# Characteristics of Wood Sawdust/Natural Rubber Composites Processed by Electron Beam Irradiation

**GABRIELA CRACIUN[1], ELENA MANAILA[1], MARIA DANIELA STELESCU[2]*, ANA-MARIA VASILESCU[2]**

[1] National Institute for Laser, Plasma and Radiation Physics, Electron Accelerators Laboratory, 409 Atomistilor Str., 077125, Magurele, Romania

[2] National R&D Institute for Textile and Leather – Leather and Footwear Research Institute, 93 Ion Minulescu Str., Bucharest, Romania

*This paper presents our experiments on obtaining and characterizing polymeric composites based on wood sawdust and natural rubber. Natural rubber was crosslinked both through a classic method – using benzoyl peroxide at high temperature, and an unconventional method at room temperature – electron beam irradiation. Physico-mechanical properties such as hardness, modulus at 100% elongation and tensile strength indicate a significant improvement as a result of adding wood sawdust to blends. Better results have been obtained using crosslinking by electron beam irradiation. The crosslinking rates of samples, measured using the Flory-Rehner equation increase as the amount of wood sawdust in blends increases and as the electron beam irradiation dose increases. The swelling parameters of samples significantly depend on the amount of wood sawdust in blends, because the latter has hydrophilic characteristics.*

*Key-words: natural rubber, wood sawdust, irradiation, physico–mechanical characteristics, crosslink density, swelling parameters*

Wood sawdust/polymer composites are of great interest because they offer several advantages over synthetic fiber composites such as: low density, improved acoustic properties, insulating properties, favourable processing properties (e.g. low wear on tools), occupational health benefits compared to glass fibers. Additional advantages include cost savings, dimensional stability, as well as positive effects on environment and agriculture with comparable mechanical properties [1, 2].

The polymers most often used in such composites are ethylene propylene diene rubber) polyethylene, polypropylenes, poly(vinyl chloride), and acrylonitrile-butadiene-styrene [3].

Sameni et al. [4] examined the mechanical and morphological properties of rubber wood fiber/reinforced thermoplastic natural rubber composites. Sombatsompop N et al. [1] studied the effects of wood sawdust (natural fiber) additions on the property changes of natural rubber containing the most important reinforcing filler, which is carbon black, where natural rubber was cured using the classic curing system namely sulfur and curing accelerators.

This paper studies the influence of wood sawdust amount and electron beam irradiation dose on physical-mechanical properties, crosslinking density rate and behaviour in aqueous environment of polymeric composites based on natural rubber and wood sawdust, where the elastomer was crosslinked by both electron beam irradiation and by means of peroxides.

## Experimental part

*Materials and methods*

All the raw materials: natural rubber (NR) Crep 1X (Mooney viscosity is 74 $ML_{1+4}$ at 100°C, 0.32% volatile materials content, 0.38% nitrogen content, 0.22% percentage of ash, 0.021% impurities content), antioxidant Irganox 1010, polyethylene glycol PEG 4000 (1.128 g/cm$^3$ density, 4-8°C melting point range), wood sawdust (dimension of max 1 mm) and dibenzoyl peroxide Perkadox 14-40B (1.60 g/cm$^3$ density, 3,8% active oxygen content, 40% peroxide content, *p*H 7) as vulcanizing agent for vulcanization at high temperature.

Sample preparation

Blends were prepared on an electrically heated laboratory roller. For preparation of polymeric composites, the blend constituents were added in the following sequence and amounts: 100 parts natural rubber (NR) roll binding (2'), embedding 3 phr (parts to 100 parts rubber) PEG 4000 and 1 phr Irganox 1010 antioxidant (2'), adding 5 and 15 phr wood sawdust respectively (2-4'), homogenisation of blends and removing from the roll in the form of sheet (4'). Process variables: temperature 25-50 ±5°C, friction 1:1.1, and total blending time 8-14'. Plates required for physico-mechanical tests with sizes of 150x150x2 mm$^3$ were obtained by pressing in a hydraulic press at 110±5°C and 150 MPa. Dibenzoyl peroxide vulcanized samples were prepared similarly to the experimental ones with the following specifications: 8 phr of dibenzoyl peroxide Perkadox 14-4B (2') as vulcanizing agent was added and the blend vulcanization was achieved in a hydraulic press at 160°C; the curing time was 19'.

Experimental installations and sample irradiation

The samples were packed in a polyethylene film and were irradiated at irradiation doses of 75, 150, 300 and 600 kGy respectively in the ALIN-10 electron beam accelerator in atmospheric conditions and at room temperature of 25°C. The ALIN-10 electron accelerator was built in Romania, National Institute for Lasers, Plasma and Radiation Physics, Electron Accelerator Laboratory-

* *email: dmstelescu@yahoo.com; Tel 040213235060*

Bucharest. The optimum values of the EB peak current $I_{EB}$ and EB energy $E_{EB}$ to produce maximum output power $P_{EB}$ for a fixed pulse duration $t_{EB}$ and repetition frequency $f_{EB}$ are as follows: $E_{EB}$ = 6.23 MeV; $I_{EB}$ = 75 mA; $P_{EB}$ = 164 W ($f_{EB}$ = 100 Hz, $t_{EB}$ = 3.5 μs). The dose from the ionizing radiation is the energy transferred to matter by ionizing radiation per unit mass of the irradiated material. The SI unit for exposure dose is joule per kilogram and it is called Gray (Gy). Any radiation – induced effect, which is reproducible and quantified, can be used as a dosimetric system [5].

Laboratory tests

The tensile properties of samples were determined using a Schopper tensile tester with a nominal rate of the traverse of the moving grip of 460 mm/min. The tensile strength was carried out according to the conditions described in ISO 37/2012, on dumbbell shaped specimens. The hardness, in units of Shore A was measured using a hardness tester according to ISO 7619-1/2011. The elasticity was evaluated with a Schob test apparatus using 6 mm thick samples according to ISO 4662/2009. Test specimens were cut off from plates of 150 x 150 x 2 mm by means of an automatic punching die.

The sol-gel analysis was performed on crosslinked NR rubber (with and without wood sawdust) to determine the mass fraction of insoluble NR (the network material resulting from network-forming crosslinking process) samples (gel fraction). The samples were swollen in toluene and extracted after 72 h in order to remove any scissioned fragments and unreacted materials. The networks were then dried in air for 6 days, and in an oven at 80°C for 3 hours, and reweighed. The gel fraction was calculated as:

$$Gelfraction = \frac{m_s}{m_i} x\ 100 \qquad (1)$$

where $m_s$ and $m_i$ are the weight of the dried sample after extraction and the weight of the sample before extraction, respectively [6-10].

The crosslink density (ν) of the samples was determined on the basis of equilibrium solvent-swelling measurements (in toluene at 23-25°C) by application of the well-known modified Flory-Rehner equation for tetra functional networks. The samples (2 mm thick) were initially weighed ($m_i$) and immersed in toluene for 72 h. The swollen samples were removed and cautiously dried to remove excess solvent before being weighed ($m_g$) and, during this operation, the samples being covered to avoid toluene evaporation during weighing. Traces of solvent and other small molecules were then eliminated by drying in air for 6 days and in an oven at 80°C for 3 hours. Finally, the samples were weighed for the last time ($m_s$), and volume fractions of polymer in the samples at equilibrium swelling $v_{2m}$ were determined from swelling ratio G as follows:

$$v_{2m} = \frac{1}{1+G} \qquad (2)$$

where:

$$G = \frac{m_g - m_s}{m_s} \times \frac{\rho_e}{\rho_s}\ ; \qquad (3)$$

$\rho_e$ and $\rho_s$ are the densities of elastomer samples and solvent (0.866 g/cm$^3$ for toluene), respectively.

The densities of elastomer samples were determined by hydrostatic weighing method, according to the SR ISO 2781/2010. By this method, the volume of a solid sample is determined by comparing the weight of the sample in air to the weight of the sample immersed in a liquid of known density. The volume of the sample is equal to the difference in the two weights divided by the density of the liquid.

The samples crosslink densities, ν, were determined from measurements in a solvent, using the Flory–Rehner relationship:

$$v = -\frac{Ln(1-v_{2m}) + v_{2m} + \chi_{12} v_{2m}^2}{V_1\left(v_{2m}^{1/3} - \frac{v_{2m}}{2}\right)} \qquad (4)$$

where $V_1$ is the molar volume of solvent (106.5 cm$^3$/mol for toluene), $v_{2m}$ is the volume fraction of polymer in the sample at equilibrium swelling, and $\chi_{12}$ is the Flory-Huggins polymer-solvent interaction term (the values of and $\chi_{12}$ are 0.393 for toluene [6-9].

*Water uptake test* - the effect of water absorption on fiber reinforced natural rubber composites are investigated in accordance with SR EN ISO 20344/2004. The samples were dried in an oven at 80°C for 2 hours and then are allowed to cool at room temperature in desiccators before weighing. Water absorption tests were conducted by immersing the samples in distilled water in beaker and kept at room temperature (23±2°C). After immersion the samples were taken out from the water at periodic intervals and the wet surfaces were quickly dried using a clean dry cloth or tissue paper. The moisture absorption was calculated by the weight difference. The percentage weight gain of the samples was measured at different time intervals. The water uptake was calculated as:

$$\text{Water uptake (\%)} = \frac{W_S - W_1}{W_1} \times 100 \qquad (5)$$

where, $W_S$ is the weight of the sample saturated with water, determined at periodic intervals and $W_1$ is the initial weight of the oven-dried specimen.

Rubber-filler interactions: the extent of interaction between rubber and filler (wood sawdust) can be analysed using Kraus equation. The Kraus theory and Kraus equation [11] have been successfully used by some researchers to assess the interfacial interaction in filler-reinforced rubber composites [12-16]. The Kraus equation is expressed as follows:

$$V_{ro}/V_{rf} = 1 - m\frac{f}{1-f} \qquad (6)$$

where $V_{ro}$ and $V_{rf}$ are the volume fractions of rubber in the gum vulcanizate and in fiber filled swollen sample, respectively, *f* the volume fraction of filler and *m* the filler polymer interaction parameter. The volume fraction of rubber in the swollen sample $V_{rf}$, was calculated by the expression:

$$V_{rf} = \frac{[(D - FT)/\rho_r]}{[(D - FT)/\rho_r] + [A_0/\rho_s]} \qquad (7)$$

where $\rho_r$ and $\rho_s$ are the densities of rubber samples and solvent (0.94-1.0 g/cm$^3$ for natural rubber and 0.866 g/cm$^3$ for toluene), respectively, *D* the deswollen weight of the test specimen (dry weight), *F* the weight fraction of the insoluble components, *T* the weight of the specimen and $A_0$ the weight of the absorbed solvent at equilibrium swelling.

## Results and discussions

*Physico–mechanical characteristics*

In table 1 are presented the results of physico–mechanical characteristics of samples crosslinked with peroxide. By introducing wood sawdust in rubber blends, it is noticed that as the amount of wood sawdust is increased, the following take place: (1) a significant increase in hardness, namely an increase of 5°ShA when introducing

**Table 1**
PHYSICO–MECHANICAL CHARACTERISTICS OF SAMPLES CROSSLINKED WITH PEROXIDE

| Characteristics/ Amount of wood sawdust | 0 phr | 5 phr | 15 phr |
|---|---|---|---|
| Hardness ( °ShA) | 45 | 50 | 52 |
| Elasticity (%) | 56 | 42 | 40 |
| Modulus at 100% elongation ($N/mm^2$) | 0.73 | 0.77 | 1.1 |
| Tensile strength, ($N/mm^2$) | 0.90 | 1.3 | 1.6 |
| Elongation at break (%) | 110 | 180 | 180 |

5 phr wood sawdust (from 45 to 50°ShA), when increasing the amount of wood sawdust at 15 phr, hardness is 52°ShA; (2) an increase in modulus at 100% elongation, tensile strength and elongation at break. These changes indicate that wood sawdust has a similar reinforcement effect to mineral fillers, at the same time improving their characteristics.

Physico–mechanical characteristics of samples crosslinked by electron beam irradiation are presented in Table 2. It is noticed that hardness increases with the increase of the irradiation dose and with the introduction of wood sawdust in natural rubber blends, because it leads to reinforcement of samples. The obtained values are lower than those of blends crosslinked with peroxides.

Elasticity varies irregularly when increasing the wood sawdust amount and with the increase of EB dose. The obtained values are comparable to those of blends crosslinked with peroxide.

Modulus at 100% elongation and tensile strength increase when increasing the irradiation dose and when introducing wood sawdust in natural rubber blends, and the obtained values are comparable to those of blends crosslinked with peroxides.

Elongation at break decreases with the increase of EB dose and with the increase of wood sawdust amount introduced in blends, indicating an increase in crosslink density. Obtained values are very high compared to those of blends crosslinked with peroxides.

*Gel fraction and crosslink density of the blends*

Table 3 shows the gel fraction (mass fraction of the network material resulting from a network-forming polymerization or crosslinking process; the gel fraction comprises a single molecule spanning the entire volume of the material sample) and cross-link density (number of crosslinks per unit volume in a polymer network) of the samples vulcanized by dibenzoyl peroxide, and EB respectively as a function of the wood sawdust content. The determination is based on the absorption of a proper solvent and subsequent swelling of the rubber [8-9]

Gel fraction value except for the natural rubber blend without sawdust irradiated with 75 kGy is over 90% for all blends and varies irregularly depending on the amount of wood sawdust in the blend, the crosslinking method or the irradiation dose.

It is noticed that, as the wood sawdust quantity increases, there is an increase of cross-link density ($\nu$) because sawdust acts as a filler in natural rubber blends and leads to reinforcement of blends. Crosslink density increases as the irradiation dose is increased as a result of a larger number of bonds forming between macro-molecular chains. Crosslink density of blends crosslinked with peroxide is higher than that of blends crosslinked with

**Table 2**
HARDNESS, ELASTICITY, 100 % ELASTIC MODULUS AND TENSILE STRENGTH OF SAMPLES VULCANIZED BY EB

| *Blend symbol* | *75 kGy* | *150 kGy* | *300 kGy* | *600 kGy* |
|---|---|---|---|---|
| | **Hardness, °ShA** | | | |
| NR (control) | 6 | 12 | 16 | 18 |
| NR+5phr sawdust | 10 | 14 | 20 | 24 |
| NR+15phr sawdust | 22 | 24 | 27 | 40 |
| | **Elasticity, %** | | | |
| NR (control) | 46 | 44 | 44 | 42 |
| NR+5phr sawdust | 44 | 48 | 46 | 44 |
| NR+15phr sawdust | 46 | 44 | 44 | 44 |
| | **100 % elastic modulus, $N/mm^2$** | | | |
| NR (control) | 0.01 | 0.02 | 0.02 | 0.04 |
| NR+5phr sawdust | 0.02 | 0.03 | 0.03 | 0.6 |
| NR+15phr sawdust | 0.5 | 0.5 | 0.6 | 0.9 |
| | **Tensile strength, $N/mm^2$** | | | |
| NR (control) | 0.01 | 1.02 | 1.06 | 3.2 |
| NR+5phr sawdust | 0.54 | 1.4 | 4.2 | 5.3 |
| NR+15phr sawdust | 1 | 2.4 | 4.4 | 4 |
| | **Elongation at break (%)** | | | |
| NR (control) | 760 | 900 | 900 | 667 |
| NR+5phr sawdust | 660 | 787 | 710 | 680 |
| NR+15phr sawdust | 420 | 627 | 670 | 660 |

electron beam. Analysing the results of physico-mechanical properties presented in table 1 and 2, it is noticed that hardness of blends crosslinked with peroxide has higher values than those crosslinked with EB, and elongation at break of blends crosslinked with EB has higher values than those crosslinked with peroxide, in accordance with values of crosslink density in table 3 and literature data [6-10, 17]. However, tensile strength of blends crosslinked with 150 kGy have comparable values to those of blends crosslinked with peroxide. This indicates that EB crosslinking can lead to materials with unique properties, which cannot be obtained through other methods. Materials with good tensile strength and elongation at break and also have low hardness and high elasticity values find applications in many economic areas [6-10, 17].

*Swelling parameters.*

The water uptake results of samples crosslinked with peroxide and by electron beam irradiation (with and without wood sawdust) are presented in figures 1-4. From these figures is observed that the percentage of water absorption in the composites NR/ wood sawdust depended on two parameters: wood sawdust content and irradiation dose. The water uptake increased with increasing sawdust content and irradiation dose. The increasing water absorption is due to the hydrophilic nature of sawdust and the greater interfacial area between the wood sawdust and the elastomer matrix [18-20]. In polymer composites with sawdust, water is absorbed mainly by the sawdust because the rubber material is hydrophobic and its water absorbability can be neglected [18-20].

*Rubber-fiber interactions*

The extent of interaction between rubber and fiber was analyzed using Kraus equation and the results are listed in table 4.

From the results in table 4, it is observed that the equilibrium solvent uptake of the samples decreased as

**Table 3**
GEL FRACTION AND CROSSLINK DENSITY OF SAMPLES

| *Sample* | G (%) (Gel fraction) | $v_{2m}$ (volume fractions of polymer in the samples at equilibrium swelling) | *v ($10^{-4}$ mol/cm$^3$) (cross-link density)* |
|---|---|---|---|
| NR+0phr sawdust / peroxide | 95.17 | 0.2463 | 2.3458 |
| NR+5 phr sawdust / peroxide | 96.30 | 0.2802 | 3.2200 |
| NR+15 phr sawdust / peroxide | 96.32 | 0.3144 | 4.3282 |
| | | | |
| NR+0phr sawdust /75 kGy | 36.24 | 0.0335 | 0.0403 |
| NR+5 phr sawdust /75 kGy | 91.70 | 0.0611 | 0.1033 |
| NR+15 phr sawdust /75 kGy | 90.61 | 0.0601 | 0.1193 |
| | | | |
| NR+0phr sawdust /150 kGy | 93.64 | 0.0877 | 0.2476 |
| NR+5 phr sawdust /150 kGy | 94.13 | 0.0912 | 0.2695 |
| NR+15 phr sawdust /150 kGy | 93.88 | 0.0997 | 0.3207 |
| | | | |
| NR+0phr sawdust / 300 kGy | 94.14 | 0.1164 | 0.4471 |
| NR+5 phr sawdust /300 kGy | 95.78 | 0.1372 | 0.6266 |
| NR+15 phr sawdust / 300 kGy | 96.05 | 0.1484 | 0.7348 |
| | | | |
| NR+0phr sawdust / 600 kGy | 95.92 | 0.2979 | 1.1076 |
| NR+5 phr sawdust / 600 kGy | 97.35 | 0.1928 | 1.3208 |
| NR+15 phr sawdust / 600 kGy | 96.48 | 0.1919 | 1.3033 |

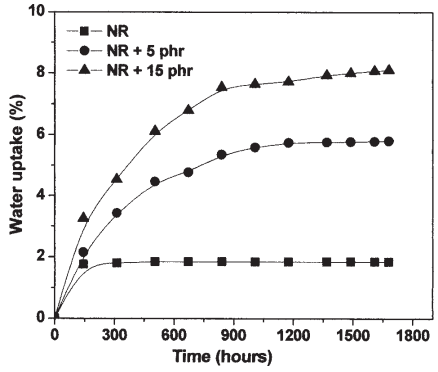


Fig. 1. Water uptake of polymeric composites based on natural rubber and wood sawdust, where the elastomer was crosslinked with peroxides

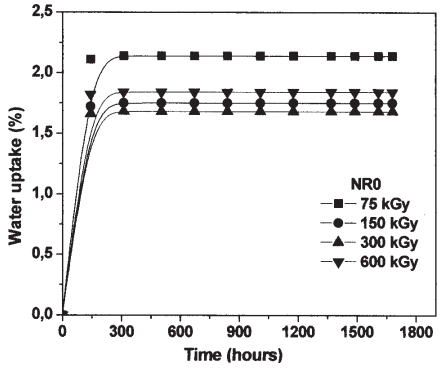


Fig. 2. Water uptake of (a) NR0/75kGy, (b) NR0/150 kGy; (c) NR0/300 kGy Mrad; (d) NR0/600 kGy

**Table 4**
$V_{rf}$ AND $V_{ro}/V_{rf}$ OF NR/ WOOD SAWDUST COMPOSITES IN TOLUENE

| *Sample* | $V_{rf}$ | $V_{ro}/V_{rf}$ |
|---|---|---|
| NR+5 phr sawdust / peroxide | 0.2912 | 0.8558 |
| NR+15 phr sawdust / peroxide | 0.3063 | 0.8137 |
| | | |
| NR+5 phr sawdust /75 kGy | 0.0583 | 4.2733 |
| NR+15 phr sawdust /75 kGy | 0.0599 | 4.1582 |
| | | |
| NR+5 phr sawdust /150 kGy | 0.0876 | 2.8434 |
| NR+15 phr sawdust /150 kGy | 0.0986 | 2.5284 |
| | | |
| NR+5 phr sawdust /300 kGy | 0.1304 | 1.9105 |
| NR+15 phr sawdust / 300 kGy | 0.1433 | 1.7385 |
| | | |
| NR+5 phr sawdust / 600 kGy | 0.1818 | 1.3706 |
| NR+15 phr sawdust / 600 kGy | 0.1836 | 1.3574 |

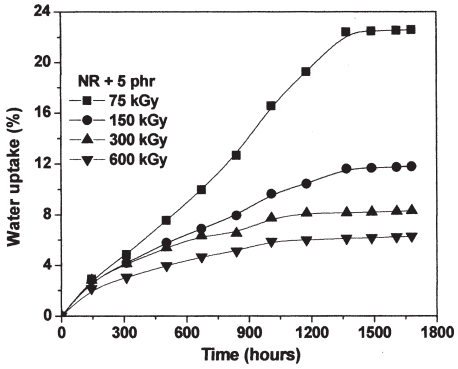


Fig. 3. Water uptake of (a) NR+5 phr wood sawdust /75kGy, (b) NR+5 phr wood sawdust /150 kGy; (c) NR+5 phr wood sawdust / 300 kGy Mrad; (d) NR+5 phr wood sawdust /600 kGy

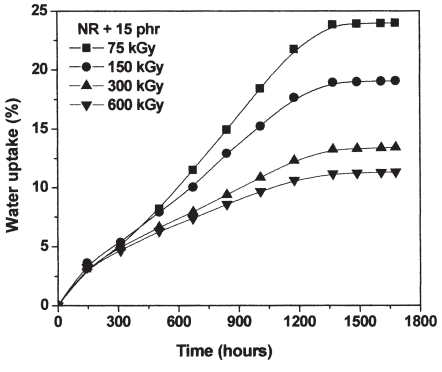


Fig. 4. Water uptake of (a) NR+15 phr wood sawdust /75kGy, (b) NR+15 phr wood sawdust /150 kGy; (c) NR+15 phr wood sawdust / 300 kGy Mrad; (d) NR+15 phr wood sawdust /600 kGy

the fiber content increased, which caused an increase in $V_{rf}$. So the ratio $V_{r0}/V_{rf}$ decreases since $V_{r0}$ is a constant. This is due to the increased hindrance exerted by the wood sawdust filler at higher loadings. The diffusion mechanism in the composite in strongly connected with the ability of rubber to provide pathways for the solvent to progress in the form of randomly generated voids. As the void formation decreases with filler content, the solvent uptake also decreases. The ratio $V_{r0}/V_{rf}$ is the degree of restriction of swelling of the rubber matrix due to the presence of fillers [11-16]. The more and more reduced values of $V_{r0}/V_{rf}$ ratio are associated with the enhanced adhesion between filler and rubber, according to the Kraus theory and Kraus equation. The decreased values of $V_{r0}/V_{rf}$ at higher loadings indicate the reinforcement effect of the wood sawdust [11-16].

## Conclusions

As a result of this study it is noticed that: (1) by introducing wood sawdust in natural rubber blends, an increase in hardness, modulus at 100% elongation and tensile strength of the blends occurs because they have the effect of reinforcing rubber blends; (2) with the increase of EB irradiation dose, an increase of hardness, modulus at 100% elongation and tensile strength of blends occurs because the crosslink density increases; (3) blends crosslinked by EB irradiation have shown higher values of elongation at break, lower values of hardness, and tensile strength of blends irradiated with over 150 kGy have higher values to those of blends crosslinked with peroxides. Gel fraction value except for the natural rubber blend without wood sawdust irradiated with 75 kGy is over 90% for all blends and varies irregularly depending on the amount of wood sawdust in the blend, the crosslinking method or the irradiation dose. The water uptake, swelling index and solubility increased with increasing fiber content. The increasing water absorption is due to the hydrophilic nature of fiber and the greater interfacial area between the fiber and the elastomer matrix.

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