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Rubber Powder – A Perspective Filler of Thermoplastics

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Summary: Rubber powder, ground at ambient temperature, was compounded with polypropylene using only 16 L/D of a co-rotating twin screw extruder. An optimum of extruder configuration and selected process parameters were found for compounds with a rubber filling rate of 50 wt.-%. Restrictions of a reactive compounding were taken into account. The dispersion of the rubber powder is not a limiting process. A compound containing 70 wt.-% rubber powder showed to be overfilled.

Keywords: compounding; particle size distribution; polypropylene; recycling; rubber

Introduction

The amount of used tyres is 600 000 tons/a only in Germany. They are mostly burnt in energetic recycling processes e.g. for cement production. A higher level of environmental protection would be an use as material. Modern techniques give the possibility to grind the rubber into particles in micrometer-scale. Such rubber powder can be an interesting filler for plastics in account to its elasticity.

An only physical compounding of rubber powder with different thermoplastics results in products with a low or intermediate mechanical behaviour. This is due to the low interaction between the surface of the cross-linked rubber particles and the polymer matrix resulting in low adhesion. A chemical bonding between the surface particles and the matrix is necessary for a substantial improvement in mechanical properties. As the rubber surface particles contain residual unsaturated double carbon-carbon bonds, a grafting of the matrix polymer onto the rubber surface initiated by radical donators, e.g. peroxides, could improve the rubber-polymer interaction.

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A process of reactive compounding of rubber powder with polypropylene in an internal mixer has been developed[1,2,3]. A variety of organic alkyl- and arylperoxides, e.g. di(tert-butyl) peroxide (DTBP) or dicumyl peroxide (DCUP), were used as radical donators for the development of the discontinuous reactive compounding process in the internal mixer. It is well known that polypropylene undergoes β-scission in the presence of radicals, in contrast to polyethylene, which undergoes a cross-linking reaction. Therefore, the additional use of the so called radical catchers or radical acceptors, such as phenolics or molecular sulphur, could prevent the unwanted scission of the polypropylene backbone. A good level of mechanical properties of the reactive compounds could also be achieved even without adding radical catchers, if the rubber particles act as free radical scavengers.

Figures 1 and 2 show the difference in the tensile strength and elongation at break obtained from stress-strain tests according to ISO 527 - standard for physical and reactive compounds (so called elastomeric alloys).

The reaction time is one of the main process parameters in case of reactive compounding. The control of the reaction time is quite easy by using an internal mixer. However, the processing costs using an internal mixer are high which leads to a low competitiveness caused by price of the products. That's why the final aim of this work is the design of a continuous compounding process for reactive polypropylene- rubber powder compounds.

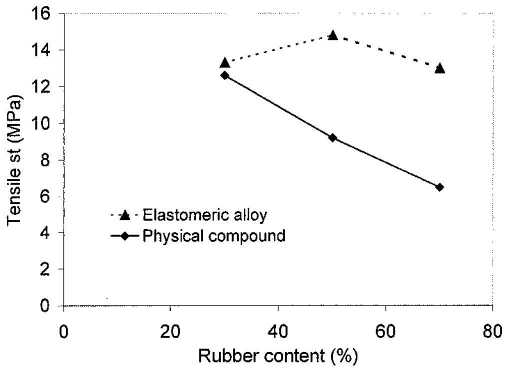


Figure 1. Tensile strength vs. rubber content of physical and reactive compounds.

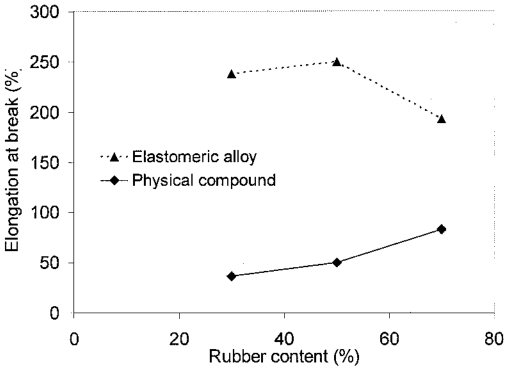


Figure 2. Elongation at break vs. rubber content of physical compounds and reactive compounds.

Experimental

Two main problems have to be solved for the design of a continuous reactive compounding process of PP with rubber powder:

- Dispersion and distribution of rubber powder in a PP-melt
- Reaction of PP with rubber powder, set off by a peroxide activator

The experiments described below are related to the powder dispersion and distribution in a twin screw extruder.

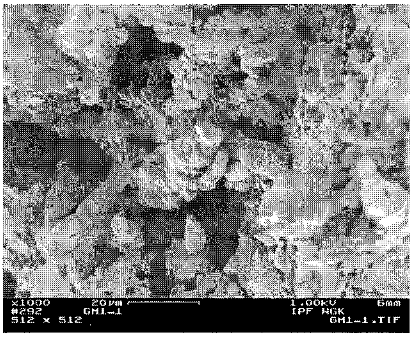


Figure 3. SEM micrograph of the rubber powder.

A rubber powder obtained from tread layers truck tyres by grinding at ambient temperature was used. The structure of the powder is shown in the SEM-micrograph of Figure 3 and the particle size distribution obtained by laser diffraction is given in Figure 4. A polypropylene-ethylene-copolymer with a melt flow rate of 8 g/10 min (190 °C/2.16 kg) and a comparatively low melting point of 139 °C was used as the thermoplastic matrix.

For the experiments a Micro 27 made by Leistritz, Nuremberg in Germany, was used as the compounding unit. This unit is a tightly intermeshing co-rotating twin screw extruder with a screw diameter of 27 mm and a variable L/D ratio from 24 up to 44. The long variant of L/D 44 will be used for the complete reactive compounding process. The L/D ratio of 24 was used in this work for the investigation of dispersion and distribution of rubber powder without the reactive processing step.

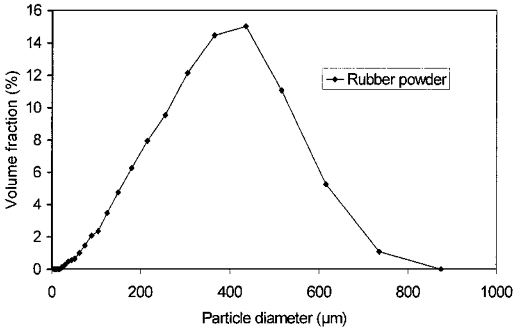


Figure 4. Particle size distribution of the rubber powder.

Both components were fed separately by gravimetric dosage units into the hopper. The fourth barrel element of the extruder (approximately 14 L/D) was closed with a closure part. This barrel element could be opened to take samples of the PP-rubber powder-melt at this place. After the process parameters were balanced, the extruder was stopped for taking samples.

The process was varied in relation to:

- Screw design (length of the kneading zone 60 and 30 mm, Figures 5 and 6)
- Screw speed (200 and 100 rpm)
- Temperature profile of the barrel T_{bmax} (2 different profiles)
- Feed rate (6 and 12 kg/h)
- Rubber powder content (50 and 70 wt.-%)

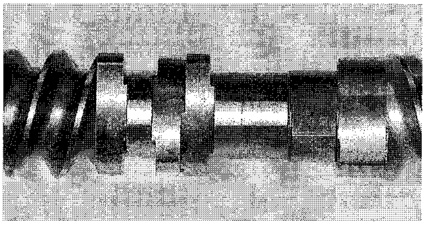


Figure 5. Kneading zone for intense kneading.

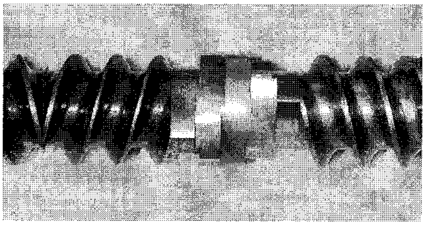


Figure 6. Kneading zone for medium kneading.

After taken the samples out of the extruder they were pressed to sheets of approximately 5 mm thick. Small pieces of the sheet were cut from one side with a cryo-microtom. The flat surfaces were partially imaged using a low voltage scanning microscope DSM 982 GEMINI Carl Zeiss. The low voltage option of this microscope made it possible to work with untreated samples. A thin gold film on the surface, usually necessary for polymers at standard voltages, leads to a very poor contrast between rubber particles and matrix polymer.

The micrographs were analysed computer aided using a special image analysing software (Analysis). Five different micrographs were used for every sample. The particle size distribution of the rubber powder before compounding was obtained using a laser diffraction sizer (Sympatec).

Results and Discussion

Four main parameters were selected to evaluate the different process parameters according to the final aim – design of a reactive compounding process:

- **Feeding/conveying behaviour:**

  The feeding of the voluminous rubber powder together with the polymer pellets in the main hopper can result in conveying problems. This variant of feeding was chosen to keep the extruder length which is necessary for physical incorporating of the rubber powder as short as possible.

- **Mass temperature**

  A small temperature window is caused by the melt temperature of polypropylene and the limiting temperature of the activator system for the reactive compounding.

- **Torque reserve**

  The reactive compounding results in an increase of melt viscosity. Therefore, the physical compounding should consume only a part of the maximum torque.

- **Dispersion of rubber powder**

  The evaluation of dispersion effectiveness using the particle size distribution in the compounds made no sense. All particle size distribution curves of the compounds were very similar, as shown in Figure 7. It was observed that none of the compounds exhibit a completely dispersed morphology, i.e. each compound exhibits a certain amount of contact points between the rubber particles. Contacts between particles will be weak zones in the final products with respect to the mechanical properties. That's why the number of observed contacts per area was used for the evaluation of the dispersion success. Such contacts can be observed in Figures 8 and 9.

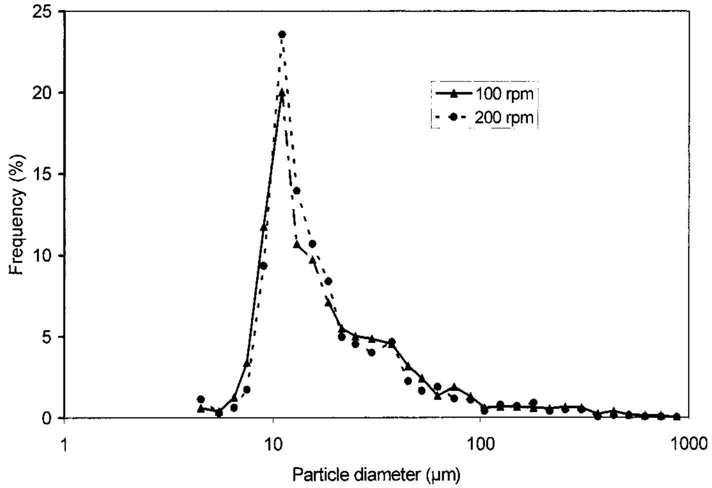


Figure 7. Examples of particle size distribution after compounding at different screw speeds (filling rate 50 wt-%, medium kneading, low barrel temperature).

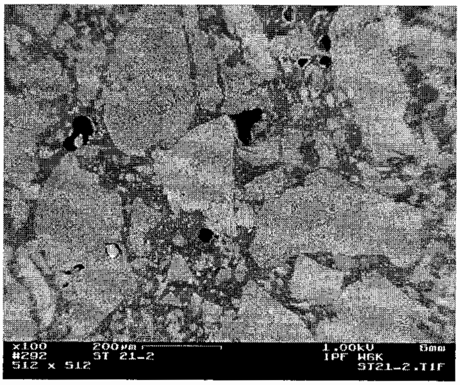


Figure 8. SEM micrograph of a compound containing 70 wt.-% of rubber powder.

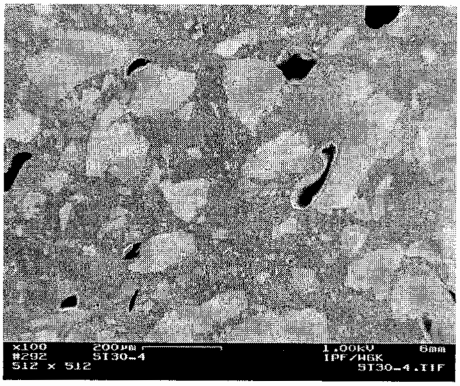
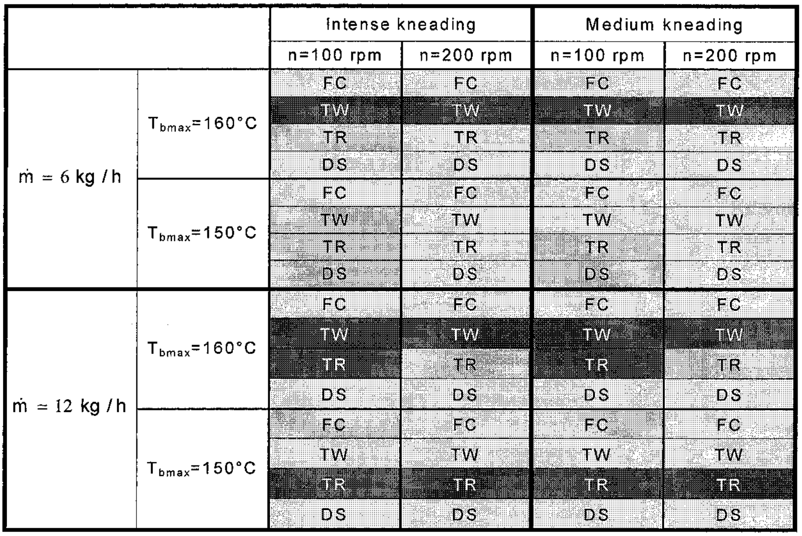


Figure 9. SEM micrograph of a compound containing 50 wt.-% of rubber powder.

Feeding/conveying behaviour (FC)	easy flow	unstable flow	tailback
Temperature window (activator system) (TW)	160 – 165 °C	166 – 170 °C	> 170°C
Torque reserve for reactive compounding (TR)	> 60%	50 – 60%	< 60%
Dispersion of rubber powder (DS)	< 20 contact points/cm²	> 20 contact points/cm²	Agglomerates > 10 particles

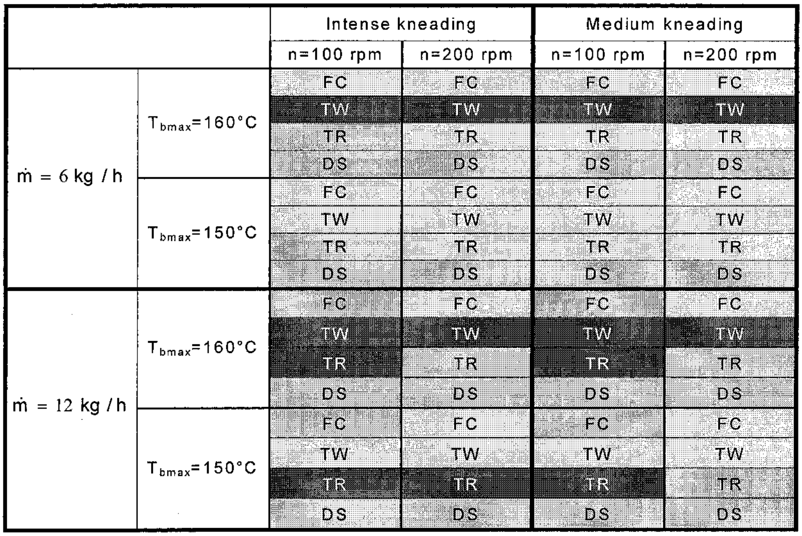
Figure 10. Classification of results.

For a better illustration of the influence of the four main parameters on the extrusion process and product quality, the results were collected into three classes as shown in Figure 10. Light grey indicates a stable process, medium grey indicates probable problems for the reactive compounding, and dark grey indicates parameter settings impossible for the reactive compounding.



		Intense kneading		Medium kneading	
		n=100 rpm	n=200 rpm	n=100 rpm	n=200 rpm
ṁ = 6 kg / h	T_{bmax}=160°C	FC	FC	FC	FC
		TW	TW	TW	TW
		TR	TR	TR	TR
		DS	DS	DS	DS
	T_{bmax}=150°C	FC	FC	FC	FC
		TW	TW	TW	TW
		TR	TR	TR	TR
		DS	DS	DS	DS
ṁ = 12 kg / h	T_{bmax}=160°C	FC	FC	FC	FC
		TW	TW	TW	TW
		TR	TR	TR	TR
		DS	DS	DS	DS
	T_{bmax}=150°C	FC	FC	FC	FC
		TW	TW	TW	TW
		TR	TR	TR	TR
		DS	DS	DS	DS

Figure 11. Illustration of possible processing windows for compounds containing 50 wt.-% rubber powder.



		Intense kneading		Medium kneading	
		n=100 rpm	n=200 rpm	n=100 rpm	n=200 rpm
ṁ = 6 kg / h	T_{bmax}=160°C	FC	FC	FC	FC
		TW	TW	TW	TW
		TR	TR	TR	TR
		DS	DS	DS	DS
	T_{bmax}=150°C	FC	FC	FC	FC
		TW	TW	TW	TW
		TR	TR	TR	TR
		DS	DS	DS	DS
ṁ = 12 kg / h	T_{bmax}=160°C	FC	FC	FC	FC
		TW	TW	TW	TW
		TR	TR	TR	TR
		DS	DS	DS	DS
	T_{bmax}=150°C	FC	FC	FC	FC
		TW	TW	TW	TW
		TR	TR	TR	TR
		DS	DS	DS	DS

Figure 12. Illustration of possible processing windows for compounds containing 70 wt.-% rubber powder.

By using this kind of classification, it is possible to express the results according the Figures 11 and 12. These figures show the strong restrictions for the design of a reactive compounding process:

- Low throughput
- Low barrel temperature
- High screw speed
- Medium filling rate

The intensity of kneading seems to be enough for different screw designs investigated.

Conclusions

Rubber powder can be dispersed using a twin screw extruder in-between a process length of approximately L/D = 14 including the feeding zone and the polypropylene melting.

A simultaneous feeding of both components into the hopper is possible even at high filling rates (70 wt.-% rubber powder).

The effective dispersion of the rubber powder enables a process design with a relatively long reaction time in the extruder. The whole process length of the used extruder is 44 L/D. It was shown, that the dispersion of rubber powder needs only around 14 L/D. A relatively long part of the process length (around 30 L/D) remains for reactive compounding, if the reactants are added downstream after the dispersion process.

Very high filling rates resulted in a lot of contacts between rubber powder particles, i.e. the system seems to be overfilled.

For design of a reactive compounding process a narrow processing window was observed.

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