Automated Control System for Radiation-Chemical Process of Composite Materials Formation*

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Abstract

A real-time control system is designed for radiation-chemical process of polymer composite materials (PCM) formation. PCM are manufactured by the bulk impregnation of solid capillary-porous materials, such as wood, gypsum, ceramics, concrete, tuff by monomers or oligomers, and subsequent polymerization under effect of bremsstrahlung or scanning electron accelerator beam with energy of 5-10 MeV, time duration of 4 mcsec., and mean beam power of 5 kW. The experimental results, which demonstrate the control system′s utilization for the production of various PCM with extended strength properties and corrosion resistance, are discussed.

1 Introduction

At Kharkov State University the radiation-chemical process of polymer composite materials formation was developed [1-3]. The general technological scheme of PCM production consists of impregnation by synthetic monomer or oligomer of articles made of capillary-porous materials such as wood, gypsum, concrete, ceramic, paper, as well as pressed granular waste of papermaking, textile and wood-working production. These are further treated by relativistic electron or bremsstruhlung beams. Such radiation treatment yields new PCMs in which the capillary-porous structure acting as reinforcement is filled with polymer. New compo-site materials (wood-plastic, gypsum polymer, organic concrete) obtained by the radiation-chemical process have qualities such as high strength characteristics, low water and moisture absorption, high stability of form and size, chemical and bio-resistance; resistance to attack of atmospheric factors, corrosive media, abrasion loads.

The comprehensive investigation of the kinetics and thermodynamics of the radiation-stimulated polymerization of monomers/oligomers in heterogeneous media was performed. It was found experimentally that the following parameters have an effect on the end composite material properties: characteristics of the radiation (type of particles, their energy, dose rate and integral dose); relative concentrations of filler and saturant; characteristics of the filler (strengths and elasticity, specific pore volume, configuration and size of pores); characteristics of applied chemicals (monomers, oligomers, solvents, initiators, sensitizes, catalysts); some external conditions (temperature, pressure). The authors developed a control processing system for optimizing the radiation treatment of compound (monomer/ oligomer capillary-porous material) by electron/ bremsstrahlung radiation.

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2 Radiation facilities for PCM formation

Let′s consider a process of fabrication for polymer modified wood as an example of the use of radiation - chemical treatment of capillary-porous materials. In our opinion, it is the favorite PCM, which has vast application. We can, through programming, modify its operational characteristics, as well as physical-mechanical properties. As a starting material for polymer modified wood, one can use soft low-grade wood poplar, aspen, alder, birch. After radiation - chemical treatment this wood acquires better physical-mechanical characteristics than those of the hardest and most expensive sorts of wood, namely, oak, beech, hornbeam.

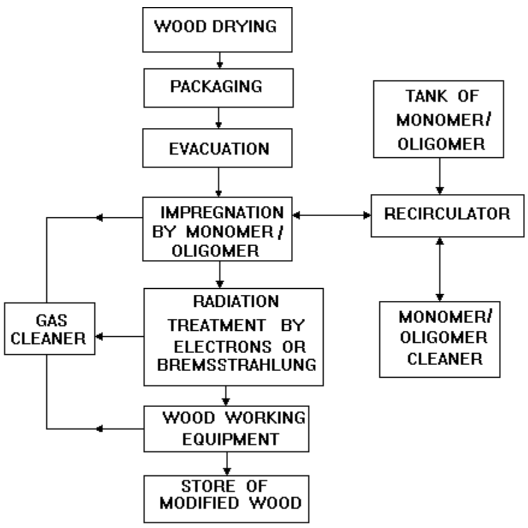

Fig. 1 Technological scheme of polymer-modified wood production.

The technological scheme of polymer-modified wood production is shown in Fig.1. The radiation facilities for polymer composite material formation incorporates a linac with scanning electron beams, microwave chamber for the drying of the materials, an evacuation system, a system for the impregnation of materials with synthetic monomers/ oligomers, a test bench for irradiation of samples, equipment for monomer/oligomer and gas cleaning, and equipment for working the PCM (cutting, grinding, and polishing). The drying of raw materials was executed in a vacuum or/and in a microwave field.

The stage of radiation treatment of the compound is the most complex. At the same time it determines the quality of the produced PCM and the productivity of the radiation-chemical installation. The main problems of this stage are the formation of a uniform three-dimensional irradiation field, and an optimal regime for the maintenance of the electron or bremsstruhlung beam. The latter varies according to prescribed low during the radiation treatment of compound. The required time, *t*, for performance of the radiation-chemical polymerization process is between 5 and 30 min. depending on the compound composition.

The experimental radiation-chemical installation, as shown in Fig.2, consists of an electron accelerator with electromagnetic scanner, an electron beam monitor, a closed container with the modified materials, and the process control system. As a source of the scanning electron beams, a linear accelerator of the type "Electronica - U003" is used with following characteristics: electron energy 5-8 MeV; electron beam current 0.5 mA; mean beam power 5 kW; pulse duration 1-4 mcs; pulse frequency 1-250 Hz; scanning frequency of electromagnetic scanner 1-8 Hz; the deviation angle of scanning electron beam was within $\pm 20^o$; the irradiation is possible both with electron and bremsstrahlung beams. The bremsstrahlung radiation was generated by an electron beam in a tantalum converter which was placed in front of the container.

3 Structure of the process control system

The process control system incorporates a set of acoustic (AS) and temperature (TS) sensors, a CAMAC crate (CC), a subsystem for the control of the radiation-chemical process (CSR-CP), a container displacement subsystem (CDS); a beam monitoring system (BMS), an electron accelerator control subsystem (ACS), a scanner control subsystem (SCS), a synchronization module (SM), and an IBM-compatible PC with corresponding software.

The accelerator control system ACS is connected to the accelerator power source APS, which consists of a high-voltage source, a modulator, a magnetron, a phase shifter, an electron injector and other elements.

The (CSR-CP) system includes AS3, TS1 and TS2 sensors, pre PA and main MA amplifiers, a DCA**,** a fast FADC - and slow SADC analog-to-digital converters. Signals from the sensors are transmitted in analog form by 100m coaxial cables to the FADC, SADC where analog-to-

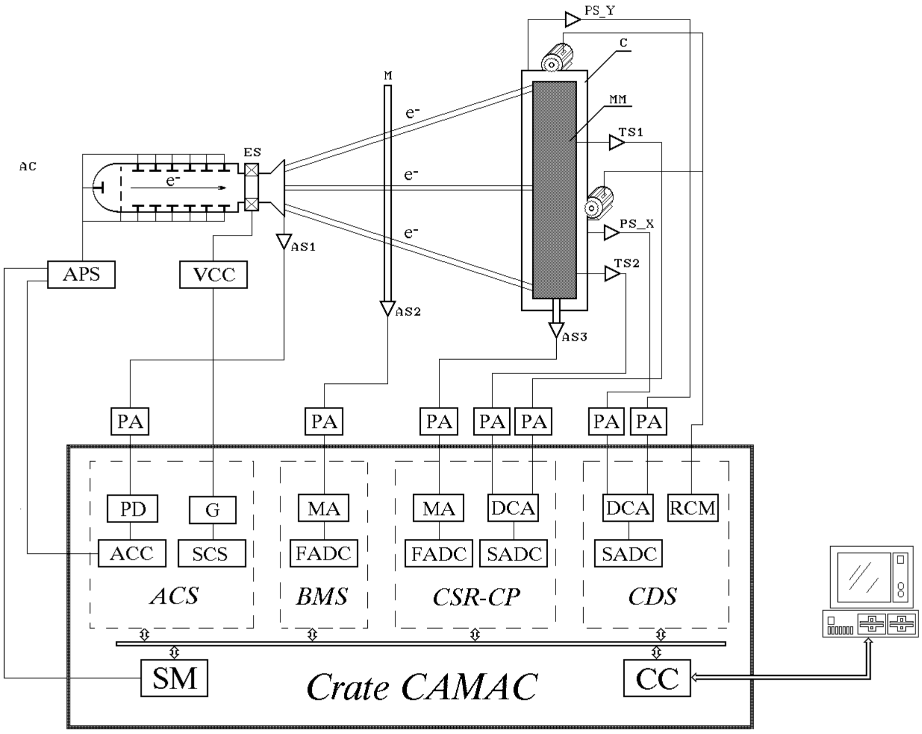

Fig. 2 Block diagram of the radiation facilities for composite materials formation.

e^- - pulsed beam of electrons; ES - electromagnetic scanner; M - scanning electron beam monitor; MM - sample of modified material; C - closed container; AS1, AS2, AS3 - acoustic sensors; TS1, TS2 - temperature sensors; PA - preamplifier; MA - main amplifier; DCA - direct-current amplifier; FADC, SADC - fast and slow analog-to digital converter; SM - synchronization module; ACS - accelerator control subsystem; APS - accelerator power source; ACC - accelerator control console; SCS - scanner control subsystem; VCC - voltage-to-current converter; G - pulsed generator; BMS - beam monitor system; CDS - container displacement subsystem; CSR-CP - control subsystem for radiation-chemical process; PD - peak detector; RCM - relay control modules; PS-X, PS-Y - position sensors; CC - CAMAC crate controller.

digital conversion is performed. The system uses both the acoustic emission and temperature distribution created by ionizing radiation in a sample MM, as a source of primary information concerning the current status of the monomer/ oligomer phase state in the PCM.

The monitor M includes thin rods as its sensitive elements with a piezoelectric detector AS2 on its butt-end, an electronic preamplifier PA, an amplifier MA, and a beam monitor system BMS.

Remote movement of the container C with modify material samples MM, is carried out with RCM by servoactuator, with controlled displacements along axes X and Y in plane (X, Y), which is perpendicular to beam axis.

Acoustic sensor AS1, PA and PD serve for emergency protection of the accelerator beam-transport pipe against damage from a high-current beam. When the acoustic signal amplitude exceeds a preset limit, the accelerator injector is disabled.

The acoustic waves are detected by broadband piezocera-mic ($\Delta f \leq 10$ MHz) sensors AS1-AS3 with sensitivity of 10 V/Pa, placed on a sample of modified material MM, on the beam monitor M, and on the accelerator elements, e.g., on foil of the accelerator outlet window. These sensors have high sensitivity and radiation resistance and their construc-tion provides protection against electro-magnetic inter-ference.

Analog-to-digital conversion is realized by fast 8-bit digital quantizer TDA8703 (Philips Semiconductor) having a maximum sampling frequency of 40 MHz. The frequency band of the restored signal on digital quantizer input is 0 to 5 MHz.

Chromel- capel thermocouples having a thermopower of 0.064 mV/ ^{0}C and nonlinearity of 8% in the temperature range of 0 to 200 ^{0}C are used as temperature sensors TS1 and TS2 placed on the sample surface. Analog-digital signal conversion is realized by 8-bit digital quantizer ADC0808CCN (National Semiconductor) with integrated 8-channel multiplexer. The conversion range is 0 to 10 V with a quantization step of 40 mV. Calibration curves of the sensors being used are stored in the computer memory and are employed for shape reconstruction of the sensor signals.

4 Control system operation

In order to obtaining high quality PCM by radiation-chemical polymerization one should maintain the compound temperature T_c in the range of 80 to 100 ^{0}C. At continuous electron beam intensity $I_e(t)$ = const temperature T_c can increase up to 200 ^{0}C due to process exothermicity, which causes PCM destruction. But we found optimum regimes of intensity variation $I_e(t)_{opt}$ when T_c does not exceed of 100^{0}C

We designed physical methods for the control and diagnostics of the kinetics of the radiation - chemical polymerization of compound. These real-time methods provide simultaneous registering the following parameters: the heat release during exothermic reaction of radiation-chemical polymerization of compound that leads to increasing its temperature T; the amplitude of the acoustic signal V generated by radiation in the sample of modified materials; dose D and dose rate of radiation.

When a pulsed electron beam passes through the sample of modified material and the accelerator constructive elements, rapid heating and thermal expanding of their materials takes place. Herewith generated acoustic waves carry information concerning beam parameters and interac-tion spot location [4, 5]. The amplitude V, time and frequency characteristics of these waves also carry information about the kinetics of the phase transition (monomer/oligomer –polymer), namely, about the stage, the phase location and the concentration, as well as thermo-physical and thermoelastic characteristics of composites [6,7].

The operation of the process control system based on the phase state monitoring of processed material MM resulting from the measurement of the sample temperature T and the acoustic pulse amplitude V on dose D, intensity j_e and time t of irradiation. These data permit controlling of the techno-logical installation parameters to support the optimal regime of the radiation treatment by the ACS, BMS, and SCS subsystems. The optimum values of the measured temperature $T_{opt}(D,t)$ and of the acoustic signal amplitude $V_{opt}(D,t)$ for various monomer/oligomer–capillary–porous material systems and regimes of bremsstrahlung or electron irradiation are stored in the memory of the control computer. They can be corrected in the course of data storing, including the automatic regime. In the case of departure of the measured parameters T(D,t) and V(D,t) of optimum values $T_{opt}(D,t)$ and $V_{opt}(D,t)$, the ACS, BMS, and SCS subsystems change scanning beam parameters to maintain these values.

5 Beam monitor system

Thermoacoustic one dimensional monitors M on the base of thin rods and plates were used in the control system of the scanning electron and bremsstrahlung pulsed beams in the technological process. The monitors are characterized by high informativity, simplicity and reliability of construc-tion. The monitor provided continuous, nondistorting acoustic dosimetry of the extensive area under pulsed radiation.

The monitor with ramifying body was displaced normally to initial direction of electron beam axis. Its body consisted of one or a few parallel titanium wires 140 cm long. The wire diameter d, as well as its material, is chosen from conditions $d << \min\{D, E_e/\chi\}$, to provide nondistorting dosimetry. Here E_e is electron energy, χ- specific energy loss of beam electron.

When a sequence of electron pulses passes through the rectilinear body of the dosimeter, a sequence of thermoacoustic waves appears in it. In the approximation of instantaneous heating $\tau_b << D/s$ the stress wave amplitude $V(t)$ generated in dosimeter body by either electron pulse, is proportional to the spatial distribution of deposited energy. The sequence of thermoelastic waves goes to a wide-range piezoelectric detector, which transforms these into electric signals and is displayed by a register device, operating in accumulation mode.

Each of the acoustic pulses, generated in the dosimeter body by the accelerator pulse sequence, carried the information about the location and the transverse distribution of the corresponding electron pulse. The registration and the processing of the output signal was executed by the BMS. As a result, a spatial profile of a radiation field j_e, caused by any periodic shape of current in the scanning magnet ES, was displayed immediately (see Fig. 3). It permitted fitting the profile j_e, to a desired shape by varying the shape of magnet current with help of a voltage-to-current convertor VCC, a pulsed generator G, and the scanner control subsystem SCS.

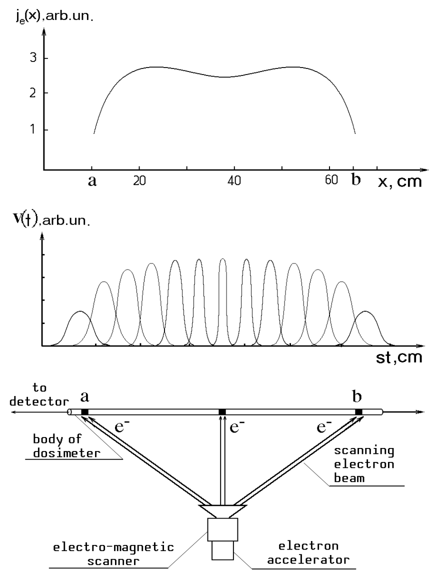

Figure 3. Scheme of utilization of a wire acoustic dosimeter for the monitoring of a scanning electron beam. The current in the scanning magnet had sinusoidal form.

6 Software

The software of the control system fulfills the following functions: delivers information concerning the initial regimes of beam parameters for any composition and dimensions of processed compound to the operator; displays information concerning the radiation parameters and calculates a dose profile in the modified material; determines the current status of the manufactured material; maintains the optimal regime of the radiation treatment; determines the termination time of polymerization process; realizes the remote manoeuvring of the modified samples; leads the experiment record; stores statistical information.

Besides, the application package performs: the control of the CAMAC crate moduli, and the graphic display of recorded data on the monitor screen, including the broadband thermoacoustic signals from the radiation-acoustic probes, and the slowly-changing signals from the temperature probes are displayed; the graphic display of the calculated values and the curves; the control and operation of the processing parameters, namely, the technological container coordinates; the registration of processing para-meters, as well as, of control-administer information; the submission of technological and reference information, the formation and control of the processing data base. The language C was used for the development of this software.

7 Radiation treatment of compounds

The large-sized samples of modified materials MM (nearly 100x50 cm) were treated by the scanning electron/bremsstrahlung beams in the radiation facilities (see Fig.2). The samples, more than 100 cm, and were treated by shifting step by step transversely to the electron beam. The uniform radiation flux distribution on the large area (nearly 100 x 40 cm) of the PCM samples was created by the scanning system with a special form of current-history in a scanning magnet.

Two-sided irradiation of the thick PCM samples or sample stacks, whose thickness is more than the electron range, was used to increase the beam utilization coefficient and to increase the installation productivity. The main goal of the optimization of the production process is to increase the capacity of the plant per unit of power of ionizing radiation. It is attained by the use of computer modeling and a precise does measurement system in the bulk of the irradiated material

Figure 4a and 4b represent the distribution of the electron and bremsstrahlung dose along the depth of the two-sided irradiated compound (wood of aspen + 70 % MMA). Curves 1, 2, 3 in Fig. 4(a) correspond to electron energies of 4, 6 and 8 MeV for two-sided irradiation. Curves 1, and 2 in Fig. 4(b) correspond to two, and one-sided irradiation of the modified wood. The bremsstrahlung radiation was generated by an electron beam with energy of 5 MeV in tantalum convertor.

Optimal thickness of the PCM articles treated by electron/bremsstrahlung beam with one-and two-sided irradiation was determined. We choose the optimal thick-ness of the PCM that corresponds to the most uniform distribution of electron/bremsstrahlung dose.

For example, for wood the optimal thickness depended on the kind of wood, type of monomer, on the degree of impregnation of wood by the monomer and on the electron energy. Optimal thickness of the PCM article with a density of 0,6-2 g/cm^3 treated by electron/bremsstrahlung beams of two-sided irradiation was in the range (3-8 cm)/(12-60 cm).

The power possibilities of the linear accelerator with electron energy 5-10 MeV and beam power 5-10 kW permitted us to design and develop an experimental installation for the manufacture of corrosion and high resistant building materials, such as parquet planks, window and door frames, construction for chemically corrosive conditions, etc. For example, such a pilot-plant equipment would be able to output 20 - 40 thousands square meters per year of modified parquet planks. The strength characteris-tics, corrosion resistance and abrasion loads of this modified parquet made of low value kinds of wood (aspen, alder, poplar, birch) exceeds natural hard woods (beech, oak, hornbeam, yew and the like).

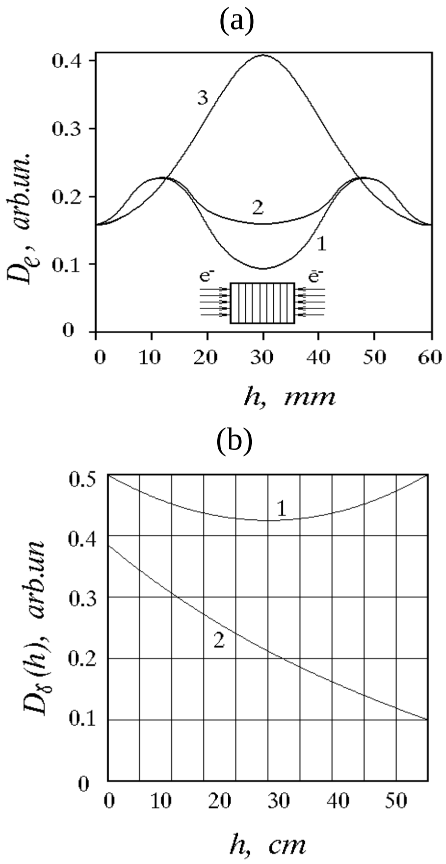

Fig.4 Distribution of electron (a) and bremsstrahlung (b) dose with depth in irradiated compound (wood of aspen + 70 % methylmethacrylate).

The high waterproof, hardness, and shape stability, high dielectric and thermoinsulation characteristics, chemical and biological durability, and perfect ornamental properties allow the materials to be used in the production of parquet flooring, window and door frames, constructions for chemically corrosive conditions and for inclement climate; manufacturing of furniture with imitation of expensive sorts of wood; construction of cottages, residential houses and industrial areas; sports and tourist equipment, household goods; production of durable components of machines and stable-in-shape cast patterns; production of electric transmission line components; production of equipment operating in rigid climatic conditions and aggressive environments; preservation and restoration of antiquities.

8 Conclusion

A versatile universal system for monitoring of radiation-chemical processes has been developed. The system is based on the use of radiation-acoustic effect, namely, the information which is carried by thermoacoustic pulse amplitude generated in the pulsed material irradiation in the course of radiation-chemical process. This information, as well as data delivered by other independent probes, and, systems measuring temperature, pressure etc., permits the control of the phase state of the manufacturing materials, of the temperature regime of processing and of the final material quality. The system can be used for comprehensive control of kinetics and thermodynamics of radiation-stimulated polymerization of monomer-oligomer in heterogeneous medium under action of pulsed electron or bremsstrahlung beams, including scanning beams. Such control permits the provision of the optimum regime of the radiation-chemical process, which leads to higher quality of the end products,

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