479

Use of Hydrolyzed Lignin for Obtaining Composite Materials

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Abstract

Possibility of obtaining a composite material from the wastes of yeast hydrolysis product – hydrolyzed lignin – and polyethylene as a thermoplastic binder has been demonstrated. The influence of the dispersion degree of the hydrolyzed lignin and of the composition of components on the physicomechanical properties of the material has been estimated. It has been established that in order to obtain a lignoplastic possessing a high strength and low water absorption, it is necessary to use finely divided lignin with a 20-40 % polyethylene content of the initial mixture.

INTRODUCTION

The problem of utilization of hydrolyzed lignin (HL) which arose with the development of industrial hydrolysis technology in the 30ies of the XX century is important today, since 30-40 % of the initial wood raw material goes to wastes whose bulk is made up by technical lignin [1]. One of the best known methods of utilizing HL is making briquettes out of it which are used as substitutes for the traditional kinds of fuel or as carbonic reducing agent in ferrous and non-ferrous metallurgy [2]. In small amounts, HL is used in agriculture for production of organomineral fertilizers, fodder yeast and additives *etc.* [3].

The widest field for comprehensive utilization of HL both as the main raw material and in combination with other materials, is constructive industry where it can be used for obtaining composition materials [4]. The use of HL for production of lignowood slabs, lignoplastic articles and heat-insulating materials [5]. As binders in such materials mainly expensive and toxic phenol- and carbamide-formaldehyde thermoreactive resins are used. This makes the technological processes time consuming and requires complicated equipment.

However, despite the partial utilization of lignin wastes formed in the hydrolysis technology, its major part is not utilized and goes to dumps, which are ecologically dangerous. Search for new ways of HL utilization is an important problem of modern science and technology. One of such ways may be production of constructive materials based on HL and thermoplastic polymers. Advantages of thermoplastics are their high plasticity, non-toxicity, and a simpler processing technology with a shorter moulding cycle as compared to those of materials based on thermoreactive resins.

The goal of the present work was a study of the possibility of creating new composite materials based on HL and polyethylene (PE) as a thermoplastic binder, and investigation of their physicomechanical and thermophysical properties.

EXPERIMENTAL

For obtaining composite materials, HL of the Rechitsa Hydrolytic Yeast Mill and PE powder with the particle diameter of up to 3 10^{-5} m and the density of 940 kg/m³ were used. The initial HL represented a humid homogeneous mass that contained particles of various sizes, including wood chips. For this reason, after drying the HL to the residual humidity of 9–15 %, large chips were sifted away and not taken into account. The granulometric composition of HL was determined by means of sieve analysis using a set of sieves with the mesh dimensions of 0.05 to 2.5 mm.

Lignoplastic specimens were obtained by hot moulding of a thoroughly stirred mixture of HL and PE at the temperature of 433-443 K and the pressure of 20-25 MPa.

Specimens of composite materials were tested for compression strength, water absorption for 24 h, water resistance for 7–90 days and heat conduction according to standard methods. Tests for compression strength were carried out on a tearing machine P-0.5, the heat conduction was estimated on a heat conduction meter $IT-\gamma-400$.

RESULTS AND DISCUSSION

The hydrolyzed lignin is a complex polydispersed mixture of hydrolytic degradation of wood, and its granulometric composition influences in a high degree the physicomechanical and technological properties of the material. The HL particle size, according the sieve analysis data, varies from a few millimetres to the size of colloid particles (Fig. 1). As it follows from Fig. 1, the most probable dispersion of HL is the fraction of 0.25–1.0 mm, since it comprises the major part of particles (54.8 % of the sample mass). This fraction contains the largest amount of properly lignin and of mineral inclusions.

The influence of HL dispersion on the properties of the material obtained on the basis of



Fig. 1. Granulometric composition of hydrolyzed lignin.



Fig. 2. Influence of HL dispersity on the properties of lignoplastic: a – density (1) and water absorption (2); b – compression strength ($\sigma_{e,s}$).

HL and PE at the mass ratio of 60 : 40 is shown in Fig. 2. One can see that as the lignin particle size decreases, the density and compression strength of lignoplastic increase, and water absorption decreases, which is determined by the chemical composition of the fractions and their sizes. The fraction of 0.32/0.05contains the largest amount of extractable colloidal lignin and water-soluble substances, which improves its physicomechanical properties. A certain increase in lignoplastic strength is observed when more coarse (2.5/1.0 mm)lignin fractions are used, which is accounted for by the fact that this fraction contains the residual cellulose part of the plant tissue having a fibrous structure and playing the role of a reinforcing substance [6].

The HL dispersion considerably influences also the water resistance of lignoplastic. More water-resistant turned out to be the material obtained from finely dispersed HL (fraction of 0.25/0.05 mm) (Fig. 3). In a week of testing, its water absorption did not exceed 2.5 %, whereas the DW of lignoplastic obtained from coarse HL fractions (2.5/1.0) was 11.5 %, and the material obtained from only HL became completely degraded. In further increase of water-resistance testing time to 30–90 days, a swelling of the specimen and its complete saturation took place, *i.e.* practically a constant water-resistance of the material was achieved. From the



Fig. 3. Dependence of water absorption on the duration of tests at various HL dispersity, mm: 1.6/1.0 (1), 1.0/0.4 (2), 0.4/0.32 (3), 0.32/0.25 (4), 0.25/0.05 (5).

data obtained it follows that in order to obtain a lignoplastic with a high strength and low water absorption, it is necessary to use finely dispersed HL with the particle size within the range of 0.4/0.05 mm.

When using finely dispersed HL, a higher homogenization of the mixture and a uniform distribution of its particles in the polymer binder melt are achieved, which ensures a better contact of mixture components and their physicochemical interaction. In further compression of the mixture, the PE which possesses a lower melting point and a higher plasticity than lignin, at its optimal content in the compression mass, completely fills up the microvoids and pores of HL and binds its particles to form a homogeneous strong monolithic structure.

Optimization of lignoplastic properties is achieved at the 20-40 % PE content of the mixture (Fig. 4). The material based only on lignin has a comparatively low compression strength (12 MPa), with a characteristic of brittle failure. The compression strength of the initial polyethylene is twice as high as that of lignin, and the material is characterized by ductile failure. From the data presented in Fig. 4,



Fig. 4. Dependence of water absorption (1) and compression strength (2) of the lignoplastic on PE content.

one can see that when PE is introduced into the HL moulding material, the physicomechanical properties of the composition material become considerably better. Thus, the water absorption of samples containing 20-40 % PE decreased by 4-6 times, and the strength increased by 2.5-3 times. Besides, the lignoplastic obtained with such a proportion of components has low heat conduction. In Table 1, results of estimation of HL- and PE-based lignoplastic heat conduction within the temperature range of 298-348 K are presented, because PE having a low heat resistance begins to melt at higher temperatures. One can see that as the temperature and the PE content increase, the lignoplastic heat conduction decreases slightly and is within the limits of 0.12-0.18 W/(m K), which is characteristic of heatinsulating materials.

CONCLUSION

The studies have demonstrated that addition of 20-40 % of polyethylene into the moulding mass of hydrolyzed lignin makes it possible to obtain composite materials with low water absorption, high compression strength

TABLE 1

Coefficient of heat conduction of HL- and PE-based lignoplastic, $W/(m\ K)$

PE content, % mass	Temperature, K			
	298	325	348	
_	0.17	0.15	0.13	
30	0.18	0.14	0.12	
40	0.18	0.17	0.15	
60	0.16	0.15	0.14	

and low heat conduction. Such materials may be used in the production of constructive articles, facing and heat-insulating materials *etc*.

In this way, the use of hydrolyzed lignin in combination with polyethylene (or its wastes) permits not only solving the problems linked with environment protection and saving of resources, but also obtaining a high-quality composite material.

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