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Utilization of air-plasma treated waste polyethylene terephthalate particles as a raw material for particleboard production



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1. Introduction

Polyethylene terephthalate (PET) has become the most favorable material for water and soft drinks bottles across the globe. Due to increased production in the last decades, the recycling of PET is of major concern [1]. The recycling is usually performed by a thermochemical procedure in which flakes are used as the input material [2]. In plastic industry, recycling is a common approach. For example, recycled low density polyethylene and aluminum of Tetra Pak were successfully used in manufacturing composite rigid boards, using a hot press [3]. This type of boards has found market niches in building industry [4,5] which may motivate utilization of PET plastic in a similar way. PET was utilized in particleboards in which PET flakes partially substituted wood in form of flakes [6] or dust [7]. Although this approach restricted the swelling of boards, the bending and internal bonding properties declined obviously. Internal bonding strength is a major quality criterion in boards [8], e.g. it determines the screw withdrawal strength [9]. Plasma treatment of composite components may help to improve the

ABSTRACT

Wood-plastic composite boards involving up to 30 wt. % of polyethylene terephthalate (PET) flakes were evaluated for its physical and mechanical properties – internal bonding strength, modulus of rupture and thickness swelling. It was found that the problem of the decline of mechanical properties can be successfully mitigated by air plasma pretreatment of PET flakes. The samples with 30 wt. % of plasma treated PET exhibited the same internal bonding strength as the control pure wood sample; samples with 15 wt. % of plasma treated PET exhibited the same modulus of rupture. The role of plasma treatment is to generate chemically active functional groups to PET flakes surface. This was verified by measuring the thermal chemiluminescence of plasma treated flakes.

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composite cohesive properties. In the case of wood composites plasma treatment increased the PVA-adhesion of particleboards and fiberboards [10]. This was followed by research of [11,12] where plasma treatment increased the quality of fibrous-plastic materials. Plastic components treated with plasma were successfully used for composites production [13,14].

In addition, plasma treatment of plastic fractions of composites was demonstrated profitable in more branches of material development. For instance, jute composite [15,16] or cement composites [17] were improved in this way. With respect to PET, the work [27] showed that oxygen containing plasma of both high and low pressure had positive effects on the surface wettability of PET films. This effect was due to the incorporation of polar functional groups of C-O and O-C=O to the film surface. A similar improvement of wettability of PET in its particulate form (i.e. chips, flakes) may be supposed; providing that a suitable plasma generator is chosen to facilitate an adequate contact between the plasma and complexshape PET flakes. Our work on this issue consisted of the following tasks: (1) production of particle boards with various PET admixtures by employing common particleboard production technologies in the laboratory; (2) utilization of the plasma treated PET flakes in particleboards in proportions of 15 and 30% of overall



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weight; (3) comparison of the properties of particleboards between untreated PET flakes and air plasma treated PET flakes.

2. Materials and methods

The research consisted of five phases: (1) preparation of PET particles by plasma activation by non-thermal plasma, (2) chemiluminescence and XPS analysis of PET flakes (3) preparation of particleboards with different replacement of wood particles by PET and (4) physical and mechanical evaluation of the particleboards according to Standards EN 310, EN 317 and EN 319, (5) scanning electron microscopy (SEM) evaluation of samples with PET particles and microscopic evaluation of ruptures in cohesive zones of PET flakes and wood particles.

2.1. Plasma treatment of particles

Plasma activation was done at atmospheric air pressure, using a Diffuse Coplanar Surface Barrier Discharge -DCSBD (Fig. 1). The DCSBD generates a thin layer (less than 0.3 mm) of non-thermal plasma over a flat dielectric plate made of 96% Al₂O₃, with the dimensions of 230 \times 95 mm. Plasma was ignited by 16 pairs of equidistantly spaced strip electrodes located at an opposite alumina plate, where the circulating transformer oil provided the electrical insulation and dissipation of heat originated in plasma. The width of an individual conductive strip was 1 mm, the distance between the strips was 1.5 mm. The system was powered by sinusoidal voltage of 10 kV_{RMS} at 15 kHz and 400 Watt input power, supplied by Lifetech VF700 power generator.

PET flakes batches of 5 g were spread over the DCSBD electrode surface and activated for 60 s, until the sufficient volume of material was obtained. The mass of 5 g allowed to spread flakes into a single layer to ensure good contact with the plasma generated for each individual PET flake (Fig. 2.). During the treatment, the flakes were slowly stirred with a flat paint brush to improve their overall contact with plasma. Routine check of wettability improvement was done by a floating test on water level. Owing to the surface tension around the flakes circumference, untreated PET flakes floated on the water surface. Plasma exposure lasting for 60 s was sufficient to lower surface tension to such extend that treated PET flakes sank to the bottom of the water beaker.

The extent of plasma created surface radicals was measured by thermally induced chemiluminescence of 50 mg PET flakes, using the "LUMIPOL 3" chemiluminometer [28] operated in inert N₂ atmosphere under the isothermal regime of 80 °C. The method is a well-established tool for assessing oxidative degradation of polymers induced e. g. thermally or by UV radiation [32]. We have used this method to monitor ongoing oxidative changes resulting from preceding plasma treatment. The measurement was used also for evaluating the plasma treatment aging effect.



Fig. 1. Scheme of plasma treatment set-up.

<u>20 mm</u>

Fig. 2. Plane view on PET flakes in contact with DCSBD plasma.

The chemical nature of surface changes was evaluated by ESCALAB 250Xi (Thermo Scientific) X-ray photoelectron spectrometer (XPS), equipped with microfocused Al K α monochromated x-ray source (1486.6 eV). The X-ray beam of 200 W with the diameter of 650 μ m was used. This X-ray spot size was found to be sufficient to compensate the natural chemical structure inhomogeneity of PET flake surface, so that the signal from various sites of the investigated surface was the same within the statistical error. Positive charge accumulated on the surface was neutralized by electron flood gun. High-resolution scans were acquired with pass energy of 50 eV and resolution of 0.05 eV. Spectra were referenced to the hydrocarbon type C 1s component set at a binding energy of 284.8 eV. The spectra calibration, processing and fitting routines were done using Avantage software.

2.2. Particleboards production

The tested particleboards were manufactured of spruce wooden particles supplied by a local particleboard producer. The average particle width was 2.25 mm, and the average length was 17.7 mm. The wood particle batches were supplemented with milled polyethylene terephthalate flakes (PET) of 1.6 mm average size. The wood particle moisture content was 5.24%, determined using a drying scale "Radwag Mac210".

PET was used in two variants: (1) untreated PET particles and (2) PET treated by air plasma (PLAS-PET). Urea formaldehyde adhesive "Prefere 4170" and hardener "Kronoadd HL 100" were used as a resin mixture. Firstly the wooden particles were mixed with resin, hardener and portion of distillated water to homogenise moisture content of wood particles batches on 11%. The mixing was done in a resinating drum for 10 min. Thereafter, the particles of PET were mixed separately for 10 min with the resin and hardener to ensure coating 8% (solid content) of weight. Finally particles of wood and PET were mixed in specified proportions (Table 1).

The prepared particle mixture was poured into boxes with bottom dimensions of 500 \times 500 mm^2 posed on the stainless steel

Table 1	
Experimental design	

Board type	Raw material	Raw material			
	PET (%)	PLAS-PET (%)	Wood (%)		
С	0	0	100		
C15	15	0	85		
C30	30	0	70		
P15	0	15	85		
P30	0	30	70		

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