Continuous Mixing of Silica based Rubber/Filler-Composites in Twin Screw Extruder

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Preface

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1. Introduction

The art of rubber mixing was started from Goodyear’s discovery of sulphur vulcanization of natural rubber in 1839 [1]. The mixing of sulphur and rubber are required to achieve satisfactory vulcanization and hence the technology of rubber mixing may be said to have started some 140 years ago. The main aim of mixing is to make a homogeneous mixture from different ingredients. Mixing is generally referred to as the distribution of single volume elements in a specified volume, where those single volume elements differ in at least one property [2]. The ingredients may differ in structure, viscosity, concentration and rheological behaviour. Apart from ingredients, mixing also depends on the type of mixing equipment. The mixing efficiency depends upon the type, design and process parameters of the equipment used.

The mixing of rubber compounds is traditionally carried out in batch wise mixing equipments, such as roll mills or in internal mixers [3-5]. As the rubbers are available as bales, the batch wise mixing equipments are used until today. Until the development of internal mixers, two roll mills were the major mixing equipment in the rubber industry for many years. Two roll mills are used exclusively in rubber compounding especially for mastication as well as for mixing chemicals. The normal internal mixer is a discontinuous compounding machine which requires high mixing energy [6]. The discontinuous mixing in internal mixer has some advantages, for example: high flexibility regarding different recipes and mixing orders. A major disadvantage of this process is that the batch mixing in internal mixer leads to differences in mixture quality from one batch to another [7-8].

In contrary, continuous mixing processes shows high consistency in mixing quality and needs low mixing energy [9]. Continuous mixing aggregates are state of the art in plastics compounding and extrusion industries for several decades [10]. The availability of plastic raw materials as granules or powders in free-flowing form is practically suitable for feeding in continuous mixing aggregates. As rubbers are available as bales, the applications of continuous mixing aggregates are not yet widely used in rubber industry. But, continuous mixing process is preferable because of cheaper labour costs and better uniformity in quality [9]. Today, a considerable variety of different continuous mixing aggregates are applied for rubber mixing such as planetary roller extruder, twin screw and multi-screw extruder [11-13]. Most of the continuous mixing aggregates originate from the principle of the screw extruder. The continuous
mixing aggregates however need rubber materials as free flowing materials for feeding continuously.

Recently, powdered and granulated rubber materials became available as feeding materials for continuous mixing aggregates. Some of them are EPDM from Dow [14] and Rubber/Filler-Composites (RFC) from Degussa AG (Evonik AG) [15-17]. Since 1998, Degussa AG has developed a number of rubber products where the essential rubber ingredients such as carbon black, silica, extender oil and other chemicals can be incorporated during the production process of Rubber/Filler-Composites [18]. The use of special RFC production techniques leads to a significant reduction of the tackiness of the rubber and additionally helps to keep the material in a free flowing form during storage for longer time. Several RFC products are today available commercially or as development products to the rubber user such as E-SBR/carbon black [16], NR/carbon black [19] and E-SBR/silica/silane [17]. These materials are available either in free flowing powder form or in granule form for feeding any continuous mixing aggregate.

During the middle of 90’s, a new system of compound formulation was introduced by the tire industry for making passenger car tire tread compounds. In the new technology, carbon black as filler is replaced by precipitated and highly dispersible silica. The silica is however highly incompatible with the rubber matrix and hence the silica surface should be modified with a coupling agent called organosilane. The mixing of silica with organosilane along with rubber in the internal mixer involves several mixing stages and long mixing time of 10-15 minutes. This is necessary to react chemically the silica and the organosilane during the mixing process. To avoid this mixing process, a new RFC material was introduced by Degussa AG where the silica is silanized during the production process of RFC [17]. Also, the filler in RFC is fully incorporated in the rubber matrix which additionally promotes less mixing time and better material quality.

Several studies were carried out with continuous mixing aggregates and free flowing raw materials such as EPDM in twin screw extruder [20], RFC containing carbon black in twin screw extruder [21], RFC containing silica in planetary roller extruder [22] and RFC containing silica in twin screw extruder [23]. These studies proved that the continuous mixing is possible with rubber compounds in comparison to discontinuous mixing. The development of a continuous rubber compounding process for the purpose of saving energy is without doubt a great challenge for the tire industry. With such Rubber/Filler-Composites containing carbon black or silica, it is one of the most promising approaches to change to continuous mixing process and it is a chance for the rubber industry in the near future.
2. **Aim of the work**

Until today, several studies were carried out with continuous mixing aggregates and free flowing rubber materials. The mixing of RFCs containing carbon black and silica in co-rotating twin screw extruder were intensively studied by M. Bogun [21]. The mixing of RFCs based on E-SBR and NR with carbon black in twin screw extruder were quite successful and the material properties were even better than the mixtures obtained from the internal mixer. But, the mixing of RFCs based on E-SBR with silica in twin screw extruder were showing poor compound and material properties compared to the mixtures obtained from the internal mixer. Along with that, the silica based compounds lead to higher outlet temperature in twin screw extruder which limited the throughput to a maximum of 37.5 kg/h. In order to overcome these difficulties in mixing of silica based RFCs, the present work is a further step carried out to improve the compound and material properties as well as to optimize the processability in a co-rotating twin screw extruder.

With respect to this, the first goal of this work is to develop a screw configuration in a co-rotating twin screw extruder optimum for mixing RFCs based on E-SBR/silica/silane. Special screw designs were configured with different mixing elements to study the influence on dispersion and distribution of silica particles. By using mixing elements such as forward, reverse and neutral kneading elements, studies were carried out to compare the mixing performance of different kneading elements influencing the compound properties such as filler-filler interaction and mooney viscosity. Along with that, the studies were comparing the mixing efficiency of different kneading elements with respect to specific energy input, residence time as well as outlet temperature of the extrudate.

The mixing performance depends not only on optimum screw design, but also on optimum dosing assembly. As in internal mixer, the ingredients should be dosed in twin screw extruder in right order. There are different barrel sections which can be modularly configured and the dosing of ingredients can be designed to take place along the extrusion direction. The screw configuration and dosing assembly should match each other, so that the incorporation of ingredients easily takes place as well as the compound gets maximum mixing and cooling simultaneously. In order to accomplish optimum incorporation of RFC, filler, plasticizer, activators, antioxidants and cross-linking chemicals, several dosing assemblies were investigated. The influences of dosing assemblies on outlet temperature, specific energy input as well as on compound properties were studied.
Continuous mixing in twin screw extruder gives the possibility to mix all the ingredients in one mixing step. The efficiency of mixing depends upon optimum screw configuration and dosing system. At the same time, the productivity depends upon the maximum throughput possible by the twin screw extruder. On increasing the throughput, the compound temperature increase drastically and it may lead to pre-mature scorching. Taking into account the outlet temperature limit, it is intended to show the possibility of increasing the throughput with one step continuous mixing process while maintaining constant material quality.

The maximum throughput is however limited in one step continuous mixing process due to the influence of higher outlet temperature. On exceeding the outlet temperature limit, pre-mature scorching of the rubber takes place and cross-linking starts during mixing in the twin screw extruder. Hence, a two step continuous mixing process is studied. Here, the first step of mixing is carried out in twin screw extruder and the second step in two roll mill. This leads to increasing the throughput further than the one step continuous mixing without sacrificing the quality of the material properties.

A further goal of this work is to study mixing parameter such as filling degree in the twin screw extruder. The degree of mixing and the material outlet temperature can be greatly controlled by filling degree. Here, the filling degrees are studied in two ways: by keeping the throughput constant and varying the screw speed as well as by keeping the screw speed constant and varying the throughput.

An additional goal of this work is to simplify the continuous mixing process. This has been developed by introducing a new RFC compound where most of the rubber ingredients including some cross-linking agents were mixed during the production process of RFC. This compound is finally available as a single mixture where it requires only one dosing system. All the ingredients are pre-mixed and hence high degree of dispersion between the components can be achieved. The maximum throughputs were studied by this new RFC compound by one step and two step continuous mixing process.

The last part of the work will discuss on continuous mixing of rubber blends based on E-SBR/BR and S-SBR/BR with silica as filler. Different possibilities for mixing blends including the addition of cross-linking chemicals in a one step mixing process were discussed. The influences of extruder parameters and dosing assemblies on the mixing performance of blends were studied. The properties of rubber blends produced in an internal mixer were finally compared.