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System of Thermostatic Control on Peltier Thermopiles and Microprocessor Control for a Portable NMR Relaxometer

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Liquid equipment for thermostatic control of samples with accuracy better than 0.1°C within the temperature range from −20 to +70°C is designed for small-sized NMR spectrometers. The difficulty in using this equipment is a thermomodule based on the Peltier effect. The temperature control system is based on a single crystal microcomputer. The equipment fits modern small-sized NMR spectrometers, which facilitates medical and biological studies. © 2002 Elsevier Science (USA)

The construction of portable NMR devices gave a powerful impetus to the interest in systems with thermostatic control of ampoules with samples in the probehead of an NMR relaxometer. The creation of such devices was possible due to the production of portable magnets on the basis of intermetallic materials with high specific magnetic energy (*1*). Portable and comparably cheap NMR relaxometers are ideal for medical–biological and ecological investigations but using them with traditional bulky systems of thermostatic control decreases the advantages of portability. The main problem of thermostatic control lies in obtaining negative Centigrade temperatures. Traditional means are (i) gaseous thermostatic control with vapor of liquid nitrogen (*2*); (ii) liquid thermostatic control with freon condensers. Both systems used in portable NMR relaxometers conventionally meet difficulties due to a small size of the NMR probehead in addition to general size incompatibility.

A liquid system with modern freon condensers, as compared to a gaseous system, has better size parameters but poorer thermostatic control range. However, in most experiments, in particular in medical–biological experiments, a comparably narrow temperature range of about −20 to +70°C is necessary. In this temperature range, liquid thermostatic control with sufficiently effective freon condensers is preferable. However, the conventional liquid system, as in the gaseous system, is not adequate for portable NMR relaxometers, considering its size.

This paper describes a thermostatic control device monitored with a single-crystal micro PC which is adequate in size for portable NMR relaxometers. Ampoules with samples are thermostatically controlled in a probehead of a portable NMR relaxometer. This device is convenient for use in medical–biological investigations. Usage of Peltier thermopiles (*3, 4*) allowed us to solve the problem of size matching of the system of thermostatic control with the portable NMR relaxometer. Peltier thermopiles are widely used in various fields of science and engineering. So far, Peltier thermopiles were not used for thermostatic control of ampoules with samples in a probehead of a portable NMR relaxometer. Therefore, the development and description of such a device seems to be useful since the efficiency of its operation depends significantly on the specific engineering solution.

The thermostatic control device consists of three parts:

1. a thermal unit;
2. a thermal jacket;
3. an electronic system for the regulation and maintenance of a given temperature.

The thermal unit (Fig. 1) serves for heating–cooling of a cooling agent up (down) to a given temperature, for maintenance of this temperature with a given accuracy, and for pumping the cooling agent in the thermal jacket. Cold sides of Peltier thermopiles (1) are in a heat contact with the side faces of the heat exchanger of the cooling agent (2). Outer hot sides of thermopiles are cooled with water heat exchangers (3). A pump (4) and an expanding chamber (5) with an inside heater coil (6) are located in the cylindrical cavity of the heat exchanger (2). The cooling agent is pumped through a spiral channel in the volume of the heat exchanger. The thermal unit is coupled to the electronic unit for control and maintenance of a given temperature.

The thermal jacket (Fig. 2) envelopes a two-way contour of the NMR probehead and is constructed as a cylinder formed by two coaxial copper tubes (1) soldered at their ends.

To decrease temperature gradients, the ring gap between cylinders along the longitudinal axis is divided by membranes (2) into a series of communicating segments. In the upper part of the thermal jacket, receptacles (3) are soldered for the control (4) and monitoring (5) of the temperature. When the pump of the thermal unit functions, the cooling agent moving up and down along the jacket passes the segments of the jacket and washes the receptacles for the temperature probeheads.

This construction is easy to manufacture, provides effective heat transfer, permits the use of liquids with resonance nuclei as

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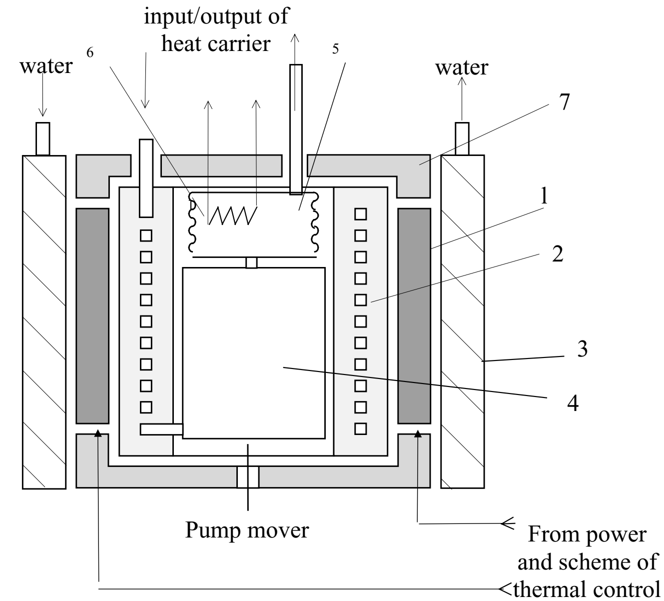

FIG. 1. The construction of the thermal unit: (1) Peltier thermal moduli; (2) heat exchanger for heat carrier; (3) water heat exchanger; (4) membrane pump; (5) dilating chamber; (6) heating spiral; (7) heat insulator.

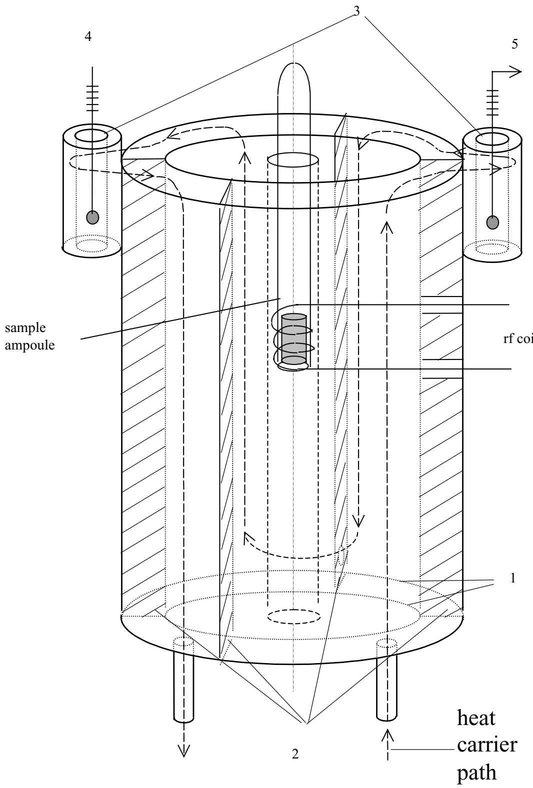

FIG. 2. The construction of the thermal jacket: (1) coaxial tubes of the thermal jacket cylinder; (2) barriers between coaxial tubes; (3) sockets made of coaxial tubes for installation of and controller; (4) temperature monitor; (5) temperature controller.

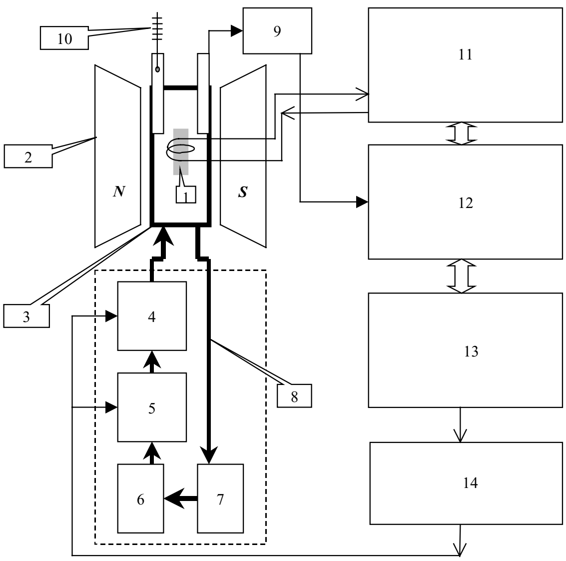

FIG. 3. Block scheme of the NMR-spectrometer with the thermal control system: (1) ampoule with the studied sample; (2) magnet of the NMR-spectrometer; (3) thermal jacket; (4) heater; (5) Peltier thermal moduli; (6) pump; (7) compensator of the temperature changes of the volume heat carrier; (8) connecting pipes; (9) thermal convertor; (10) temperature control monitor; (11) electron units of the NMR-spectrometer; (12) PC; (13) scheme of temperature control; (14) controlled power supply for the heater and Peltier thermal moduli.

cooling agents, and provides screening of the two-way contour from the gradient coils, although it somewhat lowers the quality factor of the two-way NMR contour.

The jacket is thermally insulated from the poles of the relaxometer magnet by an active thermostatic bypass. The outer elements of the probehead surface are arbitrarily heated up to the temperature of thermal control of the magnet and the thermal jacket is covered with a layer of a substance having low thermal conductivity (carbon fiber).

The block scheme of the connection of the thermal jacket and thermal unit with the temperature control electron scheme and NMR-spectrometer is shown in Fig. 3.

Temperature is regulated and controlled with a programmable controller of a digital power regulator constructed on the basis of a single-crystal microcontroller (*6*, *7*), see Fig. 4. Usage of a micro PC enables one to: exploit various algorithms of temperature maintenance, including a regime of fast approach to a given temperature and high-accuracy maintenance; combine functions of a heating regulator and temperature measuring in one unit, providing a wide range of characteristics of the heater power; increase the functionality of a system by switching additional thermal probeheads, devices for state indication, etc. by resetting programs and tables in a programmable ROM without significantly changing the basic electrical scheme. Communication means (a port of asynchronous consequent data transfer) installed in a given microcontroller allows the switching of a thermostatic control unit to any outer regulating PC equipped with ports RS-232.

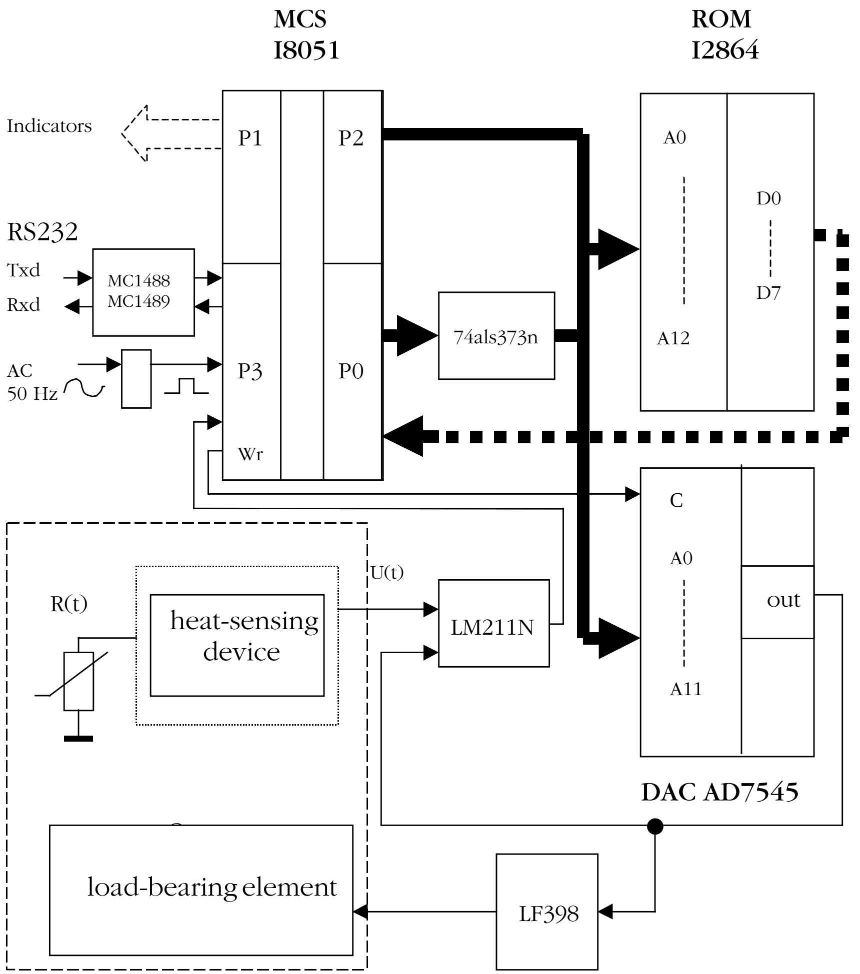

FIG. 4. Block scheme of the electronic temperature control system.

Current temperature in a thermostat is measured with a microcontroller performing an algorithm of a step-by-step balancing of a DAC signal and the output potential of a temperature transformer. The same DAC is also used to set the power transmitted into the system by heater elements. When DAC is switched into the regime of measuring temperature, the signal determining the power level of the heater is stored by the access-storage device (ASD or T/H).

The main elements of the unit regulating the thermostatic control system are a microcontroller (series MCS-51) and a 12-step digital–analog transformer (AD7545). The serving program of the thermal stabilization system is installed in the outer ROM2864. A microcontroller is synchronized by an internal generator with quartz stabilization at the frequency 12 MHz.

Bus former 74ALS373N and ROM2864 are switched according to a standard scheme and allow the microcontroller to work with the outer program memory.

To reduce the number of outer elements, the DAC registers–triggers are switched into the address space of the outer RAM of microcontroller data, and recording is performed by standard commands of data output using the signal of data quitting into the outer RAM data. An operational amplifier LF355 is used as a transformer of current-voltage of the output DAC signal. The signal determining the power level of the heater is stored in T/H constructed with a specialized integral micro scheme LF398. To eliminate the “glitch” effect of DAC this storage is performed according to the signal formed at the output of P1.0 of the microcontroller and during switching of DAC into the regime of temperature measurement.

Integral analog comparators LM211N compare the DAC signal with the output level of the temperature probehead when the microcontroller performs an algorithm of a step-by-step balancing to obtain a digital equivalent of the voltage at the thermal transformer output.

The buffer elements MC1488 and MC1489 match the electrical parameters of the communication serial port microcontroller (TTL levels) to the requirements of the RS-232C standard in a duplex regime (*5*).

SOFTWARE

The software of a controller is a background-operation system composed of three main tasks:

1. temperature measurement;
2. setting of the heater power;
3. indication of states functioning in a cyclic algorithm and data exchange with an upper-level PC working on interruptions from the communication port. For the controller to function in the autonomous regime, i.e., without the controlling upper-level PC, the program software is supplemented by sampling of the clipboard matrix switched into the free region of the outer RAM data or by an outer clipboard working via a serial port (*8*).

TEMPERATURE MEASUREMENT

In measuring potentials from the outer probeheads, the microcontroller functions according to the algorithm of step-by-step balancing, substituting the register of consequent approximations. Voltage signals at the DAC output and those at the output of the temperature probehead are compared by an analog comparator and the result of the comparison is put into P3.3 of the microcontroller. Thus, the microcontroller and the DAC form a 12-step ADC with the transformation time of 250 μs. The transformation time is determined by the time of the establishment of the DAC signal (15 mks for AD7545 used) and may be reduced if T/H with corresponding characteristics is used.

POWER CONTROL

In controlling the heater power, the microcontroller calculates the current coefficient of the power level and puts it into the DAC registers. It is stored with T/H by the recording signal formed at the output of P1.0. The controlling law was the proportional–integral–differential law given by

$$Z(t) = K_n \left[X(t) + \frac{1}{T_i} \int_0^t X(t)\,dt + T_d \frac{dX(t)}{dt} \right],$$

where K_n is the proportionality coefficient, T_i is the integration time constant, T_d is the differentiation time constant, $Z(t)$ is the measured current parameter, and $X(t)$ is the power delivered to the Peltier thermopiles and heater. The dynamical tuning of the regulator, i.e., the choice of the coefficients of the controlling law, is carried out according to (*9*). The heater current is provided either by the transistor power amplifier or by the thyristor regulation scheme. When the heater element's control is carried out by the thyristors, it is possible to use both phase-pulse and pulse-duration modulation with an inner "microcontroller" timer to set the angle of switching on the thyristors or the on–off time ratio. Synchronization with the moment of transfer of the voltage through zero is performed according to the signal at the input P3.4 to reduce the noise of switching.

STATE INDICATION

State indication is switched to port P1 and is carried out by a 10-step semiconductor digital indicator. The task of the indicator service is cyclic, organizing the output of information in a dynamical regime and presents the current heater power, temperature measured, and microcontroller state.

COUPLING WITH THE UPPER-LEVEL PC

Coupling with the upper-level PC is arranged via a channel of serial data transfer in a duplex regime with a speed of 2400 b per second on the protocol Xon/Xoff. A consequent port microcontroller functions in the regime 1. The errand format −10 b: start-bit, eight bit of data, stop-bit. The timer-counter is initialized in the regime 1 with auto-load. Data exchange is activated by the interruption signal when getting data from the upper-level PC and provides receipt of the values of the temperature set or transfer of the heater to the fixed value of the power level and transmission of the values of the current or set temperatures and powers to the upper-level PC.

SOFTWARE OF THE UPPER-LEVEL PC

Software for controlling the thermostatic system is installed as one of the tasks of providing functioning of an NMR relaxometer–diffusometer and provides manual setting of the thermal stabilization with visual presentation of the current and set parameters on the PC screen using the operation system Windows 98 (*10*).

An experimental setup was constructed on four 2-cascade thermal Peltier modules of the 2TM-127-63-6 OM type with dimensions 40 $\times$ 40 mm, heat output $Q = 35$ W at the maximal difference between the hot and cold sides of 86°C. The thermostatic volume of the NMR probehead is 24 cm^3. The volume of the cooling agent (ethanol) is 150 cm^3. The maximal heater power is 200 W. The cooling/heating mode of the temperature maintenance was provided by commutation of polarity and control of the intensity of the current through the Peltier thermal batteries and through the heater. For a given cooling agent, at the temperature of cooling water of 10°C and flow of 3l/min the thermostatic range started at -27°C. The upper boundary of $+80^\circ$C was limited by the boiling temperature of the cooling agent. The guaranteed temperature range is from -20°C up to 70°C, the accuracy of temperature maintenance is better than 0.1°C. Time of setting the low-temperature point of the range at the power of 300 W was 15 min. Due to air-tightness of the thermostatic device it is possible to use various cooling agents with an appropriate temperature range of the liquid state including toxic ones, e.g., tetrachlorethylene, CCl4.

This device is the appropriate size for modern portable NMR relaxometers–diffusometers, it is noiseless, makes NMR experiments cheaper, increases their productivity, and it may function with an accumulator.

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