimum, the control unit closes the pathway of the liquid at the outflow of the cell, and by the simultaneous opening of a blow-off valve gases are released into the atmosphere. The pathway of the gas into the atmosphere is open, until the liquid level attains the similarly fixed maximum value. Following this, the valve in the liquid pathway opens again, so that liquid free of bubbles leaves the cell.

Destruction — distillation flask

An air-heated destruction flask was constructed for the dissolution, destructive decomposition of solid samples, and for the distillation of liquids. The flask permits high heating rates without local overheating, and provides also for high cooling rate. The flask is equipped with spray-catcher, condenser, pump connection and rectification head piece. The reaction mixture can be stirred either introducing gaseous nitrogen, or with a magnetic stirrer.

Columns

Columns used for separation are made of standard polyethylene tube. The packing is seated on a quartz-wool bed. The material of the column heads is PVC (or KEL–F), inlets can be connected with threads. The columns can be thermostated. Sorption, ion-exchange or partition chromatographic separations can be realized using the columns.

Control unit

The control unit is essentially an electric — pneumatic converter, which transports the control gas to the pneumatic element of the respective microvalve, according to the program instruction when computerized control program is applied and to pressing the respective push-botton when the system is manually remote controlled.

Main characteristics of the version constructed

— simultaneous independent pneumatic control of 12 microvalves;
— 6 controlled, low-pressure (0–1.6 bar) operational gas outputs (which can be used e.g. for the pneumatic moving, mixing or rinsing of reagents);
— 4×220 V 50 Hz output (e.g. for the control of pump or magnetic agitator).

Programmer unit

Its task is the sending of control instructions to the electro-pneumatic converter unit, according to the program fixed in the magnetic storage. There is possibility for manual intervention, and for the display of pictures (flowcharts) and texts. Its main parts are: ZX Spectrum personal computer and its peripherals (magnetic tape unit, TV display), further the interface unit. The personal computer was selected of the types available in Hungary on the basis of the following considerations:

— the technical literature of the Z 80 microprocessor used in the computer, is relatively easily accessible;
— in Hungary it is one of the most widely used type of computers; it provides for the services necessary (and sufficient) for us for the lowest price.

The computer can be directly programmed in BASIC and Z80 ASSEMBLER languages, and with the aid of appropriate subprograms the faster and more efficient high-level languages FORTH or PASCAL can also be used. The peripheral units of the computer are: commercially available colour, or black- and white TV set and magnetic tape recorder.

Our present control program was written in BASIC language, while data transmission is provided by ASSEMBLER subroutines. "Switching" programs suitable for the realization of certain operational order can be written, tested and run within the control program. In all the three above cases the respective flowchart is displayed on the TV screen. So that the position of the microvalves, the state of the individual operational elements and the most important parameters can be readily followed. Thus, the system can be easily handled without programming knowledge.

In its present state the switching program enables time-programmed operation of up to 24 channels. The minimum time between two program steps is 1 second, the maximum is 18 hours. The running time of the switching program has no upper limit. Time is measured by a software cycle, its error does not exceed 0.1%. The control program can be used also for the design of the optimal switching scheme.

The construction of the interface, realizing parallel transmission, provides data transfer in two directions.

The interface makes the central processor galvanically independent of the peripheral device with the aid of optoconnecting disconnection to promote noise protection. Parallel data transfer in both directions is provided by Z80 PIO circuit for the CPU. An interrupt handler routine receives information in data input. Data output is realized in two steps via a bufferstorage register and a driver storage register. Data can reach the buffer register at any time, the only constraint is that the actual datum (bit combination) be present at the time of the write-in cycle of the driver register. The controlled outputs of the interface can control storage within the following levels: $+5 - +12$ V=; 1–200 mA so control of both TTL and relay circuits are enabled.

Using the elements and units described, in addition to suitable sensors and analog — digital converters, not only time program control, similar to the present version, but also a mode of operation with completely automatic control, meeting further demands, can be realized.

Example of application

By way of example we present a characteristic version of the processing unit, suitable for the wet decomposition and separation of neutron-activated samples by low-pressure liquid chromatography. Fig. 3 illustrates the block-diagram of the processing unit.

The sample contained in decomposition vessel F is decomposed with the reagent(s) transported by peristaltic pump P from reagent feeder(s) R1 or R2 through microvalves M5, M4, M3, M2 and M1. The reaction mixture is stirred by the introduction of low-pressure nitrogen. At the same time reagent is

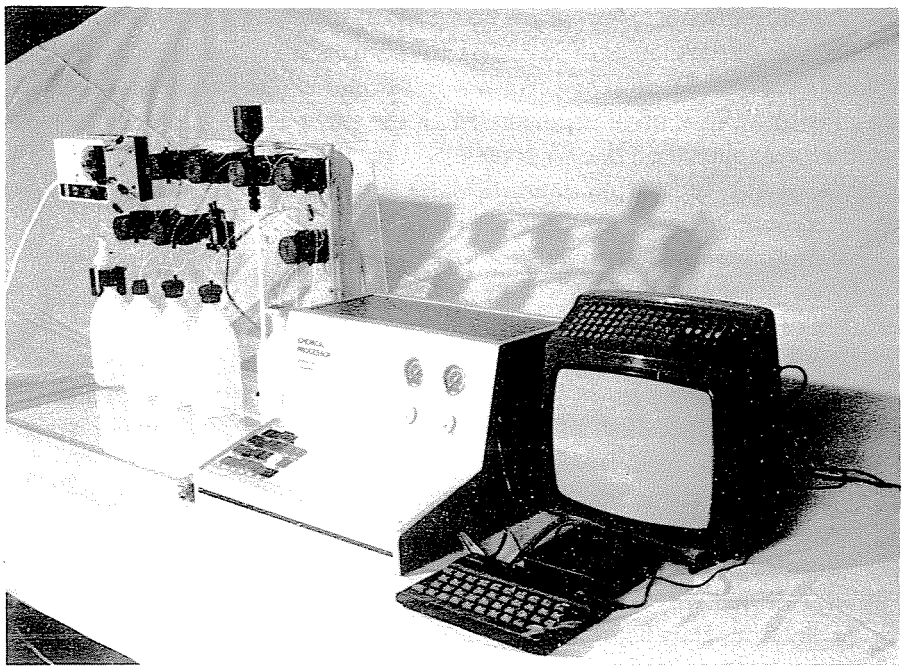

Fig. 3. Chemical processor

passed through columns C1 and C2 with low-pressure nitrogen from reagent container R3 via the microvalves M3, M9, M10, M11, M12 and M13 and the debubbler B carrying out the pretreatment of column packings. Excess reagent is collected in the vessel RE for regeneration. When decomposition is ended, pump P pumps the solution from decomposition vessel F through microvalves M1, M2, M3, M9 and debubbler B into the chromatographic column C1 and/or through valves M10, M11, M12 and M13 into column C2. Chromatographic separation can be performed with the eluents contained in containers R4, R5 and R6, the eluents being pumped with constant rate by peristaltic pump P through microvalves M6, M7, M8, M2, M3, M9, M10, M11, M12 and M13, debubbler B and columns C1, C2 to the fraction receiver FS.

In chromatographic separations the first column C1, packed with ion exchange resin, is suitable for the removal of colloidal impurities, and improves the efficiency of the principal ion-exchange column, C2.

In continuous chromatographic separation, reagent storage vessel R4, R5 or R6 can be replaced for a gradient elution headpiece.

In summary — as can be seen also from the description of the chemical processing unit, of the elements developed at the Department and of the application example — it can be stated that the automated processing system developed by us can be widely used in radiochemistry and other fields.

References

1. SNYDER, L. R.–KIRKLAND, J. J.: Bevezetés az intenziv folyadékkromatográfiába (Introduction to intensive liquid chromatography). Műszaki Könyvkiadó. Budapest 1979.
2. TÖRÖK, G.: Programozott roncsolásos neutronaktivációs nyomelemanalízis metodika, berendezés kidolgozása és alkalmazása (Programmed destructive neutron activation trace element analysis method, development and application of equipment). Dissertation for the scientific degree of a C. Sc. Budapest 1977.
3. TÖRÖK, G. et al.: Remote-controlled automatic chemical processing equipment of modul system. Hung. patent application No. 1731/83.

Dr. Gábor TÖRÖK
Prof. Dr. Lajos György NAGY
János RUIP
Miklós VODICSKA
Nóra VAJDA
Dr. József SOLYMOSI

H-1521 Budapest