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Processing Factors and Properties of Thermal Insulation Boards Made of Plant Fillers

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Abstract. Wood processing soft waste is mainly used in the production of fuel briquettes, irrecoverable (non-recyclable) waste from spinning flax and cotton are incinerated or sent to dump. The development of methods for recycling non-recyclable cellulosic waste through the product manufacturing is relevant, both from the resource conservation perspective, as well as the environmental point of view. The issues of plant waste recycling through the manufacturing of various types of products are widely developed in the Russian and foreign scientific research practice. Researchers deal with the processing of wheat, rice straw, bamboo stalks, and other cellulosic materials. There is a plenty of published information on methods of soft wood waste recycling. However, no research on recycling irrecoverable waste of spinning flax and cotton fibers had been carried out before this paper. We propose to produce thermal insulation boards based on phenol-formaldehyde resol binder using flax and cotton spinning waste and soft wood processing waste. The wet production method used here involves mixing the filler with water, a precipitant solution and a binder. After spinning the material is dried. The paper presents the results of determining the physical and mechanical properties and thermal conductivity coefficient of boards made of plant waste. The research was carried out according to the B-plan of the second order. Adequate regression mathematical models of the dependences of physical and mechanical parameters of the boards on the varying factors of the production process were developed according to the experimental data processing results. Using the developed regression models we built the response surfaces of the composite parameters: the bending strength of the boards, the thickness swelling of the boards after 24 h of exposure in water and the thermal conductivity coefficient. Nomograms of the dependencies of board parameters on the values of varying factors have been developed based on the mathematical models analysis. The nomograms are the basis for the development of practical recommendations for determining the rational values of the parameters of insulation board materials production from irrecoverable waste of spinning flax and cotton and soft wood processing waste.

Keywords: plant waste, flax, cotton, wood, composite boards, regression model, ultimate strength, static bending, thickness swelling, thermal conductivity coefficient

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Научная статья

Технологические факторы и свойства теплоизоляционных плит из растительных наполнителей

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Аннотация. Мягкие отходы переработки древесины в основном используют для производства топливных брикетов, невозвратные (неиспользуемые) отходы прядения льна и хлопка сжигают или отправляют на свалку. Поиск способов утилизации невозвратных целлюлозосодержащих отходов путем производства продукции актуален и с позиций ресурсосбережения, и с экологической точки зрения. В практике российских и зарубежных научных исследований широко разрабатывается такое направление, как утилизация растительных отходов путем производства продукции. Изучаются вопросы переработки пшеничной, рисовой соломы, стеблей бамбука и других целлюлозосодержащих материалов. Существует много публикаций о способах утилизации мягких древесных отходов. Однако исследований в области переработки невозвратных отходов прядения льняных и хлопковых волокон до работы авторов статьи не проводилось. Нами предлагается изготавливать из отходов прядения льна и хлопка и мягких отходов переработки древесины теплоизоляционные плиты на фенолоформальдегидном резольном связующем. Используется мокрый способ производства, при котором наполнитель смешивается с водой, раствором осадителя и связующего, после отжима материал сушится. Представлены результаты определения физико-механических показателей и коэффициента теплопроводности плит, изготовленных из растительных отходов. Исследование проводилось по В-плану второго порядка. Обработка экспериментальных данных позволила разработать адекватные регрессионные математические модели зависимости физико-механических показателей плит от варьируемых факторов процесса производства. По этим регрессионным моделям построены поверхности отклика показателей композита: предела прочности плит при статическом изгибе, их разбухания по толщине за 24 ч пребывания в воде и коэффициента теплопроводности. На основе анализа моделей получены номограммы зависимости показателей плит от варьируемых факторов. Номограммы являются основой для разработки практических рекомендаций по определению рациональных параметров производства теплоизоляционных плитных материалов из неиспользуемых (невозвратных) отходов прядения льна и хлопка и мягких отходов переработки древесины.

Ключевые слова: растительные отходы, лен, хлопок, древесина, композиционные плиты, регрессионная модель, предел прочности, статический изгиб, разбухание по толщине, коэффициент теплопроводности

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Introduction

Processing of plant materials, such as wood and annuals (flax, cotton, etc.) inevitably generates waste, some of which is irrecoverable, i.e. is sent to the dump or incinerated. Both ways of plant waste disposal negatively affect the biosphere. The problem of recycling waste by using it in product manufacturing is relevant worldwide. Russia generates about 4.5 million tons of wood waste per year, and despite there has been a decline in wood waste over the past three years its amount exceeds the data from 2012 according to the Federal Service for Supervision of Natural Resources (Rosprirodnadzor) [24]. Approximately 1 million tons of wood waste in Russia remains unrecycled annually [39]. However, not only Russia rich with wood resources generates a significant amount of irrecoverable wood waste. According to the Statistical Office of the European Union (Eurostat) such countries as Germany, France, Great Britain, etc., generate a vast amount of wood waste annually [6]. Germany leads here with about 400 thousand tons per year [5].

This is the reason for the high relevance of works on waste recycling. Conventionally, plant waste is used in hydrolysis production [16, 30]; since the 20th century it has been used for fuel needs [8]. Further increase in the use of wood waste as a fuel is predicted both abroad [27] and in Russia [35], which is positive from an energy point of view [15, 28]. However, this trend does not reduce the environmental impact caused by the burning of plant materials [17]. More preferable methods of plant waste recycling are bioconversion [1, 3, 4, 19, 29] and hydrolysis [18, 23, 27, 30]; plant waste recycling for the purpose of producing phenolic compounds, oligosaccharides, and polysaccharides with a low degree of polymerization is also promising [14].

Engineering progress in the field of chemical and chemical-mechanical processing of plant raw materials allows almost all biomass to be used, but different types of processing have different efficiency. The share of the final product output in the wood chemical industry (pulp and paper production, hydrolysis production, including ethanol production) is 62–68 %, while the output in the board production reaches 90 % [19]. Technologies for the production of board materials from fine plant particles enable the use of various types of lignocellulosic waste for the production of construction materials. However, the extensive damage of plant cells in waste [36] hinders its use for the production of structural building materials, excluding materials on mineral binders. There are solutions for the return of industrial waste of wood fiberboard production to the main process, but usually this is wood fiber lost with circulating water [2] or during board size trimming [20].

A promising direction for recycling plant waste is the production of thermal insulation board materials, such as soft fiberboard. T. Tabarsa remarks that the production of thermal insulation boards is important in terms of resource-saving technologies, but problematic for European countries due to the limited reserves of forest resources near industrially developed areas. So it requires the use of alternative plant materials [12, 21, 34]. Urea-formaldehyde (UFC), polyisocyanate (PMDI), and phenol-formaldehyde (PFC) resins are used as binders [21, 22].

There is a great deal of research in the field of producing composite materials from non-wood plant fibers and waste, including those based on a combined wood waste filler with the addition of plant fibers. G. Han et al. [11] and S. Halvarsson et al. [9, 10] studied the performance of wood fiberboards with wheat and reed grass added based on UFC and melamine-formaldehyde (MFS) binders. J.E.G. van Dam et al. used coconut fiber as a composite filler [7, 40]. Composites made of bamboo and rice straw have been developed [25]. J. Kanagaraj et al. [13] studied composite materials of cotton fibers and corn stalks. Work [26] presents the results of studying the physical and mechanical properties of composite materials made of kenaf fibers carried out by M.J. Saad and I. Kamal. The research results on the use of flax fiber processing waste in the production of composites are also known [31, 34].

A wide range of research on recycling plant waste into composite materials shows the relevance of this direction – development of thermal insulation boards made of plant waste and based on thermosetting binder. However, there are no developments in Russian and foreign research practice on usage of soft wood processing waste and irrecoverable waste of flax and cotton spinning as a filler of thermal insulation board composite materials. The research aims at substantiation of rational values of production factors of thermal insulation composites made of soft wood waste and irrecoverable cellulosic fiber spinning waste with the necessary physical-mechanical and operational properties.

Research objects and methods

The laboratory of the Department of Logging and Wood Processing Industries (Kostroma State University, Kostroma, Russia) develops thermal insulation board materials with a filler made of wood waste and irrecoverable waste of the flax and cotton fibers production [38].

The analogue material is wet-processed thermal insulation soft fiberboard (grade M3 according to the Russian state standard GOST 4598–2018). The material being developed is not a complete analogue of soft fiberboard, therefore, it is not possible to speak of the compliance of its parameters with the fiberboard parameters. The materials are united by the method of wet formation and drying of boards. It is impossible to obtain a low-density material with the desired complex of performance properties by the wet method only from soft wood waste. This is due to the insufficient amount of active hydroxyls in pulp microfibrils without wood grinding. Grinding of this wood material will be ineffective due to significant damage of the original wood fine material and a high yield of fiber fragments. Finely-dispersed waste from the spinning of plant annuals is formed after chemical treatment of raw materials at the stage preceding fiber spinning and subsequent repeated mechanical exposures. At the same time, the cellulose polymerization degree of materials decreases [32, 36], the mobility of cellulose macromolecules increases and hydroxyls are activated on the microfibril surface, which leads to an increase in the material intermolecular hydrogen bonds [33].

The fractional composition of the filler made of plant waste was determined by sieving in a sieve analyzer and weighing the fraction share. The results of determining the fractional composition of the filler are presented in table 1.

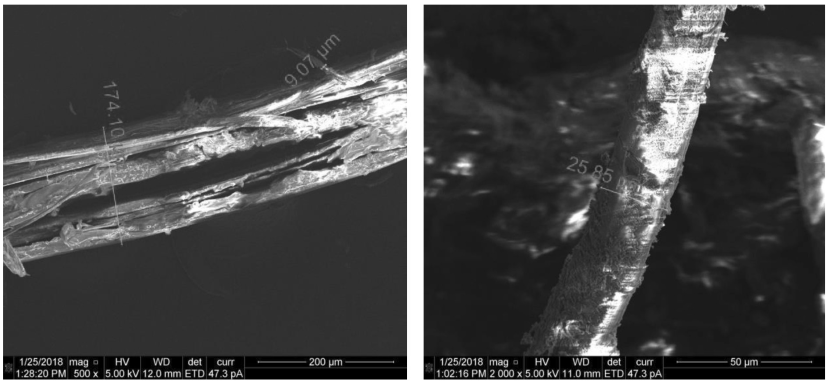
Table 1

Results of determining the filler fractional composition

Fraction	Fraction share i_{fr}, %		
	cotton waste	flax waste	soft wood waste (coniferous)
–/10	1.14	6.01	1.34
10/7	1.82	0.60	20.16
7/5	2.94	0.90	14.0
5/2	19.64	7.46	29.72
2/0.5	36.36	33.82	28.29
Tray	38.10	51.21	6.49

Irrevocable waste of flax and cotton spinning has an average length: 4.76 mm for cotton; 4.12 mm for flax.

Photographs of flax fiber spinning waste and flax waste composite made by the authors using a FEI QUANTA 3D FEG scanning electron microscope are shown in fig. 1.



a *b*

Fig. 1. Photographs: *a* – flax fiber spinning waste; *b* – plant waste composite

The particles of the plant waste filler have significant damage and a large specific surface area, therefore, the binder covers only part of the filler surface (fig. 1*b*). Table 2 shows the results of the physicochemical analysis of the plant waste.

Table 2

Composition of plant materials, %

Filler		Cellulose	Lignin	Ash content	Water-soluble substances *
Irrevocable spinning waste:	cotton	44.0	22.7	17.0	0.01
	flax	54.0	24.9	5.0	0.02
Soft wood waste:	pine	42.0	25.7	0.3	1.85
	spruce	53.8	28.0	0.2	1.70

*Soluble in hot water.

A synthetic phenol-formaldehyde binder (PFB) was used as a matrix for the composite. The binder consumption varied from 0 to 40 % of the filler weight.

The filler was mixed with water; the binder and the precipitant (aluminum sulfate $Al_2(SO_4)_3$) were added in an amount of 1 % of the resin weight. The material was placed in a mold with a grid, squeezed out at a specific pressure of 0.95 MPa, removed from the mold and placed in a drying oven. Board samples were dried for 2 h at a temperature of 100–170 °C, then conditioned for 24 h at a temperature of 20 °C. The average density of composite boards was 275 kg/m^3.

Studies at the previous stage showed a significant scatter in values of parameters of the composites being developed [37]. The B-plan of the second order was used as a method of experimental research in order to substantiate the rational values of the production process factors of plant waste material. Table 3 presents the matrix of the B_3 plan. This plan is close to D-optimal, i.e., the generalized variance of the estimates of the regression coefficients is close to the minimum. This is a significant advantage of the B-plan.

Table 3

B_3 plan matrix in coded levels of factors

No.	X_1	X_2	X_3
1	+	+	+
2	–	+	+
3	+	–	+
4	–	–	+
5	+	+	–
6	–	+	–
7	+	–	–
8	–	–	–
9	+	0	0
10	–	0	0
11	0	+	0
12	0	–	0
13	0	0	+
14	0	0	–

The assessment of the physical and mechanical properties of the boards was carried out in accordance with the Russian state standard GOST 10633–2018 "Wood-Shaving and Wood-Fiber Plates. General Regulations in Testing Physical and Mechanical Properties".

Results and discussion

The factors varied in the experiment and their levels are presented in table 4. Output values: Y_1 – static bending strength (σ_i), MPa; Y_2 – thickness swelling of boards for 24 h (P_h), %; Y_3 – thermal conductivity coefficient (λ), W/mK.

Table 4

Variable factors and their natural and coded levels

Factor	Factor level		Variation levels			Variation interval, Δ_i
	natural	coded	–1	0	+1	
Mass fraction of binder additive, %	F_m	X_1	0	20	40	20
Drying temperature, °C	T_{dry}	X_2	100	135	170	35
Share of wood waste additive, % by weight of a plant filler	S_w	X_3	0	25	50	25

Mathematical models of parameters of flax fiber spinning waste composites with the addition of soft wood wastes (in coded levels of factors) were obtained based on the results of experimental data processing. Equations show them:

$$Y_1 = 0.334 + 0.071X_1 + 0.039X_2 - 0.057X_3 + 0.026X_1^2 + 0.026X_2^2 + 0.026X_3^2 + 0.011X_1X_2 - 0.011X_1X_3 - 0.011X_2X_3;$$

$$Y_2 = 12.51 - 3.85X_1 - 1.43X_2 + 0.81X_3 - 0.561X_1^2 + 0.539X_2^2 - 0.561X_3^2 + 0.300X_1X_2 + 0.275X_1X_3 - 0.250X_2X_3;$$

$$Y_3 = 0.067 + 0.003X_1 - 0.001X_2 + 0.002X_3 - 0.002X_1^2 - 0.001X_3^2 - 0.001X_1X_3.$$

Figure 2 shows the response surfaces of the dependences of the output values on the variable factors. Namely, the dependences of the strength of the boards at static bending and thickness swelling for 24 h on the share of binder additive (X_1) and drying temperature (X_2).

The increase in the share of binder additive and the drying temperature of the boards in the entire range of factors variation increases their static bending strength. The increase of the soft wood waste share in the composite filler up to 50 % (by the filler weight) results in the reduction of the board static bending strength by 0.1 MPa, i.e., the strength is reduced by 17–30 % (depending on the combination of levels of the varying processing factors in the experiment). It is recommended to use the maximum values of the binder additive share and the composite drying temperature for the maximum share of wood waste additive in order to ensure the minimum strength of the analogue material – soft thermal insulation wood fiber boards – 0.4 MPa. Sufficient operating strength without a binder will be provided only by the combination of factors "maximum drying temperature + filler without wood waste". It is possible to use the drying temperature of boards of 100–170 ° C at any share of wood waste additive when the share of the binder additive is 20 %.

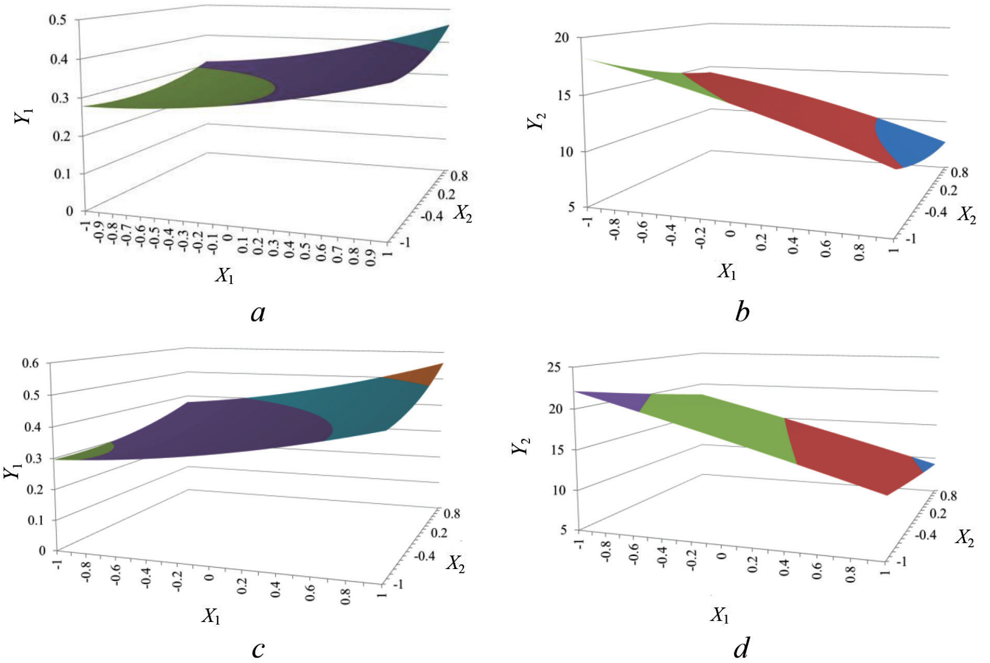


Fig. 2. Response surfaces for the composites: *a*, *b* – with the maximum share of soft wood waste in the filler (X_3 = +1); *c*, *d* – with maximum share of flax and cotton spinning waste in the filler (X_3 = –1)

An increase in the share of the soft wood waste additive to the filler results in a 10–30 % increase in thickness swelling of the boards after 24 h of exposure in water (depending on the combination of the levels of factors). A significant decrease in the thickness swelling of the boards provides an increase in the binder share. An increase in the drying temperature of the boards reduces their thickness swelling less significantly.

At any share of soft wood waste additive and any combinations of processing factors levels the thermal conductivity coefficient of the composite boards is in the range of 0.058–0.070 W/mK, i.e. the material is characterized by good thermal insulating properties. The share of the binder additive causes an increase and then stabilization of the material thermal conductivity coefficient due to the higher value of the parameter for the cured binder in comparison with the thermal conductivity coefficient of the cellulosic filler. An increase in the drying temperature of the boards, in addition to the improvement of the physical and mechanical properties of the material, also causes a 2.5–3.0 % decrease in the composite thermal conductivity coefficient.

Based on the research results a set of nomograms was developed for practical purposes. They enable to rapidly select the structure-forming components of the composite with the given properties. As an example, figure 3 shows the dependence of physical and mechanical properties on the share of binder additive for composites made of flax waste and cotton waste with the addition of various soft wood waste.

The following symbols are used in the figure: FW – flax waste; BC – bark of conifers; BH – bark of hardwood; SS – shavings of softwood; SH – shavings of hardwood; numbers indicate the shares of the filler additive.

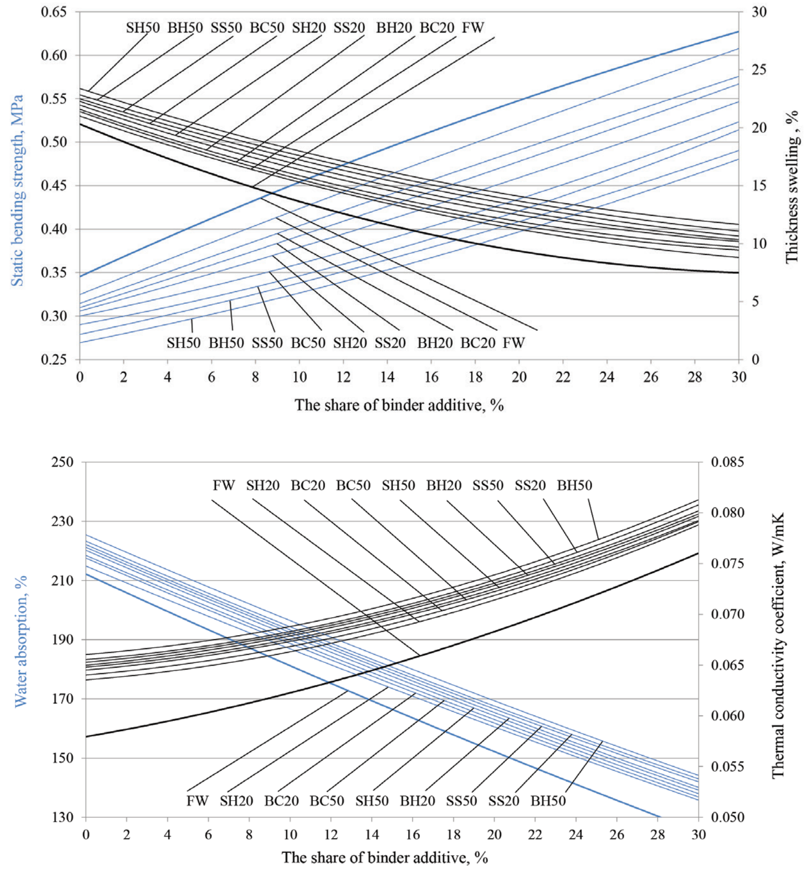


Fig. 3. Dependences of physical and mechanical properties on the share of the binder additive (PFB) for the flax waste composite with the addition of soft wood waste

Conclusion

Cellulosic waste, such as soft wood, namely wood shavings and bark of deciduous and coniferous species, and irrecoverable waste of flax and cotton spinning were used as fillers of thermal insulation boards. The lignin content in irrecoverable plant waste of flax and cotton is comparable to that in wood raw material. This is due to the presence of a significant amount of bolls, stalks (cotton waste) and shover (flax waste) in the spinning waste. The cellulose content in cotton waste is the same as in wood. The cellulose content in flax production waste is significantly higher than in wood raw materials. Flax and cotton wastes contain less water-soluble substances compared to wood raw materials. Flax and cotton plant waste has significant ash content, which is the reason for the waste contamination. The experiment showed that despite the high ash content, flax and cotton irrecoverable waste can be used for the thermal insulation board production. The high content of cellulose in the plant waste enables to create a composite structure via hydrogen bonds between the particles and covalent bonds with the binder.

The required operational parameters of the thermal insulation composite made of industrial production cellulosic waste (soft wood waste and irrecoverable fine-dispersed waste of flax and cotton spinning) are provided without the energy-consuming operation of wood material grinding necessary for the analogue material (soft fiberboard). Chemical and mechanical effects on flax and cotton plant fibers in technological processes of raw material preparation and fiber spinning provide activation of hydroxyls on the surfaces of pulp fibrils, which creates conditions for the formation of hydrogen bonds front and covalent bonds between pulp and binder.

Different levels of the production process factors depending on the composition of the filler are recommended to ensure the required physical and mechanical properties of the composite thermal insulation material with the filler based on the cellulosic waste and polycondensation phenol-formaldehyde binder.

The recommended drying temperature is 100–170 °C, when using only irrecoverable flax waste as a filler, according to the schedules the share of the PFB additive should be not less than 20 %. It is necessary to increase the PFB mass fraction up to 40 % when using the combined filler of 50 % of soft wood waste and 50 % of flax fiber spinning waste.

The thermal conductivity coefficient of the material is 0.062–0.070 W/mK at the PFB additive of 20 % and more, at any drying temperature and at the maximum share of soft wood waste additive, which indicates strong thermal insulation properties of the material.

Thus, this combination of processing factors can be recommended for the production of thermal insulation boards on phenol-formaldehyde binder with a combined filler of soft wood waste and irrecoverable waste of flax spinning.

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