

## A METHOD FOR OBTAINING A POLYMER COMPOSITE BASED ON AROMATIC POLYAMIDE AND SILICON DIOXIDE

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Received 05 November 2019

Accepted 30 July 2020

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### ABSTRACT

*The method for obtaining polymer composite (PC) based on aromatic polyamide and silicon dioxide was developed, and it consists in in situ combination of the initial polymer with the filler during its synthesis. This makes it possible to significantly simplify the manufacture of products from the tested PC by eliminating the operation of mixing the original components at the preparatory stage of processing, and in accordance with this to reduce their price. The comparative structural and physical-mechanical investigations of PC have been carried out. It has been established that PCs obtained with the use of in situ combination of the aromatic polyamide and silicon dioxide have more ordered structure and higher level of physical and mechanical properties than those of materials obtained by the standard method.*

*Keywords:* aromatic polyamide, silicon dioxide, production method, structure, physical and mechanical properties.

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### INTRODUCTION

The development of mankind is closely connected with progress in the field of materials science. The creation of new materials in most cases leads to a significant technological leap in various branches of science and technology. In accordance with the principles developed by the European Commission for key technologies (European Commission Key Enabling Technologies (KETs)) the creation of such materials is one of the most topical and urgent tasks.

The polymers and polymer composite materials (PC) based on them are some of the most available materials [1, 2]. Due to their unique properties, they have found the practical application in all spheres of human activity, and they are obtained as “materials of the future” from which nowadays they receive a wide range of products ranging from dishes and ending with rocket elements [3 - 5]. However, despite of the unique set of characteristics, the products made of polymers and based on them PC, have a number of disadvantages such as a low level of heat, heat resistance and strength. Thanks to the development of new PCs for a special purpose,

based on aromatic polyamides, polyimides, polyarylates, phenolic compounds [6 - 8], these deficiencies are partially eliminated. However, products made of such materials are distinguished by a high cost, which is a consequence of the use of expensive raw PC components and a fairly complex and energy-intensive technology for their processing to products. Therefore, the actual task is to create the PC based on special-purpose polymers, which products will have a high level of physical and mechanical properties and an attractive cost from an economic point of view.

As an initial polymer matrix for the creation of such materials, the aromatic polyamide of the brand “Phenylone C1” was chosen, which products have a high level of heat and heat resistance up to 290°C and 345°C, respectively. They have a high level of the strength characteristics - stress at compression yield strength, modulus and toughness are 230 MPa, 3000 MPa and 50 kJ·m<sup>-2</sup>. To reduce the cost of the products, made of “Phenylone C1”, and to improve the level of their characteristics, dispersed and fibrous fillers are added to the polymer matrix [9, 10], due to which the level of properties of the obtained PC is controlled.

One of the most effective fillers that make it possible to maximally improve the properties of PC, based on aromatic polyamide and reduce their cost in comparison with the initial polymer, is silicon dioxide with various modifications [11]. These fillers have a developed surface (up to  $380 \text{ m}^2 \cdot \text{g}$ ), on which there are active silanol groups that interact with the polymer matrix when processing PC to products. The heat and heat resistance of such materials is  $300^\circ\text{C}$  and  $375^\circ\text{C}$ , respectively, and the stress at the compression yield strength and the modulus of elasticity are 260 MPa and 3500 MPa.

The products made of PC, based on aromatic polyamide of the brand "Phenylone C1" and silicon dioxide, are produced by compression molding in heated forms. The method of such materials processing is due to their structure. So, the melting temperature of this aromatic polyamide ( $425^\circ\text{C}$  -  $435^\circ\text{C}$ ) exceeds its active thermal destruction temperature ( $345^\circ\text{C}$  -  $355^\circ\text{C}$ ), which excludes its processing into products by highly productive methods (injection molding, extrusion) from the viscous-flowing state.

The method of compression molding in heated PC molds based on aromatic polyamide of the brand "Phenylone C1" and silicon dioxide consists of three main stages: preparatory, basic and final [12]. At the preparatory stage operations are carried out to obtain a polymer composition followed by its briquetting. At the main stage the pressing process and cooling the finished products takes place according to a predetermined

procedure. At the final stage the machining operations are performed to convey the desired appearance to finished products. The method of processing PC based on aromatic polyamide of the brand "Phenylone C1" is rather complicated and energy-intensive, which leads to an increase in the finished products cost. Therefore, the main purpose of this work is not only to create a PC with high level of physical and mechanical properties, but it is to simplify it without losing the quality of the resulting products.

## EXPERIMENTAL

### Materials

As a matrix polymer we selected aromatic polyamide of brand "Phenylone C1", produced by OOO Uniplast, Vladimir (Russia). This material is a copolymer poly(meta- and para-phenylene isophthalamide), which structural formula is shown in Fig. 1.

In its original form the aromatic polyamide of brand "Phenylone C1" is a finely divided a brown colored powder with a basic size of the particles lied in the range of 20 - 40 microns. Products from this polymer had high level of heat resistance (to  $350^\circ\text{C}$ ) and durability (to 240 MPa).

As a filler a silicon dioxide was selected, which was obtained by the authors by a sedimentation from aqueous solution of sodium metasilicate [13]. The received filler has the developed surface with average size of particles 5 - 10  $\mu\text{m}$  (Fig. 2).

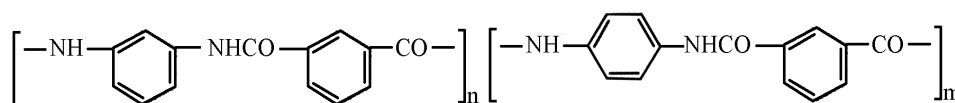


Fig. 1. The structural formula of aromatic polyamide of brand "Phenylone C1".

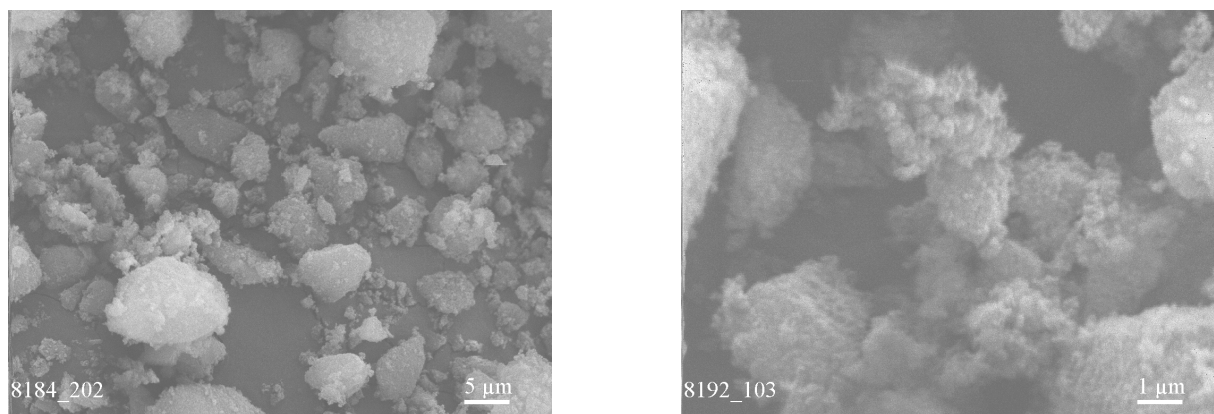


Fig. 2. Microphotographs of particles of the silicon dioxide.

## Methods

Microphotographs were obtained by the electronic scanning microscope Superprobe-733 (Jeol). X-ray diffraction analysis of PC, based on aromatic polyamide, was obtained by the X-ray diffractometer "DRON-2". Compressive stress at yield ( $\sigma_y$ ) was found according to ISO 604 on a universal tensile testing machine "Heckert FP 100/1". Vicat softening temperature of thermoplastics ( $T_{VC}$ ) was determined according to ISO 1183-1 at FWV-633/10 device.

## RESULTS AND DISCUSSION

The purpose of the work is achieved through the use of a fundamentally new method for processing PC, based on aromatic polyamide of brand "Phenylone C1" and silicon dioxide in the product, which includes combining in situ the initial components of the polymer composition during the synthesis of the filler.

In accordance with the developed method the dispersed powder of aromatic polyamide, an aqueous solution of sodium metasilicate is added, and with constant stirring a synthesis of silicon dioxide obtained from the gel formed when the reaction medium is reacted with a 10 % acid solution has been carried out. The resulting gel with chaotically dispersed particles of the dispersed polymer is dried and washed in distilled water to remove the by-products (salt, acid residues), the re-drying and grinding process has been made to produce the polymer composition in the form of a particulate press powder. Further processing is carried out in accordance with the standard method of obtaining products from PC based on aromatic polyamide of brand "Phenylone C1".

This approach to processing PC based on aromatic polyamide and silicon dioxide makes possible to simplify considerably the method of obtaining products from them by eliminating the operation of mixing the initial components at the preparatory stage of processing, thus reducing the cost of products.

The change in the method of processing the investigated PC into products will affect their structures [14]. For this investigation the micrometric images of the cleavage surfaces were obtained (Fig. 3) and X-ray diffraction analysis (Fig. 4) of the PC was carried out by the standard method and using the method of in situ combining the initial components of the polymer composition.

From the microphotographs it can be seen that the PC structures obtained by the standard method and

by using the method of in situ combining the initial components of the polymer composition are significantly different. Thus, the morphology of the cleaved surface of the PC obtained by using the method of in situ combining the initial components of the polymer composition differs by the more fragile nature of the cleavage in comparison with the material obtained by the standard method.

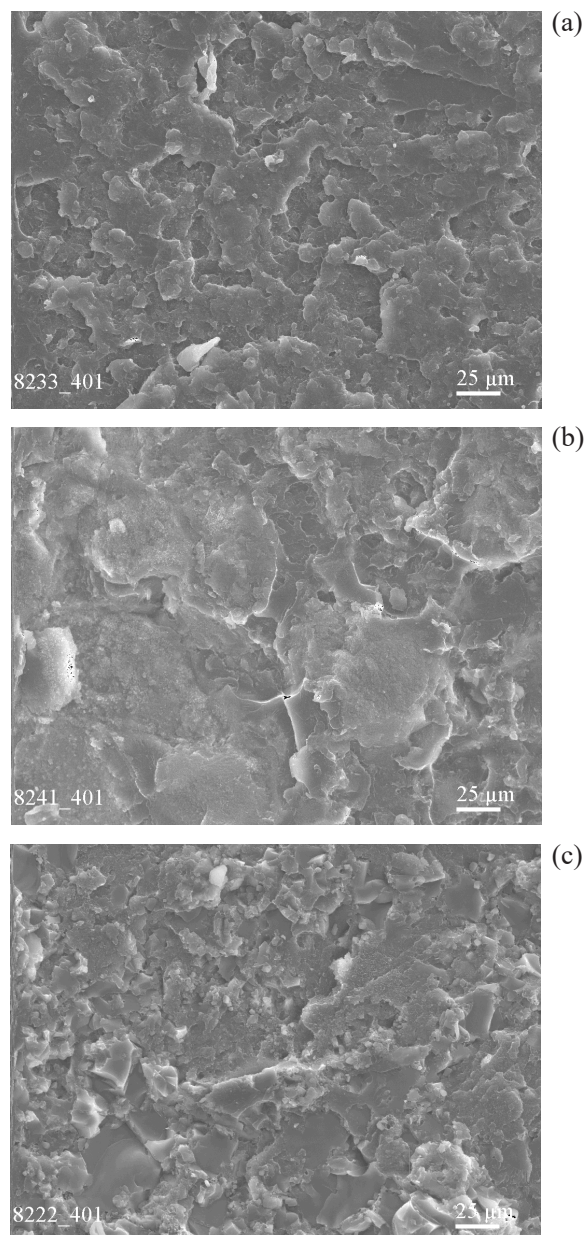


Fig. 3. Microphotographs of the cleavage surfaces (a) - of the original polymer; (b) - PC obtained by the standard method; (c) – PC prepared by the method of in situ combining the initial components of the polymer composition.

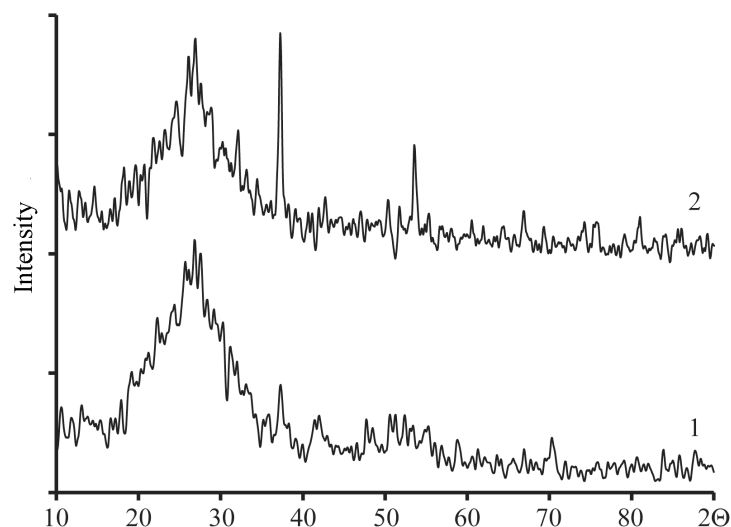


Fig. 4. X-ray diffraction of PC based on aromatic polyamide and silicon dioxide obtained by: 1 - standard method; 2 - using the method of in situ combining the initial components of the polymer composition.

X-ray diffraction patterns of PC based on aromatic polyamide and silicon dioxide obtained by different methods also have differences (Fig. 4).

From the X-ray diffraction patterns it can be shown two amorphous halo at 18 - 34 and 45 - 57 deg, which is typical for the polymer matrix. It should be noted, that these halo's is less expressed in materials obtained

by using the method of in situ combining the initial components of the polymer composition comparison with the material obtained by the standard method. The X-ray diffraction patterns of materials obtained by using the method of in situ combining the initial components of the polymer composition has more intensive peaks at 37 and 55 deg, which is typical for silicon dioxide of

Table 1. The physical and mechanical properties of PC based on aromatic polyamide and silicon dioxide.

Property	Filler content, wt. %	Method for the preparation of a PCs	
		standard	by using of <i>in situ</i> combining the initial components of the polymer composition
Compressive stress at yield $\sigma_y$ , MPa	10	243	262
	20	268	276
	30	264	281
	40	225	248
	50	186	150
	60	115	65
Vicat softening temperature $T_{VC}$ , °C	10	273	284
	20	283	292
	30	288	302
	40	290	318
	50	292	332
	60	301	over 380

different modification ( $\alpha$ -quartz and  $\alpha$ -cristobalite) [15].

The differences in the X-ray diffraction patterns point a change of macromolecular structure of PCs obtained by using the method of in situ combining the initial components of the polymer composition in comparison with the material obtained by the standard method. On X-ray patterns of PCs obtained by using the method of in situ combining the initial components of the polymer composition more intense absorption peaks are observed throughout the whole study area of angles. This indicates an increase in their degree of crystallinity in comparison with PCs obtained by the standard method. An increase in this parameter is associated with a change in the ordering degree of the basic structural elements of the examined PC. Thus, it leads to a change in the level of properties obtained from them products.

Therefore it is important to study the effect of the PC processing method on their level of physical and mechanical properties. The results of the experiments are given in the Table 1 was selected. As can be seen from the results the products from PC obtained by using the method of in situ combining the initial components of the polymer composition in terms of the level of physical and mechanical properties are 10 - 15 % higher than analogues obtained by the standard method. That is the consequence of the change in their structure and the increase in the degree of ordering of the PC structural elements when it is received and processed into products.

It should be noted that the compressive stress at yield of PC with high degree of filling (more than 50 %), obtained by the method of in situ combining the initial components have smaller values than in analogues obtained by standard method. It is connected with structural features of these materials, which consist in the formation of a continuous "inorganic frame" in them which has a significant effect on the strength properties of PC, playing the role of a force matrix in it.

## CONCLUSIONS

The results of this research show that the application of the method of in situ combining the initial components of the polymer composition for processing the PC, based on aromatic polyamide of brand "Phenylone C1" and silicon dioxide in products, allows not only to simplify the production method and to reduce the cost of the final products, but also to improve significantly their physical and mechanical properties.

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