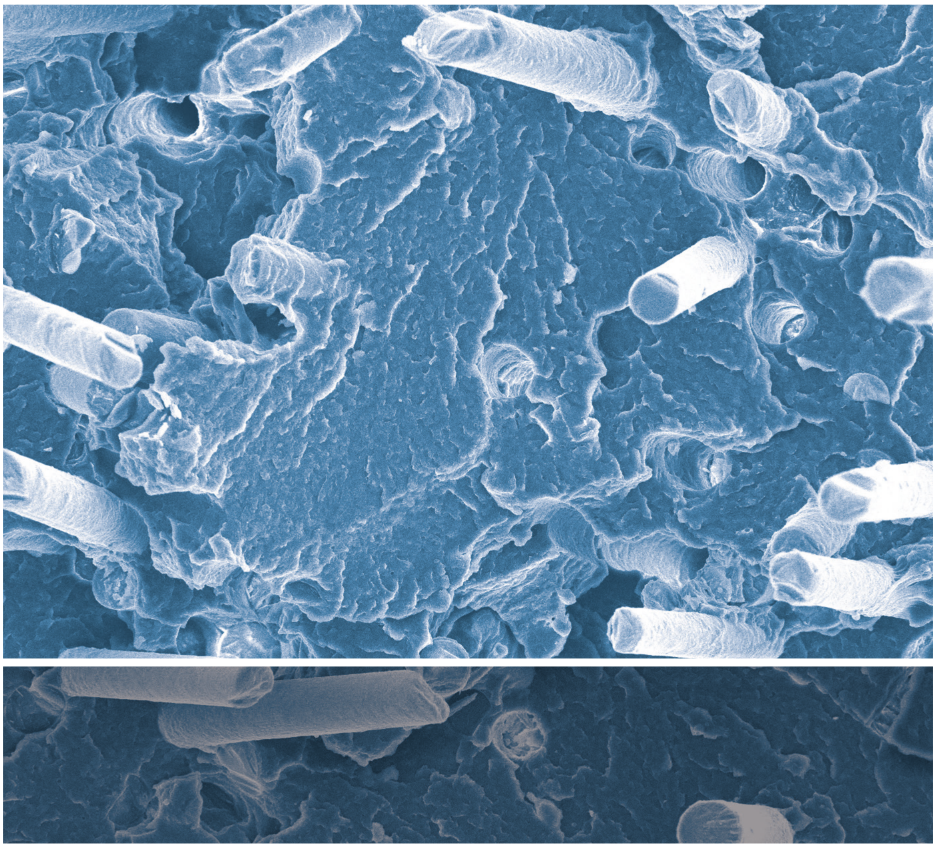
Melanie Rohde-Tibitanzl

Direct Processing of Long Fiber Reinforced Thermoplastic Composites and Their Mechanical Behavior under Static and Dynamic Load



HANSER

Rohde-Tibitanzl

Direct Processing of Long Fiber Reinforced Thermoplastic Composites and their Mechanical Behavior under Static and Dynamic Load

Melanie Rohde-Tibitanzl

Direct Processing of Long Fiber Reinforced Thermoplastic Composites and their Mechanical Behavior under Static and Dynamic Load

HANSER
Hanser Publishers, Munich
Hanser Publications, Cincinnati

The Author:
Dr. Melanie Rohde-Tibitanzl, Karlsbader Straße 7, 90480 Nürnberg, Germany

Distributed in North and South America by:
Hanser Publications
6915 Valley Avenue, Cincinnati, Ohio 45244-3029, USA
Fax: (513) 527-8801
Phone: (513) 527-8977
www.hanserpublications.com

Distributed in all other countries by
Carl Hanser Verlag
Postfach 86 04 20, 81631 München, Germany
Fax: +49 (89) 98 48 09
www.hanser-fachbuch.de

Cataloging-in-Publication Data is on file with the Library of Congress

ISBN 978-1-56990-629-3
E-Book ISBN 978-1-56990-630-9

Coverdesign: Stephan Rönigk
Printed and bound by BoD - Books on Demand, Norderstedt
Printed in Germany

PREFACE

The research presented in this book was carried out during my time as a research assistant at the Department of Polymer Engineering at the University of Bayreuth. Finance for this research project was provided by the Oberfrankenstiftung.

I thank Prof. Dr.-Ing. Volker Altstädt for providing the opportunity to work in this exciting and stimulating research field. Furthermore, I am grateful to Prof. Dr.-Ing. Dietmar Drummer for the reviewing of this work.

I am grateful to all the employees of the Department of Polymers, and Neue Materialien GmbH, for their support, the good cooperation, and the creative, productive, and social working environment. The exchange of scientific ideas, valuable discussions, and constructive and critical suggestions from Dr. Andreas Spörrer, Johannes Müller, Stephan Mörl, and Clemens Keilholz were particularly helpful. Very special thanks should be sent at this point also to Alexander Brückner, Andreas Mainz, Anne Lang, Ute Kuhn, Dirk Pessler, and Andreas Popp.

Hanka Gall, I thank you for the valuable English corrections.

Furthermore, I am grateful to all undergraduate, diploma, bachelor, and masters colleagues for their tireless efforts and dedication: Anna Ebel, Katlen Frenzel, Hannes Welz, Dominik Harant, Georg Schlageter, Anna Malyhin, Elena Kaiser, Verena Durant, Ute Gressano, Florian Jelitschek, Nikolai Witt, and Sebastian Ihlow.

Finally, I thank my parents, who have facilitated my studies and always supported me.

A very special heartfelt thank you goes to my husband Johannes Tibitanzl, who has given me full support in busy and stressful times. Without his loving care, this work would not have been possible in this form. Last but not least, I thank my children Ida Katharina and Veit Joseph Tibitanzl, who teach me every day to strengthen my patience, to put priorities right, and to be a better person.

1 INTRODUCTION

The combination of fibrous materials with polymeric matrices is widely established since several decades. The incorporation of reinforcing fibers into a thermoplastic matrix leads to excellent materials with tailored application-friendly properties allowing the substitution of numerous conventional metallic components [BSM03]. Due to advantages like outstanding performance in terms of stiffness, strength, heat resistance, warpage, mechanical behavior under impact load, recyclability, density, material costs and potential for integration, the development of this material class was extremely accelerated within the last years [Sch03]. Thereby in the year 2013 the European composites market shares 33 % of global composites industry in value and 22 % in volume.

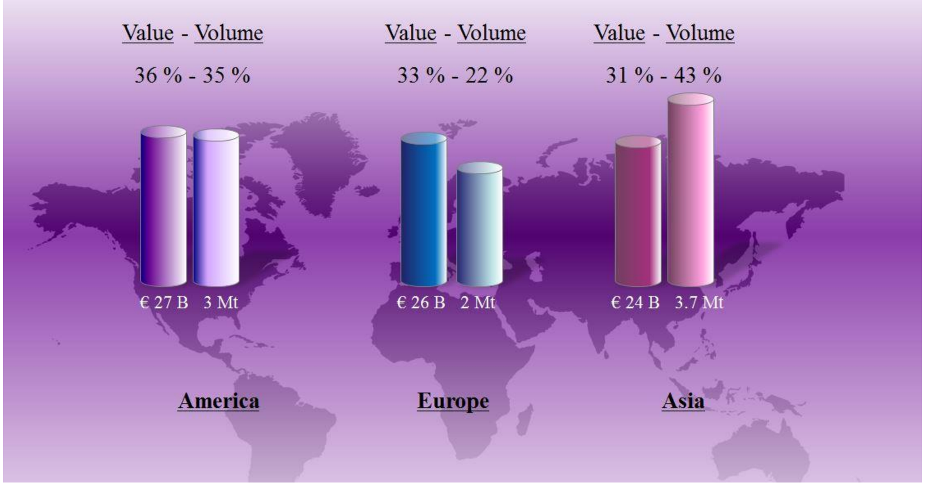


Figure 1.1: Global market share in 2013 [NN13]

The American composites market covers 36 % and 35 % respectively [NN13]. Polymer based composites correspond to 16 % of the US structural materials market (steel: 76 %, aluminum: 8 %) following 6 % annual growth since 1960. During the same period, steel growth had stagnated and growth rate of aluminum remained under 1 % per year.

The development of composites industry is mainly driven by following application sectors:

- Automotive/transportation,
- Wind Energy and
- Aerospace.

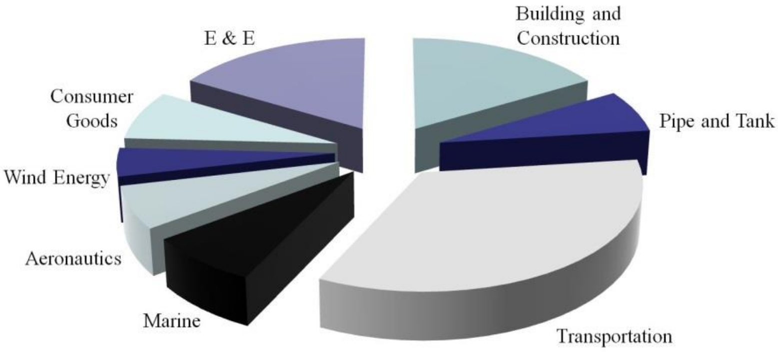


Figure 1.2: The American composites market by application in volume, 2010 [NN13]

The demand for thermoplastic composites is supposed to grow above average until 2015. Benefits like high performance to weight ratio, competitive costs, processing advantages compared to thermoset composites and better design capabilities speed up this trend.

Glass fiber reinforced PP and PA for example are superior construction materials for automotive sector having an outstanding performance. Applications like frontends, dashboard carriers, door modules and underbody structures based on these materials currently dominate this market segment [BSM03, Mar05].

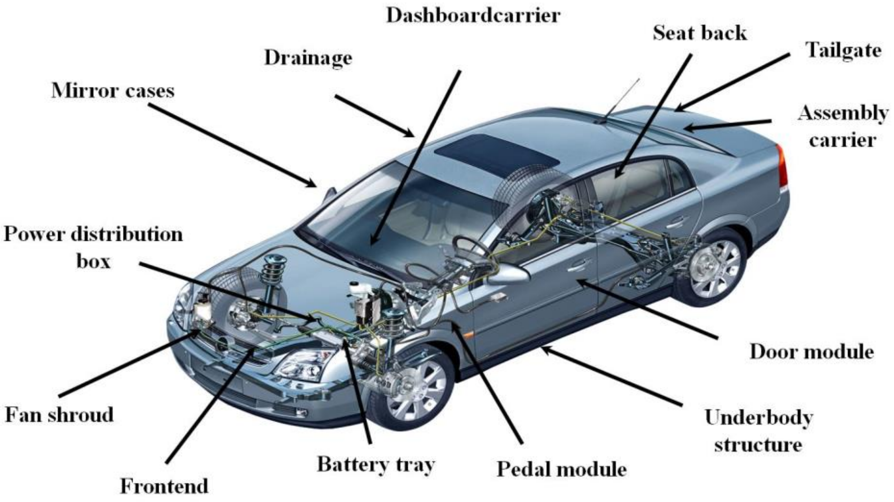


Figure 1.3: Application examples of LFRT in automotive [NN12]

Fiber aspect ratio is a key factor for mechanical behavior. Yet, fiber length is extremely affected by processing. Consequently conventional manufactured LFRT materials do often not perform sufficiently to meet the expectations. Conventional processes are classified into manual, injection, compression and continuous processes as pultrusion or extrusion. Direct processing techniques combining the advantages of twin-screw compounding and injection molding are increasingly applied to manufacture long fiber reinforced thermoplastic composites (LFRT). The fibers are prevented against degradation during processing. The molded components reveal improved average fiber lengths resulting in excellent mechanical performance.

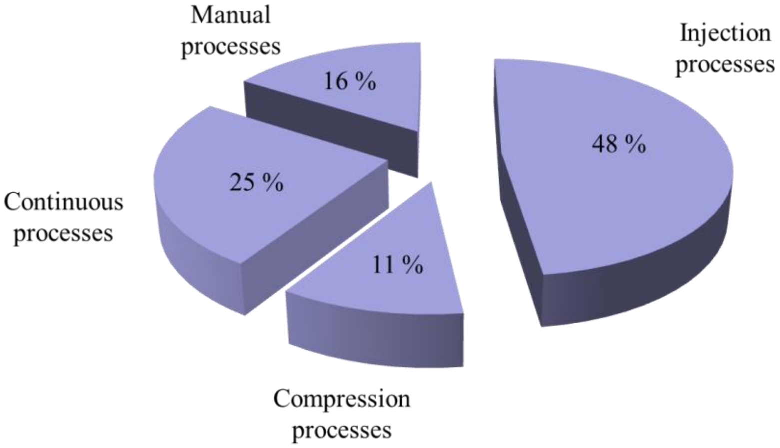


Figure 1.4: Manufacturing processes [NN13]

2 STATE OF THE ART

2.1 Processing of Long Fiber Reinforced Thermoplastic Composites (LFRT)

Compression and injection molding are the most important manufacturing processes for long fiber reinforced thermoplastic composites, *figure 2.1* [Sch08]. The fiber aspect ratio in the manufactured components is a key factor for their superior behavior under mechanical load. To use the full potential of the fiber reinforcement in a thermoplastic material, a certain fiber length has to be achieved. As glass fibers are discontinuous in length, the entire load has to be brought into the fiber via matrix and interface [Sch00]. Does the fiber length exceed a critical value, the composite failure will not occur by collapse of the interface, instead by fiber fracture. The meaning of the critical fiber length will be further explained in chapter 2.2 "Static properties of fiber reinforced thermoplastic composites".

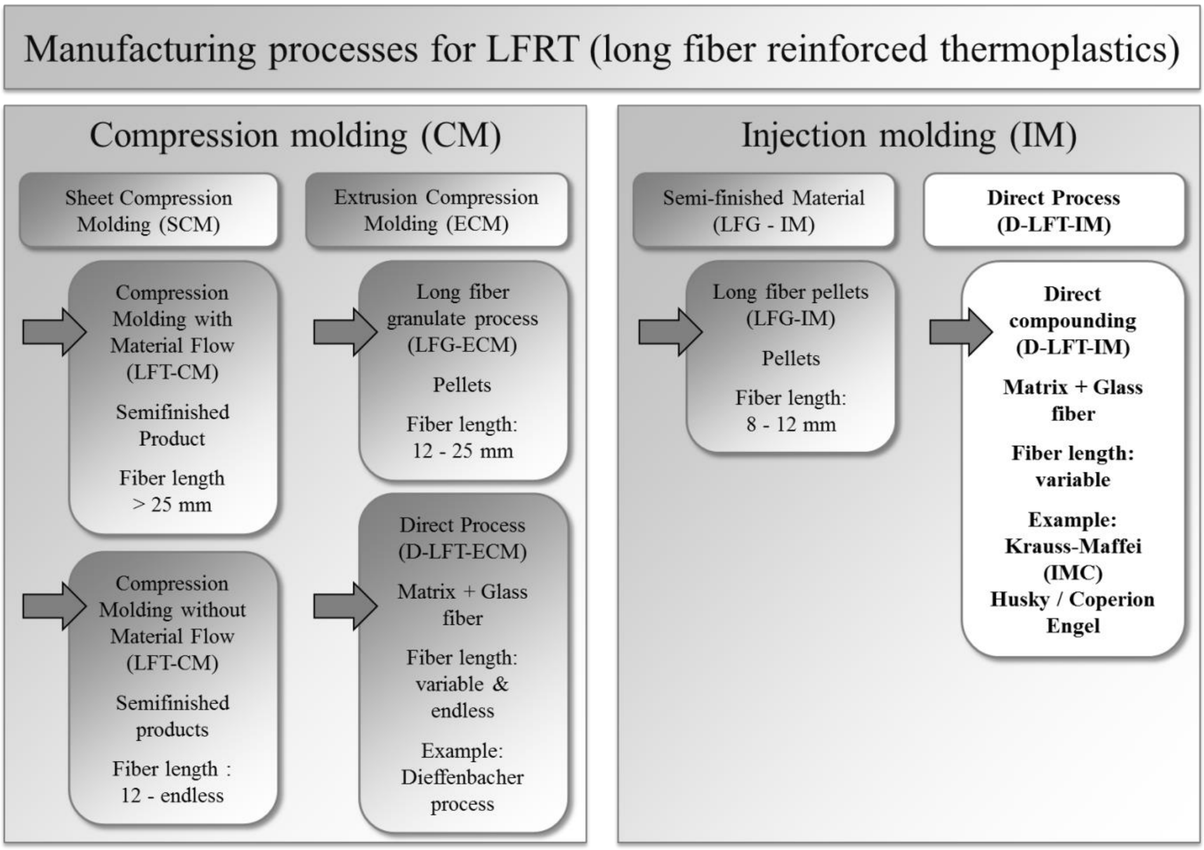


Figure 2.1: Processing technologies for LFRT [Sch08]

Injection molding is a well-established economical method for manufacturing high quality structural parts in mass production without the need for finishing. Therefore different feedstock classes are available, which are classified into [Wol93]:

- screw-compounded short-fiber reinforced grades with initial fiber length of 0.2 – 0.4 mm (short glass fiber: SGF); the short fibers are incorporated as staple fibers during previous compounding
- coextruded / coated and pultruded long fiber reinforced grades (long glass fiber: LGF) [BSM03] with

- sufficiently wetted fibers in pultruded grades and
- non-dispersed fibers in coated grades.

The impregnated fiber strands are cut afterwards into pellets of defined length (10 to 12 mm). The embedded fibers have the same length as the pellets.

The feedstock classes are shown schematically in *figure 2.2*.

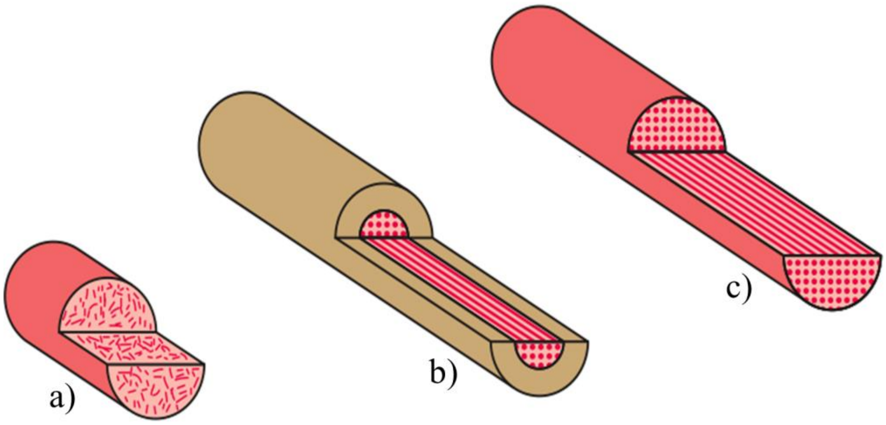


Figure 2.2: Different feedstock classes of fiber reinforced pellets: a) SGF, b) LGF coated and c) LGF pultruded [BSM03]

It is expected that parts manufactured with LGF instead of SGF will provide better mechanical performance. Yet, during injection molding massive additional fiber length degradation takes place induced by shear-intensive melting processes. The resulting fiber lengths are often found between 50 and 300 µm (SGF) or 1,000 and 2,000 µm (LGF) respectively in the molded components. Dependent on the kind of matrix, the average fiber length drops below critical fiber length after processing with dramatic effect on mechanical properties.

Besides fiber length reduction, physical limits of injection molding are characterized by low melt throughput and shot volume, batch-wise operation mode, less homogenizing capacity and restrictions in processing of highly filled formulations [BW02].

The current state of industrial applications demands superior performance of fiber reinforced structural parts in terms of mechanical behavior. However, conventional manufactured fiber reinforced thermoplastic composites do often not perform sufficiently due to reduced fiber length. Therefore multiple direct processing methods have been developed.

One of the first direct processing injection molding machines has been designed by Truckenmüller. He equipped a standard injection molding machine with a special plasticizing unit, which was able to directly feed the glass fiber rovings. The fibers are guided into a special designed vent and the screw pulls the fibers into the plasticizing unit. The vent geometry determines the feed characteristics (fiber length, wetting, fiber feed and content). The content of fiber in the molded part is regulated by screw speed, number of rovings, tex number, matrix material and vent geometry [TF91]. An uneven melt transport resulting from entangled rovings leads to considerably higher metering times compared to conventional injection molding and a non-constant shot weight. The weight average fiber length after this special-purpose processing of PA66-GF is about 4.02 mm compared to commercial materials with 0.38 mm (SGF) and 2.60 mm (LGF). However, the fiber bundles agglomerated and clusters appeared resulting from insufficient fiber distribution. To overcome these disadvantages an optimized non-return valve avoids the occurrence of fiber bundles [Tru93]. Yet, fiber length decreases in this case as a function of fiber content.

The next generation of direct processing technologies combines a twin-screw compounding extruder and an injection and clamping unit. The injection molding compounder (IMC) from KraussMaffei is one of the most relevant direct processing machines for LFRTs and was also used for the present study, *figure 2.3*. The resulting fiber lengths in the manufactured products exceed critical fiber length leading to outstanding mechanical properties. However, thermoplastic composites are always subjected to severe fiber fracture during the whole process either by injection molding or by compounding or combining processing methods.

For this reason the following paragraphs analyze the critical stages for fiber length during the process of injection molding compounding. The current state of the art unfortunately does not give enough information about fiber length degradation during injection molding compounding. Thus references will be discussed, which will explain the fiber length degradation either during injection molding or compounding. The mechanisms are assumed to be transferable for injection molding compounding.

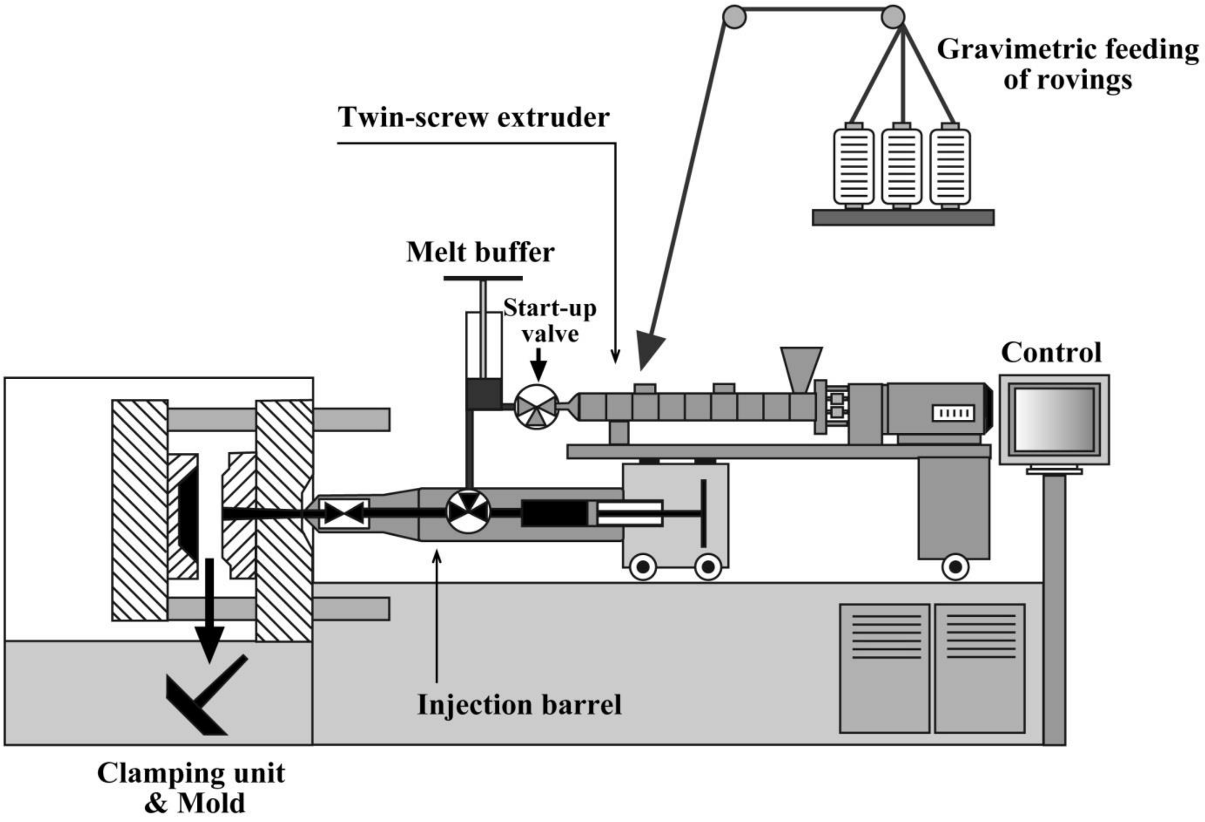


Figure 2.3: Scheme of the injection molding compounder

2.2 General mechanisms of fiber length degradation during direct processing

In general three main reasons for fiber fracture exist [Fis85, HPW+00]:

- Fiber-fiber interaction resulting from collisions, spatial hindrance, friction etc.
- Fiber-polymer interaction induces stresses without any concurrent interaction from other fibers
- Fiber-machine interaction

The physical conditions of these mechanisms are briefly explained in the following section.

Fiber fracture due to fiber-fiber interaction

Fiber concentration plays a major role for this mechanism. It starts to occur when the fibers in the melt are free to move and their centroids are close to one another. Suspensions are classified into dilute, semi- and highly concentrated systems as shown in *figure 2.4*.

In dilute or sub-critical suspensions the distance between a fiber and its nearest neighbor is larger than the fiber length L_F. The fibers are free to rotate and interactions are rare [Thi91, BK00]. At high fiber aspect ratios the volume fraction must be quite small for dilute suspensions [FT84]. As a matter of fact practically no commercial compound is a dilute suspension [FT84].

The spacing between fibers in semi-concentrated or transient suspensions is less than the fiber length L_F but larger than the fiber diameter D_F. Fiber-fiber interactions frequently occur [FT84] and the first hindering of fiber rotation due to collision is observed. Therefore the space of mobility virtually reduces to a disc [Thi91].

In highly concentrated or super-critical suspensions the distance between fibers is in the order of the fiber diameter D_F [FT84]. The fiber mobility is almost reduced to the fiber volume [Thi91] and limited in each direction. Due to high fiber content the suspension behaves like a solid [BK00]. Thereby in flow processes fiber agglomeration and orientation were observed.

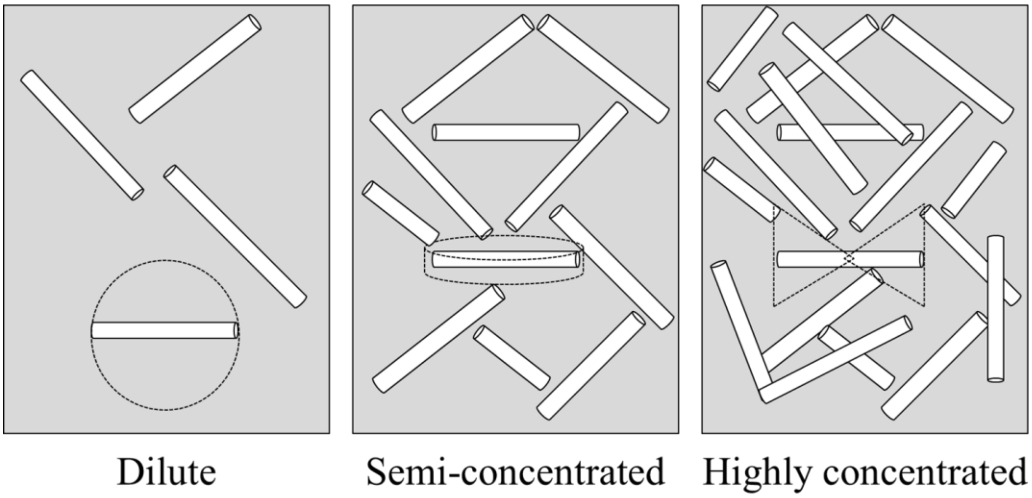


Figure 2.4: Three different kinds of suspensions dependent on fiber concentration [EG86]

These above mentioned regimes of fiber concentration are defined by fiber concentration and geometrical parameters such as fiber diameter D_F and fiber length L_F [Bat71, FT84, Thi91]. Fiber concentration is then characterized by fiber volume fraction Φ:

$$\Phi = \frac{1}{1+\frac{1+\psi}{\psi}\cdot\frac{\rho_F}{\rho_M}} \qquad \textit{(Eq. 2.1)}$$

with ψ as fiber weight fraction, ρ_F as density of the glass fibers, ρ_M as density of the matrix polymer.