DEVELOPMENT OF A METHOD TO CHARACTERIZE SCREW ELEMENTS FOR A TWIN SCREW EXTRUDER

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SUMMARY

Twin screw extrusion is a commonly used technique in the pharmaceutical industry. The material inside the extruder is modified by rotating screws. The final properties of the product depend on a variety of parameters, e.g. rotation speed, temperature and the presence of fluids or vacuum vents. A very important variable during extrusion is the screw type and the screw configuration which is used. By changing the screw configuration a product with different properties may be formed. During the development of new drug formulations it is often unknown which screws will be most suitable to get a product with the desired properties. For this reason a development of a characterization method for several screws is done. The screws can be characterized based on their geometry, but also on their distributive and dispersive mixing properties. During each experiment also pressure and torque are measured.

Some errors occurred during examination of distributive and dispersive mixing by the screw elements, but the measurements of the torque and the pressure were successful.

From the results of the experiments, it can be stated that the torque on the screws increased by an increasing screw speed and an increasing loading of the extruder barrel. For the GFA elements (conveying elements), the torque is also significantly increased by the viscosity of the substance inside of the extruder. The highest axial pressure is measured with the GFA 2-15-90 elements, followed by the GFA 2-20-90, the GFA 2-30-90 and the GFA 2-40-90 elements. The KB elements (kneading elements) show a low conveying effect, except for the KB 5-2-30-30 elements, in combination with a high loading (28 g) of a high viscous substance.
SAMENVATTING

Twin screw extrusion is een veelgebruikte techniek in de farmaceutische industrie. Het ingebrachte materiaal wordt door middel van ronddraaiende schroeven bewerkt. Hierbij zijn onder andere de temperatuur, de rotatiesnelheid van de schroeven en de aanwezigheid van vloeistoffen en vacuüm openingen van belang. Afhankelijk van de gebruikte schroeven en de gebruikte schroefconfiguratie, zal het product bepaalde eigenschappen bezitten. Bij het ontwikkelen van nieuwe geneesmiddelvormen, weet men echter niet welke schroef de juiste eigenschappen bezit om het gewenste product te bekomen. Daarom wordt een methode ontwikkeld om een aantal schroeven te karakteriseren op basis van hun drukgeneratie, en mengcapaciteiten. Ook het krachtmoment op de schroeven wordt gemeten.

Bij het nagaan van de mengcapaciteiten van de verschillende schroeven zijn enkele problemen opgetreden, maar de druk en het krachtmoment konden goed gemeten worden in deze studie.

Er kan geconcludeerd worden dat het krachtmoment op de schroeven toeneemt met de rotatiesnelheid en met de belading van de extruder. Voor de GFA elementen (transportelementen) wordt ook een invloed van de viscositeit waargenomen. De hoogste axiale druk wordt gevormd door de GFA 2-15-90 elementen, gevolgd door de GFA 2-20-90, GFA 2-30-90 en GFA 2-40-90 elementen. Tijdens de experimenten met de KB elementen (kneedelementen) wordt over het algemeen geen grote drukstijging waargenomen, behalve in het geval van de KB 5-2-30-30° elementen in combinatie met een hoge belading van de extruder met een hoog viskeuze substantie.
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USED ABBREVIATIONS

API: Active Pharmaceutical Ingredient
D: Screw element diameter
GFA: Conveying element
GFM: Mixing element
KB: Kneading element
NaCl: Sodium chloride
Q²: Predictive relevance
R²: Coefficient of determination
rpm: rotations per minute
1 INTRODUCTION

1.1 Extrusion

Extrusion is a widely used technique and has already been introduced in many industrial fields. Its applications vary from the production and processing of plastics to the production of food and drugs. There is a variety of extruders available on the market, e.g. screw extruders (single and twin screw extruders), sieve extruders, basket extruders, roll extruders and ram extruders (Hicks and Freese, 1989).

1.2 Twin screw extrusion

1.2.1 General aspects

The screw of Archimedes forms the basis for all screw extruders. This screw was invented in the classical antiquity to transfer water in an upward direction, e.g. to store it for irrigation (Oldfather, 1933) or for draining mines (Oldfather, 1939). The Archimedes screw is still in use for these purposes (Rorres, 2000). An example of an Archimedes screw is shown in Figure 1.1.

Figure 1.1: An Archimedes screw used for drainage of mines (Palmer, 1926)

The first fully developed and commercialized screw extruders were single screw extruders, used in the polymer industry, around 1930 (Bruin et al., 1978). In 1935 the single screw extruder got commercialized in the pasta industry (Rossen and Miller, 1973). Twin screw extruders were commercialized later on (Colombo, 1939).

A twin screw extruder has two screws, which are fitted side by side. There are many different types of twin screw extruders. A first distinction can be made between counter-rotating and co-rotating twin screw extruders (see 1.2.3 and 1.2.4). A second distinction can
be made between intermeshing and non-intermeshing twin screw extruders (see 1.2.5). Twin screw extruders are often preferred over single screw extruders, because of their better mixing properties (Ferns, 1974). In twin screw extruders, the material is transported from one screw to another, which gives a good mixing on a macroscopic level (Mollan, 2003). Also on microscopic level a good mixing occurs, due to high shear regions between the screws. Because of their good mixing properties, twin screw extruders often are used in compounding processes. An advantage of the intermeshing twin screw extruder compared with the single screw extruder, is the better positive conveying and thus the shorter residence time of the component in the extruder (Mollan, 2003). A short residence time is desirable for the extrusion of labile components.

Meanwhile a large number of twin screw extruders has been developed for use in the pharmaceutical industry, for example in granulation processes. The advantage of the extruder is that many processes can be combined in one system. For example, water or other fluids can be added during the extrusion process, or by increasing the temperature melt extrudates can be formed out of the drug. At the same time the extruder mixes the sample and desired chemical reactions can be carried out.

1.2.2 Design of a twin screw extruder

In each screw extruder the energy for rotation of the screws is provided by the motor. A gearing reduces the motor speed to the desired rotation speed (rpm). The gear is surrounded by oil. The temperature and thus the viscosity of this gear oil influences the power needed to turn the screws (Köster and Thommes, 2010). In a twin screw extruder, the screws are attached on two shafts which are connected to the gear. A general scheme of an extruder is shown in Figure 1.2.

![Figure 1.2: Extruder design of a Leistritz Micro 27GL-28D (Köster and Thommes, 2010)](image-url)
The barrels which surround the shafts, can be connected to a powder feeder or liquid feeder. There is also the possibility to degas or vent at the barrel. The temperature can be set on the control panel of the extruder. To achieve this temperature, the barrels can be heated electrically, or cooled via liquid. Temperature sensors measure the temperature of the barrel in different places. The measured temperature is then displayed on the control panel. At the last barrel, the extrudate is pressed out through a die, with a certain diameter. In Figure 1.3 a picture of an extruder is shown.

![Image of extruder](image)

**Figure 1.3: Leistritz labextruder Micro 27 (Leistritz)**

1.2.3 Counter-rotating twin screw extruders

In a counter-rotating twin screw extruder the two screws rotate in opposite directions. When the material enters the gap between the two screws, a high pressure is formed (Shah and Gupta, 2004). This pressure is increasing with an increasing rotation speed of the screws, and may cause degradation of the processed material. That is why counter-rotating twin screw extruders should be used at low rpm. Another disadvantage of counter-rotating twin screw extruders is the potential air entrapment (Mollan, 2003). Counter-rotating twin screw extruders are mainly used in the polymer industry, e.g. in plastic production. In this case, the extruder can be fed with plastic pellets, which are modified during the extrusion process. Depending on the used operating variables and screws a certain melting process occurs, by which desired plastics can be obtained (Wilczyński and Lewandowski, 2012).
1.2.4 Co-rotating twin screw extruders

In a co-rotating twin screw extruder much lower pressures occur, which makes it possible to work at a higher rotation speed. A co-rotating twin screw extruder has a higher mixing capacity than a counter-rotating twin screw extruder (Rauwendaal, 1981), and is the most important extruder type in the pharmaceutical industry. The difference between counter-rotating and co-rotating screws is illustrated in Figure 1.4.

![Figure 1.4: Classical intermeshing co-rotating and counter-rotating screws (Thiele, 2003)](image)

1.2.5 Intermeshing and non-intermeshing twin screw extruders

In fully-intermeshing extruders there is no open space between the screws and the barrel, which causes a good conveying of the material inside of the extruder. All the material is processed in the same way, which is essential in extrusion for pharmaceutical preparations. Non-intermeshing extruders do not form closed compartments, so there is no self-wiping of the screw, and some of the material may stick to the screw. This causes an unequal treatment of the material during the extrusion process, which is not favorable. Non-intermeshing twin screw extruders may serve well for removing large amounts of volatiles, or for processing high viscous substances, which may cause problematical torque-buildups in an intermeshing system (Mollan, 2003). An illustration of an intermeshing and a non-intermeshing system is given in Figure 1.5.

![Figure 1.5: Intermeshing screws (left) and non-intermeshing screws (right) (Steiner, 2003)](image)
1.3 Screw elements

1.3.1 General

Different types of screw elements can be used in a twin screw extruder. Generally there are four major types of screw elements, namely conveying elements, kneading elements, backward-pumping elements and mixing elements (Sämann, 2008). For each type there are many different subtypes. The geometry determines the properties of each screw. Examples for conveying, kneading and mixing elements are shown in Figure 1.6.

**Figure 1.6: Conveying elements (left), kneading elements (middle), mixing elements (right)**

1.3.2 Conveying elements

The geometry of the conveying elements enables a good transport of the product through the extruder. Each conveying element is characterized by its number of flights, its length in millimeters, and its pitch. The pitch is defined as the axial length in millimeters, required for a complete thread. Conveying elements with a large pitch have a greater conveying effect than those with a small pitch (Sämann, 2008). These elements are used to forward the material e.g. at feed openings, in order to avoid that pressure increases excessively at the openings. An example of conveying elements is shown in Figure 1.6 (left).

1.3.3 Kneading elements

Kneading elements are mostly used for dispersive mixing, but they are also used for distributive mixing processes (Sämann, 2008). They consist of kneading discs with a certain thickness (Figure 1.6, middle). Like the conveying elements there are several parameters by which the kneading elements are characterized. These parameters include the number of flights, the staggering angle between sequential kneading discs (in degrees), the pitch
direction (clockwise or counter-clockwise) and the element length (in millimeters) (Sämann, 2008). Except for their mixing properties, kneading elements can also be used for devolatilizing and draining of the substance. Kneading elements with a small staggering angle are also capable to forward the material through the extruder, but therefore their axial mixing effect is less intense. In general, kneading elements with a smaller disc width provide a better distributive mixing (Sämann, 2008) than those with thicker discs, which provide a better dispersive mixing (Yerramilli and Karwe, 2004).

### 1.3.4 Mixing elements

Some of the mixing elements are called “Igel” elements, due to their shape. They are designed for a good homogenization of the substance, with a minimum of dispersive, shear-intensive mixing. These elements can be conveying, neutral, or backward-pumping, depending on their geometry. Screw mixing elements have the same geometry as the conveying elements but there are gaps in the flights, parallel to the screw axes. They are shown in Figure 1.6 (right). The mixing occurs when the product which is conveyed through the extruder, flows back via the gaps in the flights (Sämann, 2008).

### 1.3.5 Backward-pumping elements

Backward-pumping elements give a good distributive mixing and as the name implies they transport the product upstream instead of downstream. They are used for performing a more longitudinal homogenization (Sämann, 2008). The difference between a conveying element and a backward-pumping element is shown in Figure 1.7.

![Figure 1.7: Conveying element (left) and backward pumping element (right)](image-url)
1.3.6 Zoning elements

Zoning elements do not have their own effect on the material in the extruder, but they serve as a barrier function in the extruder, for example to separate the vacuum vent from the other zones in the extruder. This example is shown in Figure 1.8.

![Figure 1.8: Illustration of the use of zoning elements during twin screw extrusion](image)

1.3.7 Naming convention of screw elements

The conveying elements are also referred to as GFA elements. The mixing elements used in this study are called GFM elements. The kneading elements are known as the KB elements. When a specific screw is described, one of these three codes is used, followed by a numerical code, which describes the geometry of the screw. Two examples of a code are shown in Figure 1.9. A conveying element is described by the code GFA, followed by three numbers. The first number is its number of flights, in this study only double flighted screw elements are used, so this number is always two. The second number describes the pitch of the screw and the third number describes the length of the screw. For a kneading element four numbers are used to describe it. The first number describes the thickness of the used kneading discs, the second number describes the flights of the kneading element, the third number describes the length of the kneading element and the fourth number describes the staggering angle between the kneading discs. The GFM elements are described by three numbers, namely the number of flights, the pitch and the length.
1.4 Mixing behavior

1.4.1 Distributive mixing

Distributive mixing implies a rearrangement of the different particles in the extruder, without a change in their morphological characteristics. The longer the distributive mixing continues, the more homogeneous the mixture. Mixing elements have good distributive mixing properties.

1.4.2 Dispersive mixing

In contrast to distributive mixing, dispersive mixing causes morphological changes of the material. Regions of high pressure are able to transform the material and may for instance give rise to particle size reduction of drops or agglomerates. Dispersive mixing occurs at kneading elements. Usually both types of mixing occur at one screw element, but depending on the screw geometry one of the two mixing types will dominate. The differences between dispersive and distributive mixing are illustrated in Figure 1.10.
1.5 Characterization of the screw elements

Based on the geometrical structure of the screw elements, an impression of the mixing and conveying qualities of the screw elements can be obtained. Yet it is difficult to predict the exact mixing and conveying properties of each screw. A pure mathematical evaluation of the screws does not succeed in predicting the effect on the drug in the extruder system (Mollan, 2003). Not only the geometry of the screws is an important parameter for the final properties of the product, but also the working conditions such as the loading of the extruder, screw speed and temperature have an influence on the final result. That is why it is so difficult to create a good extrusion setup from the beginning. Characterization of screw elements has been researched before by pharmaceutical companies. But as the outcome of this research is of high value, it is kept as secret corporate information. Therefore there is no public information available about characterization of screw elements, the planning of the screw configuration is always based on former experiments. The characterization of the screw elements, based on their geometry, their mixing capacities, and their conveying properties, could be a real timesaver for those who are starting a new extrusion process.

1.6 Pharmaceutical applications of twin screw extrusion

1.6.1 General aspects

Extrusion has a wide range of applications in the pharmaceutical industry. It can be used to produce homogeneous mixtures, granules and pellets of API’s and excipients to ensure a stable solid dosage form with defined properties.

In a granulation process, particle granules are formed, which are usually large enough to flow and easy to process (Holdich, 2002). Granules are complexes of powder particles. Pharmaceutical granules have a size from 0.1 mm to 2 mm (Holm et al., 1983). By conducting spheronization processes, pellets can be obtained out of the granules. Pellets are part of the group of granules, and are characterized by a spherical surface. There are many reasons for forming granulates or pellets, for example to avoid segregation of API’s in a mixture. Granulates and pellets also have better flow properties than a powder and for this reason they can be used to obtain an equal filling degree of capsules. Pellets are easy to coat, so they can be used in the manufacturing of enteric-coated dosage forms. Granulation is often
applied prior to tabletting of the substance, because most of the pharmaceutical powders are not easily compressed in a tablet in their original form.

Extrusion processes can be divided in wet extrusion, hot-melt extrusion and cold extrusion. These processes are discussed in the following paragraphs.

1.6.2 Wet extrusion
In wet extrusion, liquid excipients are added to the powder, to facilitate the processing. After the extrusion process the obtained extrudates are dried, in order to form a rigid product. This drying step can be done at room temperature (Hasznos et al., 1992), at elevated temperature in a fluidized bed (Chapman et al., 1986) or in an oven (Woodruff and Nuessle, 1972). The presence of a solvent in wet extrusion processes entails two disadvantages, namely the possible degradation of moisture sensitive drug products and the additional drying step, which is time consuming.

1.6.3 Hot-melt extrusion
Hot-melt extrusion is used in the pharmaceutical industry for the production of controlled release dosage forms (Follonier et al., 1995). In hot-melt extrusion, a high-melting polymer is added to the pharmaceutical substance. The extruder barrel is heated above the melting temperature of this polymer. The pharmaceutical drug particles get dispersed in the molten polymer, by rotation of the screw elements. In this way, a uniform dispersion of fine drug particles in the polymer is obtained (McGinity and Zhang, 2003). The mixture solidifies when it leaves the extruder, through the die-plate. As the API is embedded in a polymer matrix, a dosage form with modified drug release can be obtained by using hot-melt extrusion. During hot-melt extrusion for production of granulates, a pharmaceutical grade thermal polymer should be used (McGinity and Zhang, 2003), which is a polymer with a low melting temperature. By using pharmaceutical grade thermal polymers, the temperature of the extruder barrel can be kept relatively low during the extrusion process. This is of substantial importance, because of the temperature sensitivity of most drugs. Compared to batch-processes hot-melt extrusion has one major advantage, namely the short process time of the drug product in the extruder. Short process times can be required to avoid heat degradation of the drug product. Process times during hot-melt extrusion are usually between 5 seconds and 10 minutes (Crowley et al., 2007). The process time depends on the
type of the extruder, the type of screw elements, the used screw configuration and the operation variables (Crowley et al. 2007).

1.6.4 Cold extrusion

In cold extrusion no liquids are added and the processing temperature stays low, which is good for temperature and water sensitive products. Cold extrusion is done with materials that yield under mechanical stress (Thommes, 2012). In recent years, research has been done concerning cold extrusion processes, in which triglycerides are added. These processes are typical hot-melt extrusion processes, but because of the low processing temperature they can be called cold extrusion processes (Breitkreutz et al., 2003).

1.6.5 Continuous granulation

In recent years there has been an increasing interest in continuous granulation, because of its many advantages compared to batch granulations. In continuous granulation there is a continuous feed of raw material and a continuous production of granulates, while batch processes are limited to the loading of the production equipment. In continuous granulation, the granulation process is not interrupted and therefore batch-to-batch differences can be avoided. Due to this advantage and to the improved quality of the obtained granulates, continuous granulation is gaining more interest in the pharmaceutical industry (Lodaya and Mollan, 2003).

The three major types of granulation processes are melt granulation, wet granulation and dry granulation. Normally granulation is done by batch-processes, for these batch processes fluid bed granulators (Wurster, 1959) and high shear granulators (Holm et al., 1983) are used.

Different continuous granulation methods are in use. These include continuous fluid bed agglomeration (Bonde, 1997), mechanical agglomeration (Bonde, 1997) and twin screw extrusion (Ghebre-Sellasie, 2002). Using a twin screw extruder, continuous granulation is mostly done by a wet extrusion process without using a die-plate (Thommes, 2012).
1.7 Experimental work

1.7.1 General aspects

For the characterization of the different screws, several experiments are carried out. First some preliminary extrusion tests are done, to know whether the used setup gives useful results. After these preliminary tests, adjustments in the operating procedure are made, to optimize the working conditions. The obtained ‘standard operating procedure’ is used for the final extrusion experiments. The experiments are done in an extrusion barrel, closed at the front and at the back. In this type of setup screw elements with a length of only 90 mm can be used for investigation of their characteristics. By using a closed system, the effect of different loadings of the extruder barrel can be determined. Silicon oil and sodium chloride particles are processed by the rotating screws within this barrel.

1.7.2 Silicon oil

Silicon oil is a polydimethylsiloxane, its molecular structure is shown in Figure 1.11. Depending on the length of the molecules, the silicon oil has different properties, e.g. a different molecular weight and viscosity. Because silicon oil is a Newtonian fluid, its viscosity will remain constant during the extrusion experiments. In addition to the dynamic viscosity, a kinematic viscosity is defined. This is the ratio between the dynamic viscosity and the density of the substance. The kinematic viscosity is measured by using a capillary viscometer. A substance with a high density will start to flow much easier than a substance with the same dynamic viscosity, but with a low density. The kinematic viscosity can be represented by Equation 1.1.

\[
\gamma = \frac{\eta}{\rho}
\]  

Where: \( \gamma \) : Kinematic viscosity (m²/s)

\( \eta \) : Dynamic viscosity (Pa.s)

\( \rho \) : Density (kg/m³)

![Molecular structure of silicon oil](Wacker Chemie AG, 2012)
1.7.3 Torque

The torque can be seen as a force which extends to turn something around its axes. The magnitude of the torque is given by Equation 1.2. When \( \theta \) is 90°, the torque can be given by Equation 1.3.

\[
T = r \cdot F \cdot \sin(\theta)
\]  

(1.2)

With \( T \): Torque (Nm)
\( r \): Length of the torque arm (m)
\( F \): Applied force (N)
\( \theta \): Angle between the force vector and the torque arm vector

\[
T = r \cdot F
\]  

(1.3)

With \( T \): Torque (Nm)
\( r \): Length of the torque arm (m)
\( F \): Applied force (N)

During extrusion, a certain torque on the screws is applied. This torque is correlated to the force exerted by the screws and thus by the mechanical power consumption (Köster and Thommes, 2010). This power consumption correlates with the rheological properties of the substance between the screws. The more viscous this substance is, the more power will be needed to make the screws turn. In this way, measurement of the torque can be used to observe changes in the viscosity of the processed material during extrusion.

1.7.4 Statistical values

For the interpretation of the results, statistical models are used to process the data. The reliability of a statistical model is characterized by four different values, namely \( R^2 \), \( Q^2 \), model validity and reproducibility. \( R^2 \) is the coefficient of determination, which is the ratio of the variance of the predicted values and the variance of the outcomes in the experiment. \( Q^2 \) is the predictive relevance and is calculated by Equation 1.4. Values close to 1 for both \( R^2 \) and \( Q^2 \) indicate a very good model with excellent predictive power.
\[ Q^2 = (1 - \text{PRESS/SS})^2 \]  \hspace{1cm} (1.4)

With \( \text{PRESS} \): the prediction residual sum of squares.

\( \text{SS} \): the total sum of squares of Y corrected for the mean.

The validity is calculated by Equation 1.5. When the model validity is larger than 0.25, there is no lack of fit of the model. This means that the model error is in the same range as the pure error. When the model validity is less than 0.25 there is a significant lack of fit and the model error is significantly larger than the pure error (reproducibility). A model validity value of 1 represents a perfect model.

\[
\text{Model validity} = 1 + 0.57647 \cdot \log(plof) \hspace{1cm} (1.5)
\]

With \( plof \): p for lack of fit.

The reproducibility is the variation of the response under the same conditions (pure error), compared to the total variation of the response, and is given by Equation 1.6. A reproducibility value of 1 represents perfect reproducibility.

\[
\text{Reproducibility} = 1 - \left(\frac{\text{MS(Pure error)}}{\text{MS(total SS corrected)}}\right) \hspace{1cm} (1.6)
\]

With \( \text{MS} \): Variance

1.7.5 Content distribution and particle size distribution

To determine the distributive and dispersive mixing, a substance is added to the system. The content distribution and the particle size distribution of this substance are determined after each extrusion process. Sodium chloride is used because of its large particle size and its good electrical conductivity when it is dissolved in water. By using sodium chloride, the distributive mixing can be measured by conductivity measurements.

The electrical conductivity of a substance is a mark for its ability to conduct an electric signal. It is the conductance of a 1 cm³ solution and is expressed in Siemens per meter (S/m). The conductance is the reciprocal of the resistance of the solution. The aim of the conductivity measurements is to measure the sodium chloride distribution in the silicon oil, using a certain screw type. The conductivity of a solution correlates with the amount of ionic substances in that solution. As sodium chloride is a strong electrolyte, it dissociates almost
completely in water. The Na\(^+\) ions and Cl\(^-\) ions will cause a higher conductivity than pure water. This happens in a linear way: the more ions, the higher the conductivity.

The dispersive mixing can be measured by using sodium chloride particles of a known size, and measuring the particle size distribution after the experiment. Measurement methods for particle size distribution are e.g. laser diffraction and microscopy.
2 OBJECTIVES

Extrusion has become an indispensable process in pharmaceutical applications. Depending on the conditions in which the extrusion is performed, the extrusion process can be classified into Hot Melt Extrusion (Follonier et al., 1994), Wet Extrusion (Vervaet et al., 1994) or Continuous Granulation (Van Melkebeke et al., 2008).

As extrusion is used for a wide range of applications, various screw elements are available. For a given application a certain combination of screw elements is needed to get an extrudate with the desired qualities. Once the right combination of screws is found for a certain process, high amounts of the required product can be produced. The problem is that it is time consuming to find the right screw combination to obtain a specific extrusion result through trial and error. Also in financial terms this trial and error is not desired: the extruder used in this work, needs at least 2 kg of the powder which has to be extruded. When working with expensive drug products, or a newly developed active pharmaceutical ingredient, these extrusion trials can consume a lot of money.

No public information is available about characterization of screw elements, because the results of this kind of research are kept as secret corporate information in the pharmaceutical companies. Therefore it is useful to find a method for characterizing screw elements. A characterization would provide more information about the mixing and conveying properties of the screws.

To develop a method to characterize different screw elements, the torque on the screws is measured and the pressure generated by the screw elements is measured. Also the distributive and dispersive mixing properties of the screw elements are investigated.

The following screw types are tested: GFA 2-15-90, GFA 2-20-90, GFA 2-30-90, GFA 2-40-90, KB 5-2-30-30°, KB 5-2-30-90°, GFM 2-15-90.
3 MATERIALS AND METHODS

3.1 Materials

Witocan (Witocan 42/44, Sasol Germany GmbH, Witten, Germany) is used for the determination of the barrel volume. Oil (Spartan EP 150, Exxon Mobil, Germany) is used as a lubricant, to avoid friction on the screws during experiments. Silicon oil 10000 and 100000 (Wacker® AK 10000/100000 Silicon oil, Wacker Chemie AG, Burghausen, Germany) are used to fill the extruder. Sodium chloride (AnalaR NORMAPUR Sodium chloride, VWR International BVBA, Leuven, Belgium) is used for investigation of distributive mixing by the screws. Petroleum ether and cyclohexane are used as a solvent for silicon oil. Demineralized water (Laboratorium of Pharmaceutical Technology, Heinrich-Heine Universität, Düsseldorf) is used for making solutions.

3.2 Methods

3.2.1 Experimental setup

A lab scale extruder Micro 27GL-28D (Leistritz, Nuremberg, Germany) is used. For the characterization of the screw elements, the screw elements are isolated in a barrel with a length of 11 cm, this corresponds to 4D (four times the diameter of 1 screw element). The barrel is sealed from both sides with metal plates. The design of the barrel and seal is shown in Figure 3.1.

![Figure 3.1: Extruder barrel and seals (left), detail of the back seal (right)](image)

Two rods are placed in the extruder. On each rod a metal tube is placed, followed by a small brass ring, the chosen screw and a tip, which serves for tightening the screw. Different screw types are tested, these include the following: GFA 2-15-90, GFA 2-20-90, GFA 2-30-90,
GFA 2-40-90, KB 5-2-30-30°, KB 5-2-30-90°, GFM 2-15-90. The screw configuration is shown in Figure 3.2 and Figure 3.3.

Figure 3.2: Screw configuration. From left to the right: Tightening screw tips, screws (GFA 2-30-90), brass rings, metal tubes.

Figure 3.3: Detail of the screw configuration. From the left to the right: tightening screw tips, screws (GFA 2-30-90), brass rings.

The small brass rings fit within the holes in the back seal plate. The diameter of the brass rings is important because if they are too wide, they can hinder the turning of the screws and block the system. If the rings are too small, the system is not closed anymore and the content of the barrel could flow out. To avoid frictions, some oil is put on the rings before each experiment. To control if the screws are attached correctly, the extruder is turned on at 5-10 rotations per minute. When the assembling of the screws is done in the right way, the setup can continue. The extruder barrel is placed around the screws and a front plate is bolted to the barrel with a torque wrench (TORCOFIX-SE, Rahsol) until a torque of 50 Nm is formed. The pressure sensor I31-S6-MB01C1-4-D (Gefran, Provaglio d’Iseo Brescia, Italy) is fastened in the middle of this front plate. It measures the pressure in the middle between the two screws. The used pressure sensor is not available on the market yet, because it is a prototype. It has a measuring range of 10 MPa. For extrusion processes normally a pressure sensor with a measuring range of 35 MPa is used. By using the low range pressure sensor, the measured pressure is expected to be more accurate than with the broad range pressure sensor.
In each experiment, the extruder is filled with silicon oil through the topside of the barrel. The top of the barrel is closed by an upper part. To investigate the distributive and dispersive mixing, sodium chloride is added to the silicon oil in the extruder. Sodium chloride is added via the top opening of the extruder, between the two screws, as close as possible to the back seal. This is illustrated by figure 3.4.

Figure 3.4: Illustration of addition of sodium chloride to the extruder system. The screws shown in this picture are GFA 2-15-90 elements. The conveying direction is illustrated by the black arrow.

3.2.2 Determining the free barrel volume

First the total volume in the extruder barrel is determined, and afterwards the volume of each screw is determined. By subtraction of the screw volume from the total volume, the free volume in the extruder is calculated for each screw.

To measure the total volume of the extruder, 130 g of Witocan 42/44 (Witocan 42/44, Sasol Germany GmbH, Witten, Germany) is melted on a Memmert water bath (Memmert GmbH & Co, Schwabach, Germany) at 80 ° C. The extruder barrel is filled with the molten Witocan, with the Witocan at a temperature of 60.8 ° C, measured with an infrared thermometer (LaserSight D206-01-A, Optris GmbH, Berlin, Germany). After 30 minutes in the extruder barrel, the solidified Witocan is taken out and weighed. Using the density of molten Witocan, the corresponding volume can be calculated. To know the density of molten Witocan, a Helium pycnometer (Micromeretics GmbH, Mönchengladbach, Germany) is used at 50 ° C. The measuring conditions of the Helium pycnometer are described in section 3.2.3.
The volume of each screw is determined by water displacement. Therefore the screw is attached to the rod with the screw tightening tips and put into a graduated cylinder, filled with 100.0 ml water. The rise of the water level corresponds with the volume of the screw.

### 3.2.3 Density measurement of silicon oil

The density of silicon oil is determined with a Helium pycnometer (Micromeretics GmbH, Mönchengladbach, Germany). Before each measurement, the system is rinsed 10 times with helium, with a filling pressure of 134.5 kPag. The measurements are done with the same filling pressure. The measurements start when the pressure between the two chambers of the helium pycnometer is lower than 0.0345 kPag. A measurement is done 5 times per sample. Three samples are tested for each silicone oil and the density is determined by the mean of the three results.

### 3.2.4 Calibration of the torque

The calibration of the torque is done in the way described by Köster and Thommes (2010). The motor of the extruder is mechanically blocked. A lever of 1.00 m is attached to the coupling of the screws. A torque gauge T22/50 (HBM Messtechnik, Darmstadt, Germany) is placed between the motor and the gear box. Nine different masses between 350 g and 2000 g are attached to the lever, in order to calibrate the system. Each mass causes a certain torque on the screws. The signal, provided by the torque gauge is processed by an analogue/digital-transformer-card PCI-6031-E (National Instruments, Los Angeles, USA) within a personal computer. The experimental setup is illustrated in Figure 1.2 under section 1.2.2. The same torque gauge and analogue/digital-transformer-card are used for the extrusion experiments.

### 3.2.5 Development of the method for measuring content distribution of sodium chloride in silicon oil

#### 3.2.5.1 Conductivity calibration curve for sodium chloride in water

The measured conductivity of the samples is correlated with the sodium chloride content. This content can be calculated by using a calibration curve.

When there is a poor distributive mixing by the screws, the sodium chloride will be present in high concentrations on the place where it is added. That is why a calibration curve is made in a concentration range from 0 g/l to 1.5 g/l. For this calibration curve, 0.0125 g/l,
0.0200 g/l and 0.100 g/l solutions are prepared by diluting a 0.1000 g/l solution. Six other solutions are made by weighing sodium chloride, transferring it to a 100 ml volumetric flask and diluting it to the calibration mark. The weighed amounts of sodium chloride are: 0.02532 g, 0.05245 g, 0.07552 g, 0.10166 g, 0.12494 g and 0.15039 g. The solution of 1.5 g/l sodium chloride in water corresponds to a 100 mg sample with 22.5 % sodium chloride in silicon oil.

3.2.5.2 Sample preparation

After the experiments with the extruder, samples of sodium chloride in silicon oil are obtained. To measure the content of sodium chloride in silicon oil, the following sample preparation is done. To each sample, 2.5 ml petroleum ether is used to dissolve the silicon oil. 15.00 ml demineralized water is added as an extraction solvent. Then the samples are placed in an REAX2 overhead shaker (Heidolph Instruments GmbH & Co, Schwabach, Germany) for 2 hours. 10 ml of the aqueous phase is taken out of each vial and its conductivity is measured with a Mettler-Toledo SevenMulti conductivity meter. Each sample is measured three times.

3.2.5.3 Evaluation of the sodium chloride extraction procedure

To evaluate the extraction procedure, 9 samples are made as described in Table 3.1. The silicon oil used in this experiment has a viscosity of 100000 mm²/s. The sample preparation and measurement is done in the same way as described under 3.2.5.2.

Table 3.1: Composition of samples for evaluation of a sodium chloride extraction procedure

<table>
<thead>
<tr>
<th>Sample nr.</th>
<th>NaCl [g]</th>
<th>Silicon oil [g]</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>0.00507</td>
<td>0.50804</td>
</tr>
<tr>
<td>A2</td>
<td>0.00521</td>
<td>0.49666</td>
</tr>
<tr>
<td>A3</td>
<td>0.00522</td>
<td>0.49658</td>
</tr>
<tr>
<td>B1</td>
<td>0.00528</td>
<td>0</td>
</tr>
<tr>
<td>B2</td>
<td>0.00509</td>
<td>0</td>
</tr>
<tr>
<td>B3</td>
<td>0.00518</td>
<td>0</td>
</tr>
<tr>
<td>C1</td>
<td>0.00516</td>
<td>0</td>
</tr>
<tr>
<td>C2</td>
<td>0.00518</td>
<td>0</td>
</tr>
<tr>
<td>C3</td>
<td>0.00519</td>
<td>0</td>
</tr>
</tbody>
</table>
3.2.6 Development of the method for measuring particle size distribution of sodium chloride in silicon oil

3.2.6.1 Laser diffraction

The Sympatec Laserdiffractor (Sympatec GmbH, Clausthal, Germany) was used, and the suspensions are tested in a cuvette with a magnetic stirrer. Different methods are tested to measure the particle size distribution of sodium chloride in silicon oil. Petroleum ether and cyclohexane are used as dispersion media. For each measurement a reference measurement is done with the dispersion medium without particles. Dispersions of sodium chloride in petroleum ether, and sodium chloride in cyclohexane are tested. Different particle sizes of sodium chloride are tested. To test small particle sizes, the sodium chloride is ground with mortar and pestle. Sodium chloride particles with a size between 500 µm and 630 µm are obtained by sieve analysis and are used to test big particle sizes. For the small particles the R2 lens is used, this lens is able to measure particles with a diameter between 0.45 µm and 87.5 µm. For the particles with a size between 500 µm and 630 µm the R5 lens is used. This lens has a measuring range between 4.5 µm and 875 µm. Different ultrasonication times are tested, namely 0 s, 60 s and 500 s. Rotation speeds between 0 and 2500 rpm of the magnetic stirrer are tested. After the ultrasonication a break of 10 s is inserted before the particle size distribution is measured. This measurement is carried out for 10 s.

3.2.6.2 Microscopy

Sodium chloride is grinded with mortar and piston, and 0.5008 g sodium chloride is suspended in 10.2 g silicon oil with a viscosity of 10000 mm²/s. One drop of this suspension is examined with a Leica Q500/550 020-319.011 DMLB 100S light microscope (Leica Microsystems, Wetzlar, Germany). The light microscope is connected to a Leica DC 300F camera (Leica Microsystems, Wetzlar, Germany), and a personal computer with Qwin Imaging Software (Leica Microsystems, Wetzlar, Germany).

3.2.7 Preliminary extrusion experiments

3.2.7.1 Procedure

Some preliminary extrusions are done to test the suitability of the experimental setup. Before starting the experiments the extruder is running without screws for 1 hour at 400 rpm. During the whole experiment, the pressure and torque are measured. 1 signal per
second is obtained via the pressure sensor and the torque gauge T22/50 (HBM Messtechnik, Darmstadt, Germany) and the analogue/digital-transformer-card PCI-6031-E (National Instruments, Los Angeles, USA). The temperature of the extruder is set at 20°C. After adding silicon oil to the barrel, the screw speed is quickly increased to 200 rpm, in order to get a narrow distribution of the silicon oil around the screws. The screw speed is changed every three minutes. For each experiment a blank run is done, during which the screw speed is changed every two minutes.

3.2.7.2 Variables

Only four of the seven aimed screws are tested. These include the following: GFA 2-40-90, GFA 2-15-90, KB 5-2-30-30° and KB 5-2-30-90°. Per screw the high viscous and the low viscous silicon oil are tested. Three different loadings of the extruder barrel are tested, namely 12.1 g, 24.1 g and 36.1 g, which corresponds to a 25%, 50% and 75% filling of the extruder. For each loading the screw speed is increased from 50 rpm to 200 rpm, in steps of 50 rpm. After three minutes at screw speed 200 rpm, the screw speed is again decreased to 150 rpm, 100 rpm and 50 rpm.

3.2.8 Examination of different screw elements

Based on the results from the preliminary extrusion tests, a standard operating procedure is developed. The temperature of the extruder is set at 20°C. The extruder is running for 1 h at 100 rpm without any screws or barrels attached. This is to keep the temperature of the gear oil constant during extrusion. Every morning and evening a blank run is done at the different rotation speeds: the extruder is operated successively at 50, 100, 150 and 200 rpm, each time for 2 minutes. Between the different experiments the blank run is carried out by running the extruder at 100 rpm for 3 minutes. The experiment can be carried out after the blank run and the assembling of the extruder: the extruder is filled with a certain amount of silicon oil (corresponding to a 25% or a 75% filling of the extruder) and shortly increased to 200 rpm, in order to get a narrow distribution of the silicon oil around the screws. Per screw the high viscous and the low viscous silicon oil are tested in a high percentage of loading (75%) and a low percentage of loading (25%).

After each experiment, an amount of sodium chloride (equivalent to 5 % of the added loading of silicon oil) is added between the screws via the top opening of the barrel. An
illustration of how the addition of sodium chloride occurs, is shown in Figure 3.4 (see section 3.2.1). The barrel is closed afterwards and the screws are turned for 15 seconds at 50 rpm. 6 samples (A to F) are taken via the top opening of the barrel, as shown on Figure 3.5.

Figure 3.5: Sampling of the sodium chloride in silicon oil to investigate the distributive mixing.

After this sampling the extruder is run for another 15 seconds at 200 rpm, and again 6 samples are taken. The samples are used to investigate the particle size distribution of sodium chloride in silicon oil and the dispersive mixing of the sodium chloride particles.

In order to obtain a higher reliability of the results, four of the experiments are repeated. Table 3.2 shows which experiments are chosen to be repeated.

Table 3.2: Used parameters for the repetitions of four experiments

<table>
<thead>
<tr>
<th>Used screw element</th>
<th>Viscosity of the silicon oil</th>
<th>Loading of the extruder</th>
</tr>
</thead>
<tbody>
<tr>
<td>GFA 2-40-90</td>
<td>10000 m²/s</td>
<td>25 %</td>
</tr>
<tr>
<td>GFA 2-15-90</td>
<td>100000 m²/s</td>
<td>75 %</td>
</tr>
<tr>
<td>KB 5-2-30-90°</td>
<td>10000 m²/s</td>
<td>25 %</td>
</tr>
<tr>
<td>KB 5-2-30-30°</td>
<td>100000 m²/s</td>
<td>75 %</td>
</tr>
</tbody>
</table>
4 RESULTS AND DISCUSSION

4.1 Development of measurement methods

4.1.1 Determining the free barrel volume

The weight of the Witocan in the extruder barrel is determined as 111.2 g, while the density of molten Witocan, measured by Helium pycnometry, is 0.8940 g/cm³. Based on these data the total volume in the extruder barrel is calculated. This volume is 124.4 cm³.

For all elements, except for the GFM 2-15-90 element, a water displacement of 38 ml occurs. The GFM 2-15-90 elements gives a water displacement of 35 ml. The free volume in the extruder can be calculated by subtracting the volume of the screws from the total volume in the barrel, which is 48.4 cm³.

4.1.2 Density of silicon oil

For the silicon oil with a viscosity of 10000 mm²/s a density of 0.9759 g/cm³ is measured, and for the silicon oil with a viscosity of 100000 mm²/s a density of 0.9769 g/cm³ is measured. For the extrusion processes, silicon oil loadings of 12 g, 24 g and 36 g are used, corresponding to 25 %, 50 % and 75 % filling of the extruder.

4.1.3 Calibration of the torque

The results for the calibration of the torque are shown in Figure 4.1 and show a good correlation between the measured voltage and the torque generated at the lever arm.

![Figure 4.1: Calibration curve of the voltage in function of the torque](image)
4.1.4 Content distribution of sodium chloride in silicon oil

4.1.4.1 Conductivity calibration curve for sodium chloride in water

The obtained calibration curve is shown in Figure 4.2 and Equation 4.1 will be used to calculate the sodium chloride concentration in the following conductivity measurements.

\[
y = 1942.9 \, \mu\text{S.l/g} \times x + 8.143 \, \mu\text{S}
\]  

(4.1)

Where:  
\(y\): the measured conductivity (\(\mu\text{S}\))

\(x\): the concentration of sodium chloride in water (\text{g/l})

4.1.4.2 Evaluation of the sodium chloride extraction procedure

The results of the conductivity measurements of the prepared samples are listed in Table 4.1. Samples A1, A2 and A3 closely resemble the samples which are obtained after extrusion, because they contain both sodium chloride and silicon oil. Samples from B1 to C3 do not contain silicon oil. The expected concentration is calculated by the amounts of sodium chloride weighed (these are given in Table 3.1).

As the measured concentrations of sodium chloride in water are not lower than the expected concentrations, we can assume that all the sodium chloride is extracted to the aqueous phase. All measured concentrations are higher than the expected value, which has several possible explanations. The first explanation could be that the temperature for the measurements is higher than the temperature, used for making the calibration curve. A
second explanation could be a day to day variability of the conductivity measurement system. A third explanation could be the presence of more ionic substances in the glass vials or in the silicon oil itself. In the last case a new calibration curve should be made, using the same glass vials as those used for conductivity measurements of the samples and the same amount of silicon oil.

**Table 4.1: Conductivity results for three groups of samples with a different composition**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Mean Conductivity [µS]</th>
<th>Concentration [g/l]</th>
<th>expected concentration [g/l]</th>
<th>Difference between measured - expected [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>679.3</td>
<td>0.3514</td>
<td>0.3380</td>
<td>+1.8</td>
</tr>
<tr>
<td>A2</td>
<td>699.7</td>
<td>0.3620</td>
<td>0.3473</td>
<td>+2.1</td>
</tr>
<tr>
<td>A3</td>
<td>700.3</td>
<td>0.3624</td>
<td>0.3480</td>
<td>+2.0</td>
</tr>
<tr>
<td>B1</td>
<td>698.0</td>
<td>0.3612</td>
<td>0.3520</td>
<td>+1.3</td>
</tr>
<tr>
<td>B2</td>
<td>675.0</td>
<td>0.3492</td>
<td>0.3393</td>
<td>+1.4</td>
</tr>
<tr>
<td>B3</td>
<td>683.3</td>
<td>0.3535</td>
<td>0.3453</td>
<td>+1.1</td>
</tr>
<tr>
<td>C1</td>
<td>682.0</td>
<td>0.3528</td>
<td>0.3440</td>
<td>+1.2</td>
</tr>
<tr>
<td>C2</td>
<td>685.3</td>
<td>0.3546</td>
<td>0.3453</td>
<td>+1.3</td>
</tr>
<tr>
<td>C3</td>
<td>680.3</td>
<td>0.3520</td>
<td>0.3460</td>
<td>+0.8</td>
</tr>
</tbody>
</table>

**4.1.5 Development of the method for measuring particle size distribution of sodium chloride in silicon oil**

**4.1.5.1 Laser diffraction**

The used petroleum ether does not have the necessary quality for laser diffraction, with the result that the impurities in the petroleum ether adhere to the surface of the cuvette and disturb the measurements. Sodium chloride of a small particle size can be measured in cyclohexane after 60 s of ultrasonification with a rotation speed of the magnetic stirrer at 500 rpm. Particles with a size between 500 µm and 630 µm show sedimentation and cannot be measured with this method. The magnetic stirrer has to be turned at 2500 rpm to get the sodium chloride in suspension. At this rotation speed disturbances in the glass vial occur, which interfere with the measurement. Sample measurements at low rpm would always give the same particle size distribution, because only the small particles in dispersion would be measured. For this reason other methods have been sought to measure the particle size.
4.1.5.2 Microscope

In microscopy, sodium chloride particles are clearly visible. The image of the particles, as seen through the microscope, is transferred to the personal computer.

![Sodium chloride particles in silicon oil as seen through the light microscope: size 500 µm to 630 µm (left), ground sodium chloride particles in silicon oil (right)](image)

As seen in Figure 4.3, the particles from 500 µm to 630 µm are clearly distinguishable, which allows a correct particle size measurement. The image of the grinded sodium chloride shows that the sodium chloride particles tend to attach to each other. This makes it difficult to measure the particle sizes. The samples are stored in vials, so in future research other methods can be investigated to measure the particle size of the sodium chloride particles suspended in silicon oil. One option might be to use a CAMSIZER® XT (Retsch Technology GmbH, Haan, Germany). This machine can be used for dry and wet analysis (Retsch Technology, 2012). A wet analysis of the samples can be carried out after the addition of e.g. cyclohexane, in order to dissolve the silicon oil and decrease the viscosity of the sample.

4.2 Preliminary experiments with the extruder

In the preliminary experiments, four of the seven screw elements are tested. The raw data of two experiments are shown in Figure 4.4. The left graph in this figure is obtained from an experiment with the GFA 2-15-90 screw, the right graph is obtained from an experiment with the KB 5-2-30-90° screw. In both experiments the high viscous silicon oil is used.
Figure 4.4: Raw signal from preliminary extrusion experiments: Screw GFA 2-15-90 (left) and Screw KB 5-2-30-90° (right), both experiments with the high viscous silicon oil (viscosity of 100000 mm²/s). Screw speed (black), Torque (blue) and Pressure (red) are plotted versus Time. 25 %, 50 % and 75 % represent the loading of the extruder barrel.

The peaks at the left part of the graphs are caused by an increase in screw speed to test whether the screws are attached correctly. Between two subsequent loadings, the speed is set at 0, to open the extruder and add more silicon oil. After adding the new loading of silicon oil, the screw speed is turned up to 200 rpm in order to equilibrate the system, and to avoid drift in the torque and pressure measurements.

For the experiment with screw GFA 2-15-90, there is an obvious correlation between the torque and the screw speed, and between the pressure and the screw speed. With a higher loading, a higher torque and pressure are achieved at the same screw speed.

For each loading, the screw speed is increased from 50 rpm to 200 rpm and back decreased from 200 rpm to 50 rpm. The values for the pressure and torque at a certain loading and a certain screw speed do not depend on whether the screw speed is being increased or decreased. At some points a small difference in torque and pressure is observed, for example at the 50 % loading at 150 rpm. This difference can be explained by a small difference in screw speed: a slightly higher screw speed gives a slightly higher torque and pressure.

In both experiments no pressure is seen with a loading of 25 %. At higher loadings there is a difference between the two shown experiments. In the experiment with the GFA 2-15-90 element a much higher pressure is generated, compared to the experiment with the KB 5-2-30-90° element, where almost no pressure can be observed. This can be explained by a large
conveying effect of the GFA 2-15-90 elements, and poor transporting capacity of the KB 5-2-30-90° elements.

In the following graphs torque or pressure are plotted in function of the screw speed. The data used for torque and pressure are calculated by the mean value of the last 30 measured values before changing the screw speed. This is to take into account a possible equilibration phase after changing the screw speed.

Figure 4.5: Blanked torque in function of the screw speed (left), Pressure in function of screw speed (right). Experiments with conveying elements are shown. Different pitches of the screws, loadings of the extruder barrel and viscosities of the loading are shown.

In Figure 4.5 (left), the blanked torque obtained from the experiments with the GFA elements, is plotted in function of the screw speed. The influence of the pitch, the loading and the viscosity of the loading on the torque can be read from this figure. The curves for the GFA 2-15-90 elements are represented by broken lines and the curves for the GFA 2-40-90 elements are represented by continuous lines. As seen on Figure 4.5 (left), the GFA 2-15-90 elements induce a higher torque than the GFA 2-40-90 elements. The torque increases with the loading and with the viscosity of the used silicon oil. A linearity between the torque and the screw speed is observed, but also some unexpected values appear, e.g. with the GFA 2-40-90, loading 25% and viscosity 100000 mm²/s, where the torque seems to be negative. The unexpected results can be explained by an insufficient way of blanking. If the blank run is not carried out under the same conditions as the experiment, wrong values for the experimental torque can be obtained. One condition which is probably the most important, is the temperature of the gear oil. This temperature rises when high screw speeds are
applied. A high temperature decreases the viscosity of the gear oil and as such also the torque. The insufficient blanking of the torque is also observed in the experiments with the kneading elements (Figure 4.7, left).

The measured blank values for the torque are shown in Figure 4.6. This figure demonstrates that the blank values at a chosen screw speed vary over a certain torque range. The blank runs in the preliminary tests are not always sufficient to blank the torque in a correct way. Therefore more blank runs are done during the new extrusion experiments.

![Graph showing blank torque](image)

**Figure 4.6:** Blank torque (measured torque without screws) at different screw speeds. The legend shows for which screw element the blank is used, ‘10000’ or ‘100000’ refers to the viscosity of the loading, which is respectively 10000 mm²/s or 100000 mm²/s.

![Graph showing pressure and screw speed](image)

**Figure 4.7:** Blanked torque in function of screw speed (left), Pressure in function of screw speed (right). Experiments with conveying elements are shown. Different pitches of the screws, loadings of the extruder barrel and viscosities of the loading are shown.
Figure 4.5 (right) and Figure 4.7 (right) illustrate the pressure in the front of the extruder barrel, in function of the screw speed. Several screws, loadings and viscosities are compared. The pressure and screw speed show a linear correlation. The pressure is much higher with most of the GFA elements (Figure 4.5), the scale of this graph is ten times larger than the scale of the graph which is describing the KB elements (Figure 4.7). The highest pressure is given by GFA elements with a small pitch (GFA 2-15-90) and a high loading (75 % filling) of the extruder barrel.

Based on these preliminary results, some changes in the experimental setup can be done, in order to optimize the measurements. A loading of 25 % gives no pressure at the front of the extruder. This can be explained by the high volume at the front of the extruder, between the screw tips. This volume is 13 ml. It is possible that by using the low loading, almost all the silicon oil is transported to the front of the extruder and is not having any contact with the screw elements. In that case it is not possible to generate any pressure, even with a strong conveying element. Therefore Teflon rings with a volume of 10.5 ml are made to cover the screw tips, by which the dead volume at the front is reduced. The front of the extruder barrel with and without Teflon tips is shown in Figure 4.8. Because of this volume reduction, new loading weights are calculated. The used weights of silicon oil to fill the extruder barrel are now 9.5 g (25 % loading) and 28.5 g (75 % loading).

Figure 4.8: The front of the extruder barrel. Without Teflon tips (left) and with volume reducing Teflon tips (right)
4.3 Examination of different screw elements

4.3.1 General aspects

The influence of the pitch, the loading of the extruder and the viscosity of the used silicon oil on torque and pressure is investigated, using a statistical regression model. The torque and pressure have a linear correlation with the screw speed. Therefore the rate between the torque and the screw speed, and the rate between the pressure and screw speed are calculated. These rates are assumed to be large for experimental setups which cause a high torque or pressure generation.

The distributive mixing is investigated by the results of the conductivity measurements on the six samples taken in each experiment. Starting from the measured conductivity, the content of sodium chloride in silicon oil can be calculated by using Equation 4.1 (section 4.1.4.1) and the mass of the original sample. The content can be expressed as a mass percentage of sodium chloride in silicon oil. The relative standard deviation of this percentage is calculated for each set of six samples. A relative standard deviation of 0 would correspond with an equal distribution of sodium chloride in silicon oil over the whole system. A large relative standard deviation is equivalent with a poor distribution of the sodium chloride in the extruder barrel, and therefore with a poor distributive mixing.

4.3.2 Conveying elements

4.3.2.1 Influence of experimental setup on the measured torque

As mentioned in 4.3.1, the rate between the measured torque and the used screw speed is calculated for each experiment. The values for these rates are plotted in a three dimensional scatter plot (Figure 4.9). The three axes of this scatter plot are represented by the pitch of the screws in mm, the loading of the extruder in g and the viscosity of the used silicon oil in mm²/s. The values for the rate between the torque and screw speed in each experiment are represented by color codes: a red color indicates a high value for the torque rate, a deep blue color indicates that no increase of the torque is occurred. Based on these color codes in the scatter plot, it can be seen that a high torque is generated during experiments with the high viscous silicon oil (100000 mm²/s) and a high loading of the extruder barrel (a loading of 28.5 g, which corresponds to a 75 % filling of the barrel). These experiments are marked with a red circle in Figure 4.9.
Figure 4.9: Rate between the torque and the screw speed for different experiments. Three axes are used, one for the pitch of the screws [mm], one for the viscosity of the silicon oil [mm²/s] and one for the loading of the extruder barrel [%].

This effect is also shown on Figure 4.10 (right): the torque rate is significantly increased by the loading of the extruder, and by the viscosity of the silicon oil. Screws with a lower pitch seem to cause a higher torque, but this is not statistically significant. The product of the viscosity and the loading also fits in the model, because it shows a statistically significant influence on the torque rate. The reliability of the model is shown in Figure 4.10 (left). The values for $R^2$, $Q^2$, the model validity and the reproducibility of the model can be read from this figure.

With a lower pitch, a higher torque is expected, because screws with a low pitch should give more shear stress between the screws and the extruder barrel. However, in these experiments the pitch does not have a significant influence on the torque. The reason for this might be that the extruder barrel is closed at the front and the processed silicon oil cannot flow away from the screws. When the silicon oil is conveyed by the screws and it reaches the front plate, it will be pushed back towards the screws, which causes a high torque. As the system is closed in each experiment, this effect will occur for each pair of screw elements, which makes it difficult to distinguish the different screw elements based on the torque.
4.3.2.2 Influence of the experimental setup on the generation of pressure

The values for the rate between the pressure and the screw speed are plotted in a three dimensional scatter plot (Figure 4.11). This is done in the same way as the scatter plot which displays the torque rate (Figure 4.9, section 4.3.2.1). Figure 4.11 shows that high pressure is only generated in experiments with the high viscous silicon oil, except for the experiment with the combination of the 15 mm pitch conveying elements and a high loading of the extruder. In the latter case, pressure is also generated using the low viscous silicon oil. As this is only seen for the experiments with the GFA 2-15-90 elements, it demonstrates the good conveying properties of these elements.

A regression model for the pressure rate in function of the screw speed is shown in Figure 4.12 (right). In this model, only the experiments with the high viscous silicon oil are encountered. It can be seen that a low pitch and a high loading cause a significant increase of the pressure. Also the squared pitch and the product of pitch and loading are included in the model. Even though the squared pitch is not a statistical significant influence, it is not excluded from the model, because it is necessary for a good fitting of the model (Figure 4.12, left). An execution of the model is shown in Figure 4.13, where a clear correlation between the loading, pitch and the pressure rate is visible.
Figure 4.1: Pressure rate in function of the screw speed for different experiments. Three axes are used, one for the pitch of the screws [mm], one for the viscosity of the silicon oil [mm²/s] and one for the loading of the extruder barrel [%].

Figure 4.12: Left: A summary of fit for the regression model for the pressure rate. $R^2$ (Coefficient of determination), $Q^2$ (Predictive relevance), model validity and reproducibility for the model are shown. Right: Scaled and centered coefficients for predicting the rate between the pressure and the screw speed, experiments with conveying elements and high viscous silicon oil. Coefficients for the pitch of the screws (1), the loading of the extruder (2), the squared pitch of the screws (3) and the product of viscosity and loading (4) are shown.
Figure 4.13: Execution of the regression model for predicting the rate between the pressure and the screw speed, at different loadings of the extruder and pitches of the used conveying elements. Only experiments with the high viscous silicon oil are encountered.

4.3.2.3 Influence of the experimental setup on the distributive mixing

In the experiments with the GFA 2-15-90 elements and a low loading (9.5 g) of the extruder, it was not possible to take samples A to D (see Figure 3.5, section 3.2.8). This is because all the silicon oil and sodium chloride was conveyed to the front of the extruder barrel. The calculated relative standard deviation for the content distribution of sodium chloride in silicon oil is between 0.1893 and 1.2341. Here, the results from the experiments with the GFA 2-15-90 and a low loading of the extruder barrel are excluded. As shown in Table 4.2, the results for the distributive mixing show a high repetition error, by which they lose their reliability. For this reason, the results for the content distribution cannot be used for the characterization of the screw elements.

Table 4.2: Results for content distribution of sodium chloride in silicon oil

<table>
<thead>
<tr>
<th>Screw element</th>
<th>Loading [g]</th>
<th>Viscosity [mm²/s]</th>
<th>RSD</th>
<th>RSD for repetition</th>
</tr>
</thead>
<tbody>
<tr>
<td>GFA 2-15-90</td>
<td>29</td>
<td>100000</td>
<td>1.2341</td>
<td>0.4179</td>
</tr>
<tr>
<td>GFA 2-40-90</td>
<td>10</td>
<td>10000</td>
<td>0.2515</td>
<td>1.0138</td>
</tr>
</tbody>
</table>
4.3.3 Kneading elements

4.3.3.1 Influence of experimental setup on the torque on the screws

For the experiments with the kneading elements, the torque is significantly increased by a high loading of the extruder. The torque is not significantly changed by the staggering angle of the kneading elements or the viscosity of the used silicon oil. The model and its validity are shown in Figure 4.16.

![Figure 4.16: Left: A summary of fit for the model shown in Figure 4.16. R² (Coefficient of determination), Q² (Predictive relevance), model validity and reproducibility for the model are shown. Right: Scaled and centered coefficients for predicting the torque rate, experiments with kneading elements and high viscous silicon oil. Coefficients for the staggering angle of the kneading element (1), the viscosity of the used silicon oil (2) and the loading of the extruder (3) are shown.]

The closed system which is used to carry out the experiments might explain why the torque is not significantly influenced by the staggering angle of the kneading elements. The silicon oil which is processed by the kneading elements cannot flow away in this experimental setup, which probably causes a higher torque generation than in an open system. This effect will occur for both kneading elements (KB 5-2-30-30° and KB 5-2-30-90°), which makes it difficult to distinguish them based on the torque. In future research it might be helpful to investigate the torque in an open setup, to see possible differences between the various screw elements.

The viscosity of the used silicon oil does not have a statistically significant effect on the generation of torque. A higher viscosity is expected to cause a higher torque, but maybe the used viscosity range is too small to demonstrate this. Normally extrusion processes are
carried out with substances in a viscosity range 100 times higher (Ilo et al., 1996) than the one used in these experiments.

4.3.3.2 Influence of experimental setup on the pressure generation

From all the extrusion experiments with the kneading elements, there is only one in which a pressure is generated (Table 4.3). This is the experiment with a high loading of the extruder, using the high viscous silicon oil and the KB 5-2-30-30° kneading elements.

Table 4.3: Pressure results for the experiments with kneading elements

<table>
<thead>
<tr>
<th>Viscosity (mm²/s)</th>
<th>Pitch</th>
<th>Loading (g)</th>
<th>RATE Pressure(Mpa)/rpm</th>
<th>R² Pressure</th>
</tr>
</thead>
<tbody>
<tr>
<td>100000</td>
<td>90</td>
<td>10.2</td>
<td>1.214E-06</td>
<td>0.5807</td>
</tr>
<tr>
<td>10000</td>
<td>90</td>
<td>10.7</td>
<td>2.279E-06</td>
<td>0.8107</td>
</tr>
<tr>
<td>100000</td>
<td>90</td>
<td>28.0</td>
<td>2.837E-06</td>
<td>0.9444</td>
</tr>
<tr>
<td>10000</td>
<td>90</td>
<td>11.5</td>
<td>-9.159E-07</td>
<td>0.4785</td>
</tr>
<tr>
<td>10000</td>
<td>90</td>
<td>28.4</td>
<td>-9.918E-07</td>
<td>0.5677</td>
</tr>
<tr>
<td>10000</td>
<td>30</td>
<td>9.0</td>
<td>2.584E-06</td>
<td>0.9165</td>
</tr>
<tr>
<td>10000</td>
<td>30</td>
<td>28.9</td>
<td>2.069E-06</td>
<td>0.8926</td>
</tr>
<tr>
<td>100000</td>
<td>30</td>
<td>9.3</td>
<td>2.071E-06</td>
<td>0.9041</td>
</tr>
<tr>
<td>100000</td>
<td>30</td>
<td>28.8</td>
<td>3.049E-03</td>
<td>0.9781</td>
</tr>
<tr>
<td>100000</td>
<td>30</td>
<td>28.0</td>
<td>1.309E-03</td>
<td>0.9361</td>
</tr>
</tbody>
</table>

4.3.3.3 Influence of the experimental setup on the distributive mixing

The measured relative standard deviation for the content distribution of sodium chloride in silicon oil lies between 0.5939 and 1.6523. As in the experiments with the GFA elements, a high repetition error is observed, this is shown in Table 4.4.

Table 4.4: Results for content distribution of sodium chloride in silicon oil

<table>
<thead>
<tr>
<th>Screw element</th>
<th>Loading [g]</th>
<th>Viscosity [mm²/s]</th>
<th>RSD</th>
<th>RSD for repetition</th>
</tr>
</thead>
<tbody>
<tr>
<td>KB 5-2-30-30°</td>
<td>29</td>
<td>100000</td>
<td>0.9869</td>
<td>0.6766</td>
</tr>
<tr>
<td>KB 5-2-30-90°</td>
<td>10</td>
<td>10000</td>
<td>1.3002</td>
<td>1.9428</td>
</tr>
</tbody>
</table>

4.3.4 Mixing elements

Because only one mixing element is investigated, no statistical models can be proposed. Compared to the conveying elements and kneading elements, the rate between the torque and the screw speed is lower with the GFM 2-15-90 elements. This can be explained by the gaps in the flights of the GFM elements, which cause less shear stress between the screws.
and the barrel. No pressure is observed. The measured relative standard deviation for the content distribution of sodium chloride in silicon oil lies between 0.8546 and 1.8508. This is an unexpected high result because mixing elements are expected to provide a good distributive mixing. Probably the used method to measure the sodium chloride content distribution is inadequate, this can be demonstrated by the high repetition error (See Table 4.2 and Table 4.4 under sections 4.3.2.3 and 4.3.3.3).
5 CONCLUSIONS

From the results of the experiments, there could not be made a distinction between the distributive mixing properties of the different screw elements. A reason for this might be the used sampling method, namely a sampling at the top of the screws. The particle size reduction is not measured in this work, but the samples are stored and can be measured in a further study.

The used setup is appropriate for torque and pressure measurement. Based on the results of the performed experiments, it can be stated that the torque on the screws is increased by an increasing screw speed and an increasing loading of the extruder barrel. For the GFA elements, the torque is also significantly influenced by the viscosity of the substance inside of the extruder. This effect is not significant for the KB elements, which might be explained by the low viscosity range which is used in the experiments. The pitch or the staggering angle of the screws is not significantly influencing the torque on the screws in the used setup. This might be explained by the closed extruder system which is used to investigate the different screws.

For the conveying elements the highest pressure is measured with the GFA 2-15-90 elements, so it can be interpreted that these elements have the best transporting capacity. With most of the experiments with the KB elements no pressure was seen, which corresponds with a low conveying effect. Only in the experiment with the KB 5-2-30-30° elements and a high loading (28 g) of the high viscous silicon oil (100000 mm²/s), a measurable pressure is generated which implies that the KB 5-2-30-30° elements have conveying characteristics.

In future research it could be helpful to investigate the torque on the screws in an open system, using higher viscosity ranges, in order to observe the torque on the screws in a more realistic setup. The development of another method to investigate the distributive mixing properties of the screws is recommended.
6 LITERATURE


