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TRIBOLOGICAL AND PHYSICAL PROPERTIES OF POLYPROPYLENE FILLED BY NATURAL FIBRES

WŁAŚCIWOŚCI TRIBOLOGICZNE I FIZYCZNE POLIPROPYLENU WYPEŁNIONEGO WŁÓKNAMI NATURALNYMI

Key words:

natural fibres composite, polypropylene, coconut fibre, flax fibre, cellulose fibre, fleece fibre

Słowa kluczowe:

włókna naturalne kompozytowe, polipropylen, włókno kokosowe, włókna lniane, włókna celulozy i wełny

Summary

This paper deals with the tribological and physical property analysis of polypropylene filled by selected natural fibres of vegetable and animal origin

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(coconut, flax, cellulose and fleece fibres) from the application point of view. Tribological properties of injection parts are evaluated through surface roughness and friction coefficient. Physical properties are evaluated in light of melt flow behaviour, the strength characteristics of injection parts, flammability, and shape stability both under lower and higher temperatures.

INTRODUCTION

Polymeric materials and their composites belong to the very progressive kind of research. They are part of every advanced technology and these materials are often irreplaceable. Truly very new trend rests in the substitution of glass fibres by natural fibres that are the subject of interest by the polymer processors for environmental reasons. Natural fibre application is an important material alternation that is also traditionally focused on the automotive industry, which continuously represents a pressure in improving utility properties of plastics and composites with polymeric matrixes. Compared to other fibre materials, there are many advantages in natural fibres used in composites production, as e.g. low weight, low wear, low flammability, biodegradability, and mainly low price,

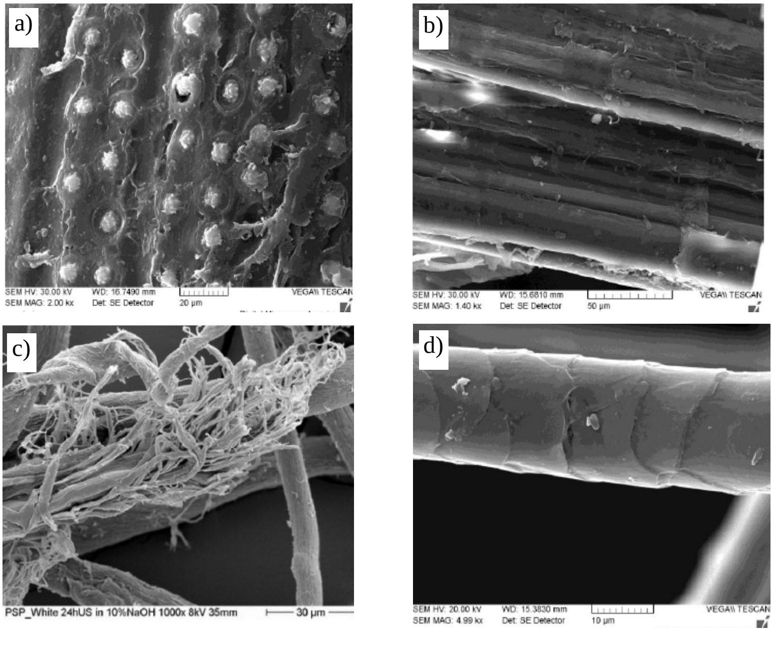

Fig. 1. Scanning electron microscope images showing a coconut (a), flax (b), cellulose (c) and a fleece fibre (d)

Rys. 1. Obraz mikroskopowy (SEM) z włókien kokosowych (a), lnianych (b), celulozowych (c) i wełny (d) which is independent of the price of oil. Due to the relative movement of plastic mechanical parts, there is constant friction and wear, which strongly influence their service life. Friction properties of polymers and their wear are influenced by quite a number of factors, e.g. choice of material couple (polymer-on-polymer contacts or non-polymer-on-polymer contacts) **[L. 1]**, crystallinity **[L. 2]**, surface treatment of the part, surface roughness **[L. 3]**, sliding speed, service temperature **[L. 3]**, specific pressure **[L. 4, 5]**, additives (in dependence on their types and amount) **[L. 5–7**], the utilisation of regrinds **[L. 8, 9]**, and so on. The tribological and physical properties of plastics are, in many cases, improved just by additives – mainly by mineral particles (e.g. talc), graphite, metal powders and fibres, solid-state lubricants, and so on. Natural fibres´ (both vegetable and animal origin) influence on polymers tribological properties has not been widely published up to this time, because natural fibres are presently used as a reinforcement of shaped parts in automotive interiors. However, it is necessary to stress that such parts are practically produced just by the moulding of biocomposites not by their injection. Natural composite injection has not been a largely investigated area, but it has huge possibilities and potential in application.

PREPARATION AND INJECTION MOULDING OF COMPOSITE SAMPLES

A choice of polymer was necessary to take into account that the maximum temperature under which is possible to process natural fibres is 210°C **[L. 10]**. The composite material was prepared with PP matrixes (PP is the most common polymer used for the production composites in automobiles) added to natural fibres of coconut, flax, cellulose and fleece fibres **(Fig. 1)** within ratio 10% up to 30% by weight on the extrusion line of cold granulation at a temperature of 180°C to 200°C. We will refer to this as NF composite). These prepared granulates were prepared with an injection moulding machine as test samples for the subsequent evaluation of tribological and physical composite properties.

For the composite material preparation, it was necessary to remove fine organic impurities and greases and to modify their length by milling at the shear mill (n = 3000 min^{-1}) with trapezoidal holes of 0.75 mm (the optimal length of milled fibres is within 0.5 up to 2 mm) with minimal moisture content. For composite production, it was necessary to provide sufficient adhesion between the hydrophilous fibres and hydrophobic polypropylene matrixes. Because of this, in cooperation with company DOW Corning, a newly developed and patented additive *Smart* was used (1.5% on the basis of silicon + 0.2% on the basis of dialkyl peroxides) **[L. 11]**, whose components were added in the liquid form directly into the double-screw extruder together with the natural fibres and PP matrixes (Thermofil E020M).

FRICTION COEFFICIENT OF NATURAL FIBRE COMPOSITES

Presently, there is no valid international standard to determine plastic parts the friction coefficient of plastic plates with a thickness over 1 mm. Therefore, the measurement was carried out with deviations in part dimensions and the value of contact area normal force according to ISO 8295, i.e. the standard which specifies the methodology for the determination of the friction coefficient between sheeting and a thin plate (with thickness up to 0.5 mm). For experimental measurement, a device with a fixed horizontal test desk from anodised aluminium (Ra = 0.8 μm) and a moveable jig for placing testing samples (with dimensions 50 x 50 x 4 mm) and weight were used, so that the loading force gravity centre (F_N = 106.6 N) was always in the testing sample centre ensuring uniform pressure between planes **(Figs. 2 and 3)**.

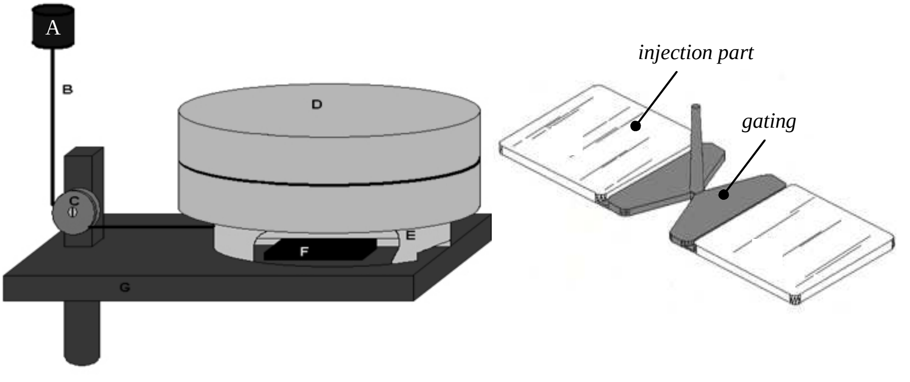

Fig. 2. Friction testing equipment concept (left) and testing sample (right)
Rys. 2. Koncepcja urządzenia do wyznaczania współczynnika tarcia (z lewej) i wypraska (z prawej)

The friction force was recorded during sliding movement of the solid at constant velocity (v = 100 mm/min, **[L. 3]**) in relation to displacement **(Fig. 4)**. The measured values were used to determine the static friction coefficient (from the maximum friction force value after the start of displacement) and the dynamic friction coefficient (determined from the average friction force after passing static friction at a displacement 60 mm then between displacements 10 p to 70 mm after the start of measurement). Static friction coefficient has a great influence on the values of the eject forces and injection part quality. High eject forces can result in the deformation of the injection part and high residual stresses in the injection part, and so on. Static and dynamic friction coefficient values are presented in **Tables 1** and **2**.

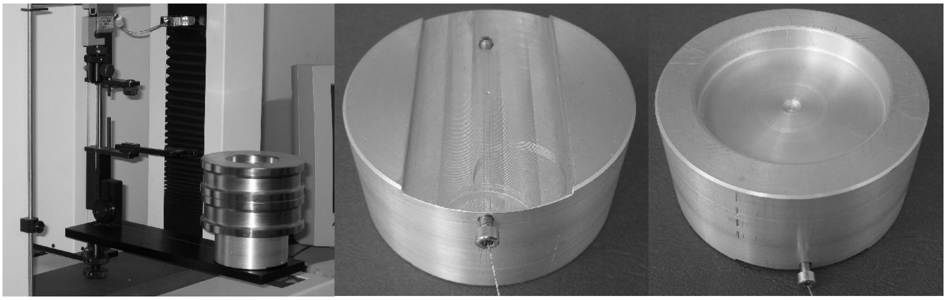

Fig. 3. Device for friction measurement with the detail of the jig design
Rys. 3. Urządzenia do pomiaru siły tarcia

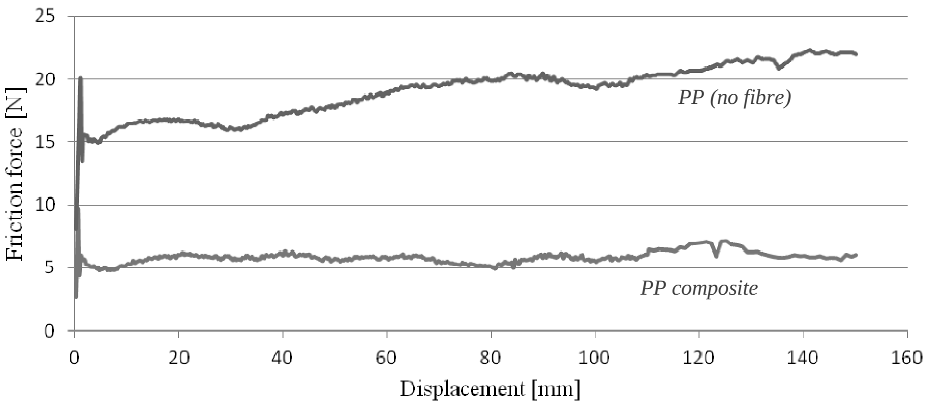

Fig. 4. Friction force of NF composite with coconut fibres
Rys. 4. Siła tarcia kompozytu z włókien kokosowych

Table 1. NF composites and static friction coefficient
Tabela 1. Statyczny współczynnik tarcia kompozytów z włókien naturalnych

Material		***Static friction coefficient "μ_S"***		
PP (no fibre)		0.20 ±*0.01*		
PP + Smart (no fibre)		0.11 ±*0.01*		
Natural fibre composite	***Fibre***	***10 wt. %***	***20 wt. %***	***30 wt. %***
	coconut	0.10 ±*0.01*	0.06 ±*0.01*	0.06 ±*0.01*
	flex	0.06 ±*0.01*	0.05 ±*0.01*	0.06 ±*0.01*
	cellulose	0.09 ±*0.01*	0.05 ±*0.00*	0.05 ±*0.00*
	fleece	0.11 ±*0.01*	0.08 ±*0.01*	0.08 ±*0.01*

Table 2. NF composites and dynamic friction coefficient
Tabela 2. Dynamiczny współczynnik tarcia kompozytów z włókien naturalnych

Material		***Dynamic friction coefficient "μ_D"***		
PP (no fibre)		0.17 ±*0.01*		
PP + Smart (no fibre)		0.06 ±*0.00*		
Natural fibre composite	***Fibre***	***10 wt. %***	***20 wt. %***	***30 wt. %***
	coconut	0.05 ±*0.01*	0.05 ±*0.00*	0.04 ±*0.01*
	flex	0.04 ±*0.01*	0.04 ±*0.00*	0.04 ±*0.00*
	cellulose	0.04 ±*0.01*	0.04 ±*0.00*	0.04 ±*0.00*
	fleece	0.08 ±*0.01*	0.04 ±*0.01*	0.06 ±*0.01*

SURFACE ROUGHNESS OF NF COMPOSITES

The composite material processed by injection moulding on small plates with the dimensions of 50 x 50 x 4 mm was subjected to surface roughness evaluation in relation to the type and amount of natural fibres in the polypropylene matrix. Three basic parameters were chosen for evaluation, which unambiguously determine surface parameters, i.e. Ra (arithmetical mean roughness), Rz (ten-point mean roughness) and Rt (maximum height of the profile). Experimentally measured surface roughness parameters using a Mitutoyo Surfest SV-2000 are presented in **Table 3**.

Table 3. Surface roughness of NF composites
Tabela 3. Chropowatość powierzchni kompozytów z włókien naturalnych

Material			***Ra*** *[μm]*	***Rz*** *[μm]*	***Rt*** *[μm]*
PP (no fibre)			0.5 ±*0.1*	3.3 ±*0.7*	5.0 ±*1.2*
PP + Smart (no fibre)			0.8 ±*0.2*	6.8 ±*1.9*	9.9 ±*2.7*
Natural fibre composite	***Fibre***	***wt. %***	***Ra*** *[μm]*	***Rz*** *[μm]*	***Rt*** *[μm]*
	coconut	*10*	1.4 ±*0.3*	10.8 ±*2.2*	16.8 ±*3.4*
		20	1.5 ±*0.6*	10.8 ±*2.5*	17.2 ±*5.3*
		30	1.6 ±*0.3*	11.9 ±*1.9*	18.0 ±*3.2*
	flax	*10*	2.1 ±*0.3*	14.6 ±*2.1*	19.6 ±*3.2*
		20	1.5 ±*0.3*	11.4 ±*2.5*	17.8 ±*4.9*
		30	2.0 ±*0.9*	14.1 ±*2.3*	19.4 ±*2.2*
	cellulose	*10*	1.6 ±*0.4*	11.5 ±*2.6*	16.4 ±*3.3*
		20	1.3 ±*0.4*	9.9 ±*2.0*	14.6 ±*4.4*
		30	2.1 ±*0.6*	13.9 ±*3.6*	20.1 ±*4.4*
	fleece	*10*	1.4 ±*0.3*	11.4 ±*2.2*	17.5 ±*3.6*
		20	1.4 ±*0.2*	9.9 ±*1.6*	14.6 ±*2.8*
		30	2.2 ±*0.5*	14.5 ±*2.9*	20.3 ±*4.3*

PHYSICAL PROPERTIES OF NF COMPOSITES

Physical (or more precisely physical-mechanical) and temperature properties of NF composites (**Tables 5** and **6**) were evaluated on parts produced by injection moulding according to ISO 294-1 and dimensionally corresponded to relevant international standards under the conditions presented in **Table 4**. There were also evaluated thermal properties on injection parts for automotive industry **(Fig. 5)** and these values are presented in **Table 7**.

Table 4. Testing conditions
Tabela 4. Warunki badania

Properties	*Symbol*	*Test standard*	*Conditions*
Yield tensile stress	σ_y	ISO 527/1A/50	50 mm/min
Tensile elongation at break	ε_B	ISO 527/1A/50	50 mm/min
Secant tensile modulus	E	ISO 527/1A/1	1 mm/min
Flexural stress	σ_{fM}	ISO 178	2 mm/min
Flexural modulus	E_{fM}	ISO 178	2 mm/min
Charpy notched impact strength	a_{CA}	ISO 179-1/1eA	2.9 m/s
Melt volume-flow rate	MVR	ISO 1133	180 °C/10 kg
Temp. of deflection under load	T_{ff}	ISO 75	1.8 MPa

Table 5. Physical properties of NF composites
Tabela 5. Fizyczne właściwości kompozytów z włókien naturalnych

Material			σ_y *[MPa]*	ε_B *[%]*	*E [MPa]*	σ_{fM} *[MPa]*	E_{fM} *[MPa]*
PP (no fibre)			27 ±*0.1*	39 ±*6.6*	1603 ±*33*	35 ±*0.2*	1287 ±*49*
PP + Smart (no fibre)			30 ±*0.4*	38 ±*2.9*	1419 ±*89*	39 ±*0.3*	1427 ±*38*
Natural fibre composite	*Fibre*	*wt. %*	σ_y *[MPa]*	ε_B *[%]*	*E [MPa]*	σ_{fM} *[MPa]*	E_{fM} *[MPa]*
	coconut	*10*	31 ±*0.2*	10 ±*0.6*	1763 ±*29*	44 ±*0.2*	1513 ±*152*
		20	31 ±*0.4*	10 ±*1.2*	1731 ±*36*	45 ±*0.1*	1630 ±*26*
		30	32 ±*0.5*	7 ±*0.7*	1985 ±*17*	49 ±*0.2*	1884 ±*33*
	flax	*10*	35 ±*0.5*	11 ±*1.2*	2304 ±*33*	48 ±*0.1*	1955 ±*71*
		20	36 ±*0.3*	9 ±*0.5*	2405 ±*32*	50 ±*0.3*	2219 ±*46*
		30	44 ±*0.3*	7 ±*0.2*	2944 ±*38*	60 ±*0.3*	2868 ±*81*
	cellulose	*10*	33 ±*0.2*	11 ±*1.6*	2073 ±*29*	47 ±*0.3*	1939 ±*58*
		20	34 ±*0.3*	11 ±*0.9*	2216 ±*28*	50 ±*0.1*	2184 ±*40*
		30	73 ±*0.3*	7 ±*1.1*	2863 ±*32*	58 ±*0.3*	2676 ±*45*
	fleece	*10*	33 ±*0.3*	12 ±*0.7*	1815 ±*32*	48 ±*0.4*	1704 ±*73*
		20	32 ±*0.3*	12 ±*1.2*	1596 ±*69*	48 ±*0.2*	1665 ±*50*
		30	37 ±*0.3*	8 ±*0.2*	1928 ±*24*	51 ±*0.2*	1710 ±*42*

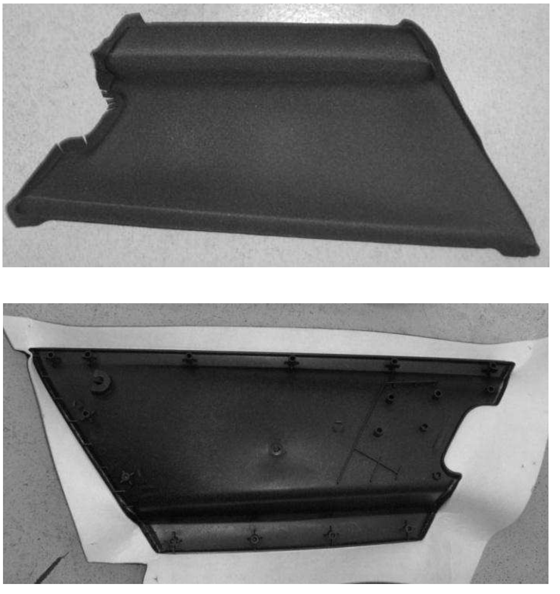

Fig. 5. NF composites injection part (In-mould)
Rys. 5. Wypraska kompozytowa

Table 6. Physical properties of NF composites
Tabela 6. Fizyczne właściwości kompozytów z włókien naturalnych

Material			a_{CA} [kJ/m^2]	*MVR* [cm^3/10 min]	T_{ff} [^{o}C]
PP (no fibre)			8.8 ±*0.6*	254.2 ±*7.6*	58 ±*2*
PP + Smart (no fibre)			10.1 ±*0.6*	21.0 ±*2.2*	63 ±*2*
Natural fibre composite	***Fibre***	***wt. %***	a_{CA} [kJ/m^2]	*MVR* [cm^3/10 min]	T_{ff} [^{o}C]
	coconut	*10*	5.6 ±*0.6*	16.7 ±*2.0*	66 ±*1*
		20	6.4 ±*0.2*	13.2 ±*1.2*	68 ±*2*
		30	5.4 ±*0.5*	9.4 ±*0.8*	72 ±*1*
	flax	*10*	5.7 ±*0.2*	7.6 ±*0.6*	70 ±*1*
		20	5.5 ±*0.2*	4.3 ±*0.6*	72 ±*2*
		30	6.2 ±*0.2*	3.2 ±*0.3*	80 ±*1*
	cellulose	*10*	4.9 ±*0.3*	5.3 ±*0.3*	69 ±*2*
		20	5.1 ±*0,3*	3.8 ±*0.4*	72 ±*1*
		30	5.1 ±*0.3*	1.6 ±*0.2*	82 ±*2*
	fleece	*10*	5.6 ±*0.2*	8.5 ±*0.5*	70 ±*1*
		20	5.5 ±*0.2*	7.5 ±*0.4*	72 ±*1*
		30	5.6 ±*0.3*	3.6 ±*0.4*	77 ±*2*

Table 7. Thermal properties of NF composites
Tabela 7. Właściwości termiczne kompozytów z włókien naturalnych

Test method	Measured value of the NF composites			
	coconut 20 wt.%	**flax** 20 wt.%	**fleece** 20 wt.%	**cellulose** 20 wt. %
Thermal resistance	Standard (desired state): without distortion			
- 30°C / 24 hod.	all right	all right	all right	all right
+90°C / 16 hod.	all right	all right	all right	all right
Flammability	Standard (desired state): ≤ 100 mm/min			
TL 1010	17.8	17.2	9.6	18.1

CONCLUSIONS

From the experimentally determination results concerning NF composites with polypropylene matrix tribological properties, it is possible to state that natural fibres of vegetable and animal origin have a positive influence on the final value of both the static and dynamic coefficients of friction **(Tables 1 and 2)**. The strong decrease in the friction coefficient was measured due to the addition of an adhesive agent whose individual components were added into polypropylene in the liquid form during the production of the granulates. The lowering of the friction coefficient was also achieved by adding natural fibres – mainly flax fibres. With an increasing portion of natural fibres, there was a decrease in the friction coefficient. The final decrease in the static friction coefficient had a positive influence on the phase of injection part removal from mould cavity during injection moulding and also on parts shape accuracy and the residual stress value in the injection part. It is possible to state that natural fibres increase surface roughness **(Table 3)** but there is no clear relationship to the percentage of natural fibres.

Natural fibres increase strength and stiffness of injection parts and decrease their ductility and melt flow rate **(Tables 5 and 6)**. The highest change of strength properties, but also shape stability under higher temperatures (measuring deflection temperature at loading – **Table 6**), was achieved for composite with cellulose fibres and flex fibres.

Regarding NF composite application in the branch of the automotive industry, it is possible to state that injection parts fulfilled the requirements concerning shape stability under lower and higher temperatures and fire safety according to VW requirements. However, for total evaluation of NF composites application in the automotive industry, it will be necessary to carry out other tests, e.g. environmental resistance, determination of puncture impact behaviour of rigid plastics, fogging test, emission test (TVOC), outdoor tests, and so on.

Initial results already predicate that the biggest problem will be just about the odour of injection parts from NF composites.

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Streszczenie

Niniejszy artykuł prezentuje właściwości tribologiczne i fizyczne analizy polipropylenu wypełnionego włóknami pochodzenia naturalnego, roślinnego i zwierzęcego (głównie kokosowe, włóknina, len i włókna celulozowe). Właściwości tribologiczne wtryskiwanego detalu są oceniane pod kątem chropowatości powierzchni oraz współczynnika tarcia. Właściwości fizyczne są oceniane pod kątem zachowania przepływu roztopionego materiału, cech wytrzymałościowych wtryskiwanej wypraski, palności i stabilności kształtu wypraski zarówno przy niskich, jak i w wysokich temperaturach.