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# ELECTRON-BEAM PROCESSING OF WOOD FIBRE REINFORCED POLYPROPYLENE

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## Abstract

High aspect ratio ($L/D > 100$) cellulosic fibres can be used as efficient reinforcement for different thermoplastics.

A procedure has been developed for wood fibre reinforced thermoplastic composites, applying a relatively small amount of reactive additive that compatibilize the synthetic and natural polymer components. Electron Beam (EB) treatment is a productive method of creating active sites on both matrix polymer and fibrous reinforcement, bound together through reactive additive.

The EB-processed, wood fibre reinforced polypropylene composite has not only a high modulus of elasticity, but also significantly higher flexural and tensile strength and improved thermal tolerance over the conventional wood fibre – polypropylene blends. The relatively low melt viscosity of our wood fibre reinforced polypropylene makes it easily processed not only by extrusion but also by injection molding. In the procedure recycled raw materials (e.g. fibres from recycled paper pulp or byproducts of the woodworking industry) can be used and the product is a fully recyclable composite.

The original procedure elaborated in Hungary was further developed and tried out in pilot scale at Canada. EB treatment, extrusion and thermoforming of the extruded sheets has been performed and applied to produce internal automobile parts, with the cooperation of Atomic Energy of Canada Ltd. (AECL) Whiteshell Research Laboratories. Technical feasibility and improved product quality have been demonstrated. The justification of high upfront investment costs of the EB processing line remained in discussion.

*Keywords:* polymer composites, fibre reinforced thermoplastics, Electron-Beam treatment.

## Introduction

The development of the fibre – reinforced polymer composites is moving recently toward the low cost composites [1]. High 'aspect ratio' ($L/D > 100$) cellulosic fibres are effectively used as reinforcement for different thermoplastics [2]. The *reactive* radiation processing, applying monomers and oligomers of high reactivity, and accelerated electron beam (EB) as energy source, offers unique methods to bind together high strength cellulosic fibres and thermoplastic synthetic matrix.

The promising results obtained with *preirradiated* wood fibre and polypropylene were published earlier [3]. A broader approach, applying different initiation methods has also been patented [4]. In the present work we report on the results of the last years of our investigations sponsored by the Atomic Energy of Canada Ltd. [5, 6]. In these works *mutual* (simultaneous) electron beam (EB) treatment has been applied to achieve

- *chemical bonds* between the components (wood fibre /WF/ and polypropylene /PP/) through
- *grafted* side chains and crosslinking bridges, built up using multifunctional, double bond containing reactive additives (RA) in
- *randomly* initiated chain reactions started by reactive centers created by EB.

The radiation response of the ingredients is well described in the literature. It has been demonstrated by KASHIWABARA and SEGUCHI [7] as well as by WILLIAMS [8] that the free radicals in PP are living long enough to count them as reactive centers even after days of irradiation. Similarly, the electron treatment createsreactive centers on the main components of the wood in different ways [9]. The multiple use of reactive oligomers in radiation chemical technology – as coating components, crosslinking agents or composite ingredients – has also been recently described [10].

## Methods, Materials, Equipment

The principle of the present technology is a kind of *reactive extrusion* [11] by which the composite ingredients mixed together and EB-treated (simultaneously) at room temperature, will activate their reactive centers mainly in the melt-extrusion procedure at high temperature. Actually, many of the polymer modificating procedures, e.g. industrial graft- copolymerization technologies are performed by such reactive extrusion.

In most of the present experiments the polypropylene PROFAX 6301 of HIMONT Canada (MFI = 19.2 g/10 min at 230 °C /2.16 kg) has been applied, originally in powder form. In a later phase the PP reference material of the Woodstock Co. (MFI = 2.78) has been used, similarly in 'barefoot, virgin' powder form, that is without any stabilizer or antioxidant.

For *wood fibre* – after surveying over 10 different types of wood and cellulose fibres, – first we selected the CF-3000-MW type purified (bleached) wood cellulose fibre from Central Fibre Corp. Canada, having the best aspect ratio (length/diameter) over 100. Later we applied unbleached, lignin containing wood fibres of slightly brownish color and much smaller aspect ratio (around 10).

The *reactive additive* (RA) applied in a relatively small proportion (about 1%) to make chemical bonds between the main constituents of the composite – is the key element in our composite processing. Its selection is based on several criteria.

The RA should have a *high reactivity:* easy entering into polymerization and crosslinking reaction of free-radical mechanism. The radiation-reactivity of the RA should be high enough to produce complete crosslinking with a dose between 8 to 25 kGy. The lower level (8 kGy) is dictated by practical point of view of (commercial) resin stability. An oligomer–monomer mixture which would polymerize much below that dose, could be unstable for storage: it could easily enter into spontaneous, thermal polymerization at room temperature. This is not only value-loss risk, it is also a safety question, because of the exotherm heat of polymerization. The upper limit of dose requirement is determined by the radiation sensitivity of the cellulose component of the wood fibre. The risk of some degradation in the molecular weight of cellulose over 16–20 kGy should be taken into consideration.

The *completely cured* RA as an EB-treated, hardened material, should be an impact-resistant, resilient and tough plastic. Regarding the chemical structure, multi-component copolymer system is advisable, having several types of chain segments linked together into an elastic network system. Different length of chain segments, 'spacers' are generally helping the elasticity. Reasonable impact strength is required with ductile failure mechanism. Tensile strength, modulus of elasticity, heat stability should also be considered.

All those features of the crosslinked RA could be different in bulk and in thin layer between the components of the composite. The best adhesives are not always producing good mechanical features in a bulk. Adhesion and elasticity as bridging function is the most important.

*Viscosity* of uncured RA is of importance for the easy distribution of a small amount of RA (generally 1%) to disperse evenly on a large amount of dry blend (of PP and WF). Solvents can be of great help, but may be unacceptable for local safety reasons. Spray-application may facilitate the homogeneous distribution of RA. The biggest factor, however, influencing the efficiency of the distribution of the RA resins is the very high activation energy of the viscosity. 20–30 °C increase in temperature decreases the viscosity of 10 times. A typical RA at 90 °C may have a viscosity 1000 times less than at room temperature.

The RA resin should show a satisfactory wetting on the ingredients of the composite. WF and PP are very different in polarity: PP is a-polar, while WF is highly polar and hygroscopic. Good wetting is based on adequate surface tension, depending on polarity relationships, viscosity, temperature, etc. We achieved good wetting with oligomer–monomer mixtures of polyesters and acrylates, proven for their good adhesion.

If the RA has an effect of *swelling* on the main components of the composite, it is possibly a benefit in anchoring the fibre into the matrix. The effect is expected to be positive, even if only one component of the RA is able to swell polymer constituents.

The *vapor pressure* of the oligomers, having a molecular weight over 1000 amu is almost negligible, so the oligomers are not volatile at all. The acrylic monomers have different volatility. The methyl methacrylate e.g. with its boiling temperature of 100 °C is practically useless as a component for RA as it would be easily volatilized at room temperature as well, and would certainly escape at the high melt-mixing temperatures of thermoplastics, typically over 170 °C. Even styrenes (boiling temp. 146 °C) is of limited suitability. Fortunately among the highly reactive di- and trifunctional acrylate monomers of medium molecular weight (200–500 amu) there are several of very low vapor pressure (below 0.02 Hgmm at room temp. and below 0.15 Hgmm even at 100 °C). Those monomers are practically non volatile and very suitable as components for RA.

The final bonding effect, based on the *reactive lubrication* is an overall, balanced result of all these earlier factors. Besides those technical criteria, we have to satisfy some equally important other ones, such as

- low skin irritation and other human safety aspects;
- fire hazard and other technological safety aspects;
- commercial availability, storage and transport capability;
- price/performance ratio.

*Table 1* shows the oligomers and monomers we used in the formulation of different reactive additives. *Fig. 1* shows the EB response of a typical RA formulation, as reflected in the exotherm reaction heat of a small (2.2 g) sample.

The I 10/1 electron accelerator of the AECL Whiteshell Laboratories used in our experiments as an EB source is basically a pilot-scale model of the factory scale IMPELA accelerators. The accelerating voltage of the machine in our experiments was 9.5 MV, the average beam current being 80 $\mu$A, the typical beam power was 0.76 kW. At those main parameters and the other standard conditions the output radiation dose is depending on the scanning width and the speed of conveyor which is transporting the target material under the electron beam. *Fig. 2* presents the dose in function of this conveyor speed. A typical dose of 10 kGy for the processing of WF–PP blends is delivered at 50 cm scan width at a conveyor speed of 0.25 cm/sec = 0.15 m/min. During the pilot scale processing, a typical bag filled with the dry blend of WF–PP compound, having a dimension of $45 \times 75 \times 7.5$ cm$^3$ was suitable for a homogeneous penetration and was

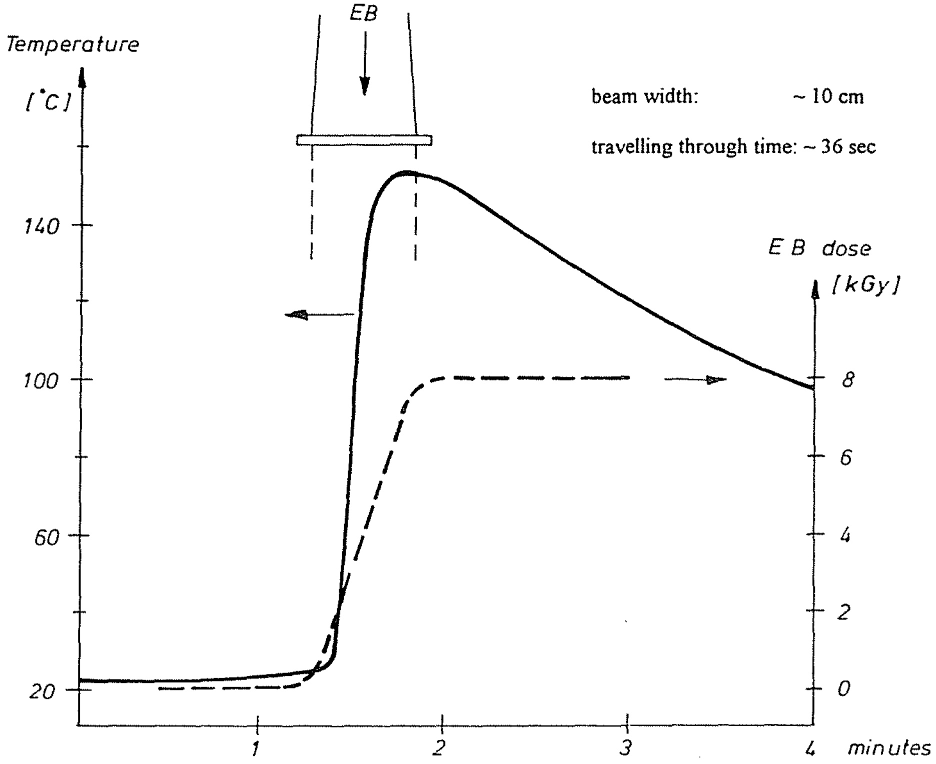

*Fig. 1.* EB curing of RA 451

irradiated typically in 5 minutes. The dose distribution in depth of our dry blend is shown on *Fig. 3*. The maximum output capacity of the I 10/1 in our case was over 100 kg/h of EB treated dry blend. A full scale IMPELA of 50 kW beam power could treat about 50 times more material (5 to/h, 20.000 ton/year) with such a dose.

The preparation of the *dry blend* of PP, WF and RA has been performed in a fluid mixer. The FM–4 mixer of Henschel Co. in our laboratory had 4 L of container capacity, with a maximum speed of rotors of 5000 r/m. At such a speed the expediently shaped rotors on the bottom of the vessel keep the dry components in a 'fluid state' and at the same time the friction between the granules is so high that the temperature is increasing rapidly (within about 5 minutes) above 80 °C without any external heat. The industrial scale mixing equipments are similar in construction and in function, yet they are 300 times bigger in size.

**Table 1**
Components of Reactive Additive formulations for WF–PP composites

| | |
|---|---|
| Oligomers used in Reactive Additive (RA) formulations | |
| C 111 Epoxidized soya oil acrylate | of SARTOMER Co. |
| C 114 Epoxy acrylate | of SARTOMER Co. |
| C 934 Aliphatic urethane acrylate oligoester | of SARTOMER Co. |
| C 964 Resilient urethane-ester acrylate | of SARTOMER Co. |
| C 965 Resilient urethane-ester (more flexible) | of SARTOMER Co. |
| C 9503 Resilient urethane-ester (earlier type) | of SARTOMER Co. |
| C 3000 Epoxy diacrylate | of SARTOMER Co. |
| C 5000 Polybutadiene diacrylate | of SARTOMER Co. |
| D 470–36 Vinylester type unsaturated polyester | of DOW Chem. Co. |
| D 470–60 Unsaturated polyester | of DOW Chem. Co. |
| Monomers used in Reactive Additive formulation | |
| S 238 Hexane diol diacrylate (HDDA) | of SARTOMER Co. |
| S 297 Butylene glycol dimethacrylate (BGDMA) | of SARTOMER Co. |
| S 306 Tripropylene glycol diacrylate (TPGDA) | of SARTOMER Co. |
| S 604 Dipropylene glycol (mono) methacrylate | of SARTOMER Co. |
| S 9040 Propoxylated neopentyl glycol diacrylate | of SARTOMER Co. |
| S 399 Pentaerythritol pentaacrylate | of SARTOMER Co. |

The *melt mixing* of the blend after the electron treatment occurred in a laboratory-scale twin-screw extruder, 'Prep-center' of BRABENDER Co. It has two intermeshing, counter-rotating screws of 25 mm diameter and a L/D (length-diameter) ratio of 20. The extruder has four different programmed heating zones, interchangeable, heated dices, electronic display of the variable screw speed, temperatures and internal pressure, and it is allotted with suitable follow-up equipment for making granules from the strands.

The *injection moulding* of the samples was performed on a conventional injection moulding machine of Battenfeld Co., having a maximum injection capacity of 350 g. A typical charge of 600–700 g of a composition was enough for 12–14 injections in our mould. The injection moulding temperature was 175 °C, the mould temperature was 41 °C, the typical cycle time 42 s.

The *mechanical properties* were measured on an MTS electronic testing machine with computerized data processing. For the dynamic mechanical analysis the DMA tester of Rheometrics Inc. (type RSA II.) was used in dynamic bending (dual cantilever) mode of operation. The complex modulus of elasticity ($E^*$), the storage modulus and the loss factor
(tan delta) were measured in a temperature range of $-50$ °C to $+150$ °C.

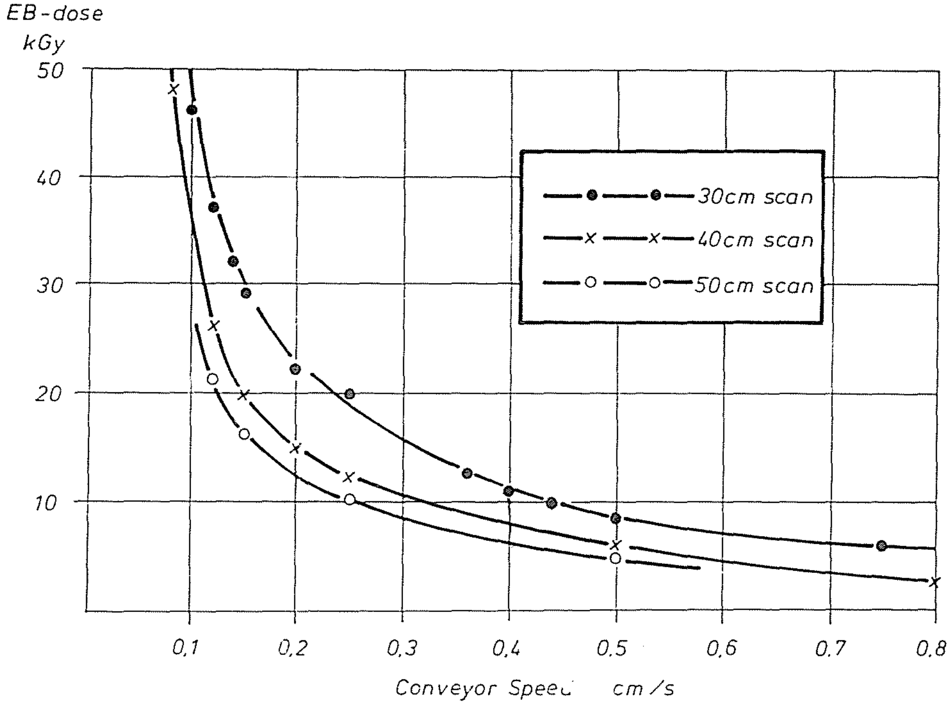

*Fig. 2.* EB dose delivered by the AECL's I 10/1 accelerator

*Table 2* shows the temperature values at the $E^* = 1$ GPa level, which gives a good comparison of heat tolerance.

## Results

The most detailed set of results comparing radiation processed and simply mixed WF – PP composites been published recently [6]. There we discussed the effect of mixing methods, of RA concentration, of the electron dose etc. *Table 2.* shows some of our WF–PP composites selected to demonstrate the adversely interconnected properties of flexural strength and impact strength. It is clearly seen on *Fig. 4* that if we want to increase the impact strength (notched IZOD) through adding Ethylene Propylene copolymer (XSV 954 of Himont) or EPDM rubber (NORDEL 2444 of DuPONT), flexural modulus and strength as well as tensile modulus and strength are decreasing. In fact the relationship is almost strictly inversely proportional, as it is seen on *Fig. 5.* We have to admit, however, that the chemical bonding between the fibrous reinforcement and synthetic matrix is well

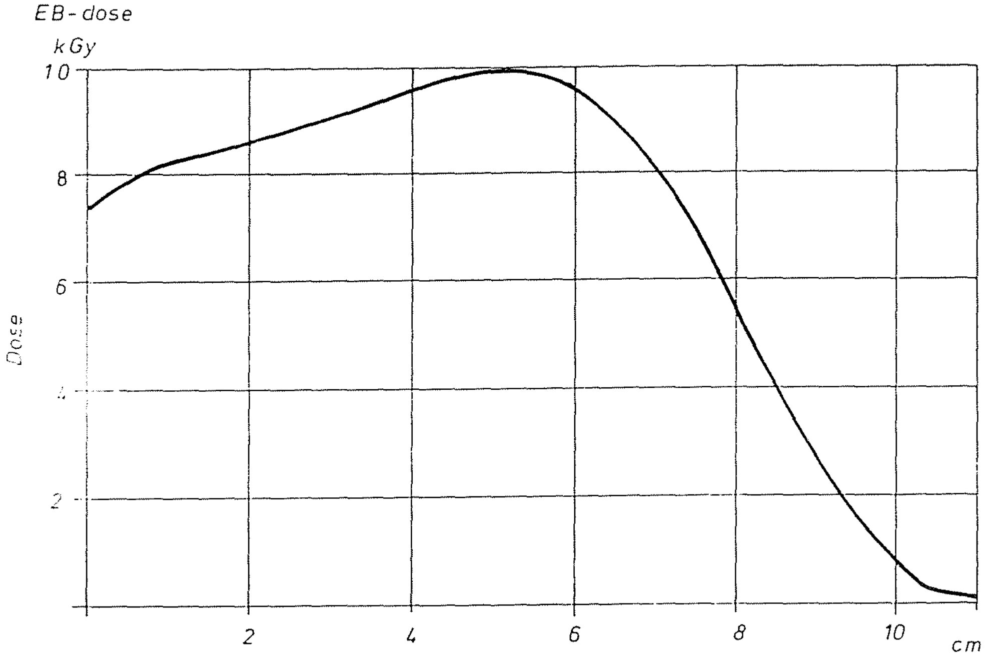

*Fig. 3.* EB dose distribution in depth in a 'dry blend' of 0.44 g/cm$^3$ density

reflected in the fact that not only the moduli, but also the flexural and tensile strength values are highly improved over the non-treated WF–PP blend.

A particularly interesting feature of the EB-treated WF–PP is the increased melt flow index (MFI), opening the way toward a high-speed plastics processing mode: the injection moulding. It seems to be a clear benefit over the non-irradiated WF–PP products existing on the plastics market, commercialized by such giant companies as GOR-Solvay, BASF, ICI, etc.

*Table 3* gives some details how MFI and the 'critical' notched impact strength are depending on the EB dose, in case of the PP alone, of WF–PP blend as well as of EB + RA processed WF–PP composites made according to our procedure with two different RA formulations.

The effect is marked on *Fig. 6.* The relatively low EB dose of 20 kGy is high enough to change the MFI of a PP powder by an order of magnitude (from 2.8 to 34.6). The effects of RA + EB compared to the effect of EB (without RA) on the WF–PP systems seem to be similar at first. There are some indications, however, that the lubricating effect of RA is dominating

**Table 2**
Composition and properties of EB-processed WF–PP composites

| Formula No. | 680 | 683 | 684 | 524 | 562 | 604 | 537 |
|---|---|---|---|---|---|---|---|
| Composition % | | | | | | | |
| PP | 100 | 65 | 65 | 58 | 52 | 52 | 33 |
| EP (XSV 945) | - | - | - | 7 | 13 | - | |
| EPDM | - | - | - | - | - | 13 | 32 |
| Wood fibre | - | 35 | 35 | 35 | 35 | 35 | 35 |
| Reactive addittive | - | - | 1.8 | 1.8 | 1.8 | 1.0 | 1.8 |
| EB dose, kGy | - | - | 5 | 10 | 10 | 8 | 10 |
| Properties: | | | | | | | |
| Flex. strength MPa | 37.8 | 44.0 | 62.5 | 46.3 | 47.3 | 28.6 | 21.7 |
| Flex. modulus GPa | 1.41 | 3.04 | 3.46 | 4.35 | 4.06 | 2.19 | 1.82 |
| Tens. strength MPa | 37.3 | 35.3 | 48.8 | 36.1 | 33.5 | 23.3 | 20.8 |
| Tens. modulus GPa | 1.87 | 3.93 | 3.89 | 3.79 | 3.68 | 2.58 | 1.05 |
| Impact strength (notched, IZOD) J/m | 12 | . . | 11 | 12 | 14 | 21 | 32 |
| Heat tolerance °C (at $E^x = 1$ GPa) | 51 | . . | 142 | 102 | 111 | . . | 63 |
| MFI (2.16/230 °) g/10 mm | 20 | $\sim 1.0$ | 4.4 | 4.2 | 2.9 | 1.5 | 0.2 |

at low doses. It is precisely the dose range (8–10 kGy) where our WF–PP composites are efficiently processed. At higher dose (20 kGy) the RA may have some slight benefit in 'healing' the wounds, compensating the radiation-caused degradation on the PP.

*Table 3* shows that the effect of EB is as critical on the MFI as on the impact strength. The brittleness of the WF–PP composite may arise from the increased dose. This may be related to a higher crosslink-density as well, however, the excess radiation damage of the PP less protected in powder form (which is required for an efficient mixing with WF) is the most probable cause of this effect.

## Discussion of Results, Conclusions

The electron-processing or EB-treatment is a highly efficient method of creating chemically active sites on all the components of the wood-fibre – polypropylene composite system. Long living free radicals and thermally activable peroxy groups are accumulated on the cellulose as well as on the lignin after radiation treatment. The polypropylene is an excellent matrix for radiation-initiated graft copolymerization because of formation

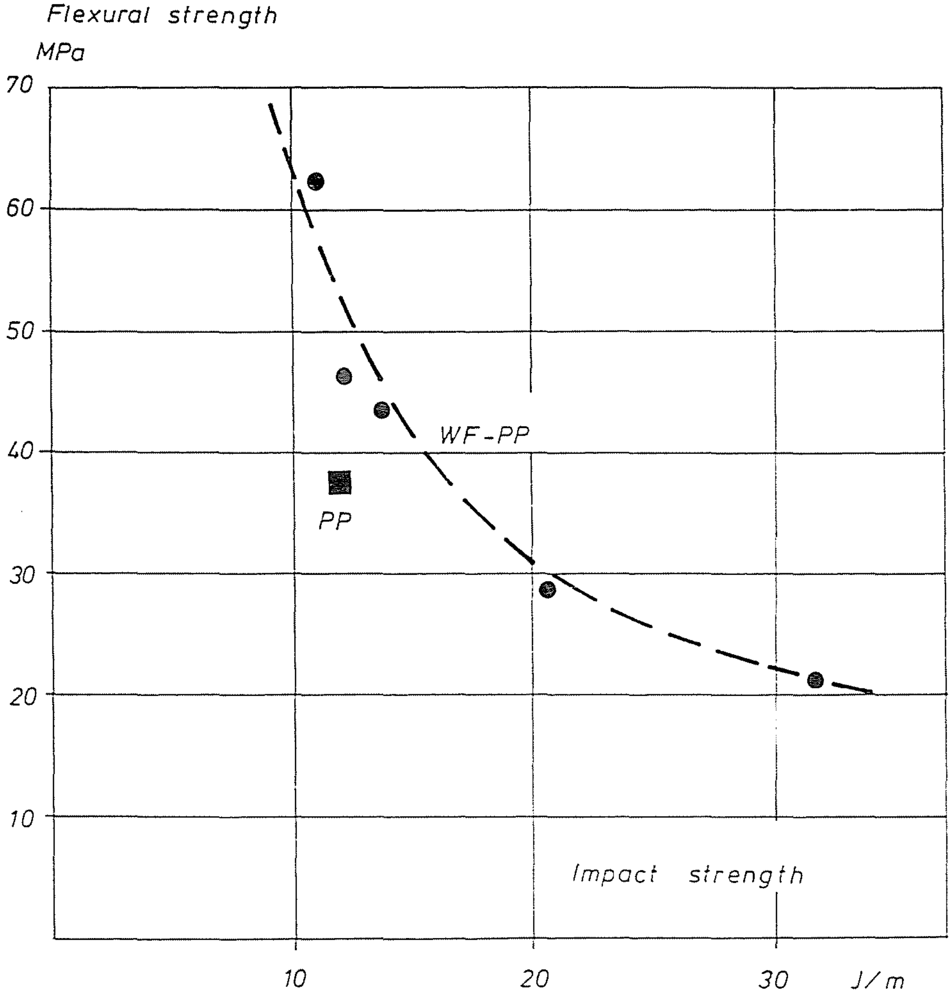

*Fig. 4.* Flexural- and impact (notched, IZOD) strength of the WF - PP composites (PP: HIMONT Profax 6301)

of similar free radicals and peroxy groups. These chemically reactive species can serve as bonding sites for suitably selected reactive additives, capable of high-speed polymerization chain reactions.

The role of the reactive additive is multiple. The benefits of such additive can be observed in all the major steps of composite process-

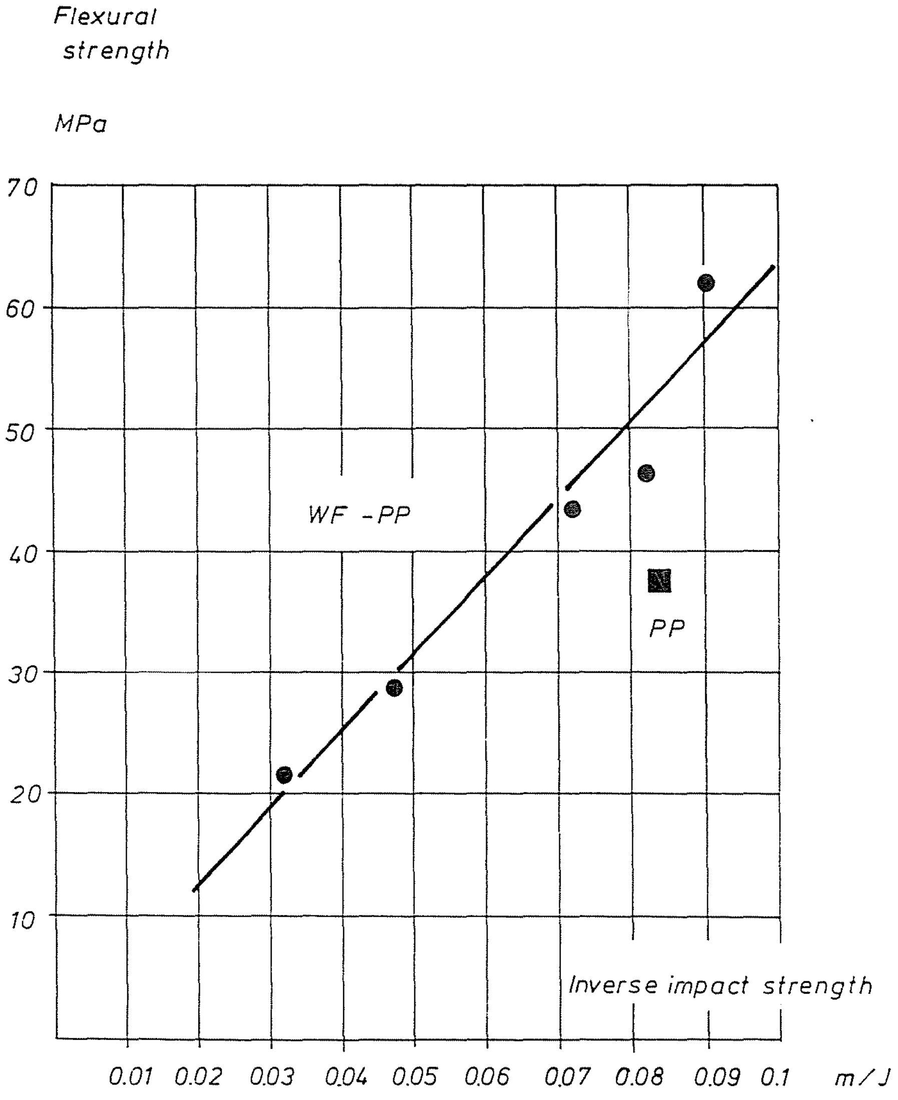

*Fig. 5.* Flexural strength of the WF - PP composites in function of the inverse impact strength

ing as well as in the properties of the final product. The benefits during the composite manufacturing are clearly surface-tension related. The suitably selected oligomer–monomer mixture of optimum viscosity and polarity helps achieving a better dispersion of fibres through a better wetting. Dur-

**Table 3**
Effect of the electron beam (EB) dose on the melt-flow rate and impact strength of PP and WF–PP composites

| | | EB-dose (kGy) | | | |
|---|---|---|---|---|---|
| | | 0 | 9 | 13 | 20 |
| PP powder 'Woodstock' (barefoot, virgin) | MFI | 2.78 | 16.8 | 25.9 | 34.6 |
| | Impact strength: | 38.1 | 25.7 | 29.0 | 31.9 |
| Mixture of WF and PP (50/50) (no RA added) | MFI | 0.41 | 0.62 | 1.36 | 2.66 |
| | Impact strength: | 25.3 | 18.4 | 17.7 | 20.5 |
| EB-treated WF–PP (50/50) (with 1% RA 416) | MFI | 0.61 | 1.40 | 1.62 | 2.48 |
| | Impact strength: | 28.3 | 20.0 | 18.4 | 17.0 |
| EB-treated WF–PP (50/50) (with 1% RA 451) | MFI | 0.41 | 0.93 | 1.66 | 2.23 |
| | Impact strength: | 23.3 | 17.4 | – | 16.2 |

MFI measured at 230 °C, 2.16 kg load, in g/10 min unit
Impact strength: notched Izod, J/m

ing the early stage of melt mixing, the still unreacted additive may act as a lubricant. However, the most important benefits are reflected in the enhanced properties of the final composite.

Our present understanding concerning the mechanism working in the composite processing in the *mutual* graft copolymerization processing mode is the following.

– During the first step, the *dry blending* of the powder-like components, the RA acts as an efficient wetting agent, coating the surface of the fibrous as well as of the particulate components of the future composites. A partial swelling of both main components may occur, since both PP and WF are easily penetrable to the low molecular components of the RA.

The *EB treatment* of the dry blend at room temperature produces frozen-in free radicals and peroxy groups on all the components. The free radicals could initiate the chain reaction of the RA immediately. However, probably most of them remain temporarily unreacted, firstly because of the free radicals in the crystalline part (more than 65%) or the PP remains frozed-in and totally inaccessible below the crystalline melting point (165 °C). The peroxy (and hydroperoxy) groups remain also unreacted at room temperature, because of their high activation energy of dissociation (over 30 kcal/mol).

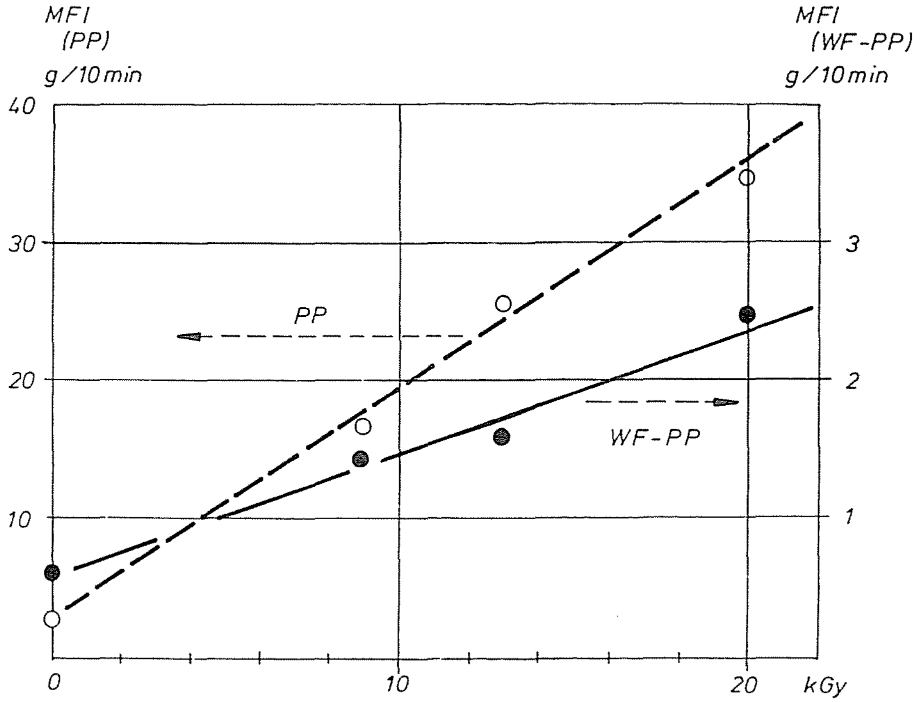

*Fig. 6.* The effect of EB dose on the melt-flow index (MFI) of PP and WF - PP composites

The main reaction between the polypropylene, reactive additive and wood fibre occurs mostly in the subsequent *melt-mixing* step, in the extruder. At the early stage, in the first zone of the screw the unreacted RA acts as a lubricant, helping the fusion of the PP particles at relatively low temperature (175 °C) and short residence time (in about 150 s), saving very much the heat-sensitive WF. This temperature and time nevertheless is more than enough to activate most of the peroxy groups and almost all the frozen-in free radicals. So, most probably the melt-mixing is the moment, and the extruder is the place, when and where the grafting and/or graft-crosslinking is accomplished.

The main features of this mechanism are well reflected in the physical and mechanical properties of the final composite. The increased homogeneity of the product is more difficult to measure, but is unquestionably observed.

The grafting and (partial) crosslinking through RA bridges between WF and PP may influence negatively the impact strength. As it is well known [12], the capacity of absorbing the elastic/impact/energy is inversely related to the modulus of elasticity. To achieve an optimum crosslink density of suitably elastic 'vulcanizing' bridges, before entering into the stage of 'bakelite' type of rigidity, requires a fine tuned balance of many parameters. The whole industry of reinforced (filled, blended) plastics is based on that type of balanced optimization of interactions. The ways to overcome the problems of possible reductions of impact strength are multiple:

- selecting adequate matrix (co)polymer of suitably high impact strength;
- formulating RA of appropriate elasticity;
- incorporating some more elastic fibrous component along with the WF;
- incorporating shock-absorber, heterophase elements into the composite, as in the manufacturing practice of high-impact thermoplastics, such as high-impact polystyrene, polyamide, etc. for stopping the propagation of micro-cracks (craze) in the moment of impact. It is known that many of those high-impact polymers are manufactured in well controlled partial graft copolymerization reactions (e.g. ABS copolymers, acrylic type PVC modifiers).

The function of the graft reactions is recognized in several recent works on wood fibre reinforced thermoplastics [2]. The advantageous pretreatment of WF with reactive isocyanates or silanes before incorporating into the composite is based on grafting reactions. The substantial advantage of the EB-initiated technology is that this procedure is *integrating* the graft-crosslinking reaction of the suitable additives into the melt-mixing step of thermoplastic *processing*. Our composite-making procedure is a sort of reactive extrusion, where part of the chemical processing occurs in the extruder, without any additional heat-expenditure.

The favorable effect of crosslinking agents, such as benzoyl peroxyde or dicumyl peroxyde in WF-thermoplastic composites was also described recently [2]. The EB crosslinking seems to offer some particular benefits in composite making. The EB treatment involves all the ingredients in the transformation, as it is penetrating in-depth, in all the components of the (future) composite, which is hardly possible for the chemical initiators.

The EB-processing of WF–PP composites has been tried out on *pilot scale* as well, in a cooperation of the Atomic Energy of Canada Ltd. (AECL) Whiteshell Laboratories and the American Woodstock Co., Sheboygan, Wisconsin, USA. Dry blend mixing of WF/PP (with RA) has been performed on factory scale equipments, producing altogether 1360 kg experimental material. EB treatment has been made on the I 10/1 EB-machine at Whiteshell, Canada. Extrusion and thermoforming of the composite sheets has been performed at Sheboygan, USA. Experimental interior trim parts have been made successfully for different models of Ford, Chevrolet and Cadillac cars.

Technical feasibility and improved product quality have been demonstrated. Improvement was observed in extrudability (with less energy consumption) in thermoformability (lower temperature) and in the fact that the EB-treated WF–PP was suitable for injection moulding while the earlier WF–PP products were not. Impact strength was somewhat (20%) decreased but flexural- and tensile strength were increased by 20% and the thermal tolerance of the product was more than 10 °C improved. The justification of the high upfront investment cost of a full-scale EB processing line remained undecided.

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