Reduction of cure time in elastomer processing

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A program for calculating the temperature increase in rubber compounds during the injection phase was created and verified in experiments in course of the FFG-Bridge research project "Cure time reduction" at the Montanuniversität Leoben. Another tool helps the user to evaluate the potential cure time reduction at an early stage before process optimization. In a software-supported cure time reduction while securing the complete curing of the molded parts a significant cycle time reduction and minimized energy consumption can be achieved. The second objective of the project was to analyze the potential for cure time reduction using coupled shear and elongational heat generation in conical dies and predict this effect by means of the previously mentioned calculation program. Furthermore, the compression heating in rubber compounds was scientifically investigated for the first time.

1. Introduction

The FFG-Bridge research project "Cure time reduction" was initiated to investigate ways and means to minimize cure times during rubber processing without affecting adversely the final part properties. Special attention was paid to predict the potential for cure time reduction in advance by using coupled shear and elongational heating in a conical die, to the evaluation of dissipative heating in rubber compounds and to the development of a calculation program for the prediction of cure time reduction.

2. Characterization of shear and elongational heating

2.1 Calculation model for the coupled shear and elongational heating in conical dies

The analytical equations for the shear and elongational heating were derived from the energy equation. Thereby, the analytical approach was based on the methods established by Cogswell [1] and Binding [2]. In order to determine the flow field at the inlet, a simplification of the flow patterns known as lubrication approximation has been carried out. This approach was essential for an analytical solution of the problem without complex 3D simulation. Apart from the elongational deformation in the flow direction, the deformation in the radial and tangential direction also proved to be scientifically significant. The simplified energy **equation 1** was used for describing the dissipation in conical dies.

Due to the non-isothermal flow conditions, it would be almost impossible to find a continuous analytical solution for the temperature increase along the die. Therefore, the die was divided in segments and the temperature increase was summarized along the flow direction (in the segments), see **figure 1.**

One of the major conclusions to be drawn from the experimental studies came from the observation that dead volume was formed on the edges of the dies when the angles (θ) exceeded 45°. Hence the maximum external angle considered in the calculation was limited to 45°.

The temperature increase in the individual sections of the die is calculated with the help of **equation 2.**

Equation 1

$$\underbrace{\rho c_p\left(\frac{\partial T}{\partial t}\right)}_{a} = \underbrace{\tau_{zr}\left(\frac{\partial v_z}{\partial r}+\frac{\partial v_r}{\partial z}\right)}_{b} + \underbrace{\tau_{rr}\frac{\partial v_r}{\partial r}}_{c} + \underbrace{\tau_{\phi\phi}\frac{v_r}{r}}_{d} + \underbrace{\tau_{zz}\frac{\partial v_z}{\partial z}}_{e} \qquad (1)$$

a) Witnessed change of energy over time
b) Shear heating
c) Elongational heating in r-direction
d) Elongational heating in ϕ-direction
e) Elongational heating in z-direction

Equation 2

$$\Delta T_i = \Delta T_{Shear-i} + \Delta T_{r-Elongation-i} + \Delta T_{\phi-Elongation-i} + \Delta T_{z-Elongation-i} \qquad (2)$$

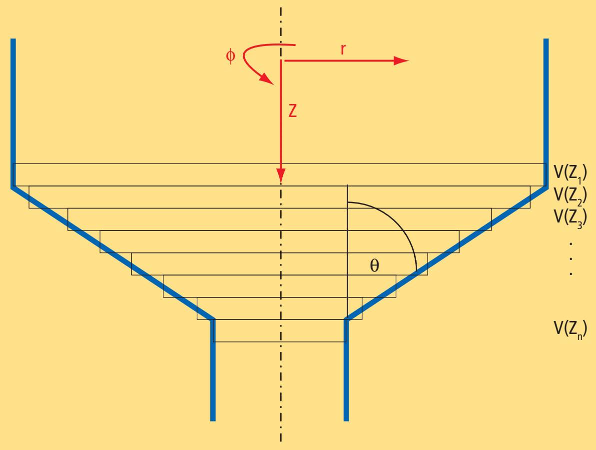

Fig. 1: Division of the die in small segments in order to simplify the calculation; figure from [3]

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In that context ΔT_i represents the total temperature increase in the segment, $\Delta T_{Shear\text{-}i}$ represents the shear contribution, $\Delta T_{r\text{-}Elongation\text{-}i}$, $\Delta T_{\Phi\text{-}Elongation\text{-}i}$ and $\Delta T_{z\text{-}Elongation\text{-}i}$ represent the elongational contributions in the directions r, Φ and z.

2.2 Model experiments for verifying the calculation model for shear and elongational heating

The objective of the experiments was to get a reproducible measurement of the bulk temperature after the flow through a conical die in order to verify the prediction of the calculation model presented in section 2.1. Bulk temperature measurements were carried out using a test mold which was specifically designed for that purpose. Six different conical nozzles were manufactured with varying die angles in the following steps: 20°, 30°, 45°, 60°, 75°, and 90°.

Figure 2 shows the temperature increase as a function of the die angle at various injection speeds (continuous lines). The hatched lines show the values obtained using the new "Calculation model by Perko" [3]. A high degree of correlation with an average error below 5 % could be found.

Conclusion: Substantial temperature increases of up to 70 °C were obtained in the experiments. The bulk melt temperature depends on the injection speed and the die angle within the range from 20° to 45°. A further increase of the die angle does not have an impact on the bulk temperature.

2.3 Developing a prototype for cure time reduction by means of conical dies

The preceding work investigated the combined shear and elongational heating in conical dies as well as the calculation in a new calculation model. For the implementation of these results, a prototype mold for cure time reduction has been developed **(fig. 3)**.

Test parts were produced under different processing conditions with this mold. Hardness (a), compression set (b) and tensile strength (c) were tested. Thereby, the compression set in the middle of the molded part proved to be a good indicator for the degree of cure at that position.

After a number of test series, the injection work has been proven to be the major impact factor for the temperature rise and on the achievable cure time reduction. **Figure 4** shows the compression set of parts produced with a uniform injection work and two different dies (die angles 20° and 60°).

The experiments with constant injection work showed no significant differences in the compression set when different dies were used. This implies that the injection work has the major influence on the temperature rise and consequently to the optimal curing time as well. Tests with different compounds as well as a variation of the die length showed similar results.

The results of this workpackage proved the dominating influence of the injection work on the curing time. Therefore, an optimization of the conical die geometry leads to no improvement of productivity. However, based on the results from this project, potential for cure time reduction can be identified and quantified more easily in industrial processes. In that context, the developed calculation programs provide substantial help.

Conclusion: The understanding of the rheological and thermodynamic phenomena during the injection phase could be improved substantially. It was possible to prove that the injection work is the main impact factor on the bulk temperature and the potential cure time reduction.

3. Cure time reduction using compression heating

3.1 Calculation model

The compression heating in rubber compounds was scientifically characterized for the first time in order to investigate the

Fig. 2: Bulk temperature as function of the die angle at different injection speeds; comparison of measurement and calculation; NBR test compound.

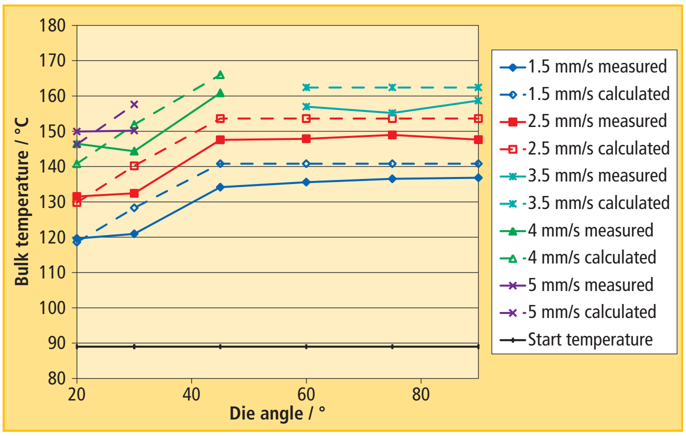
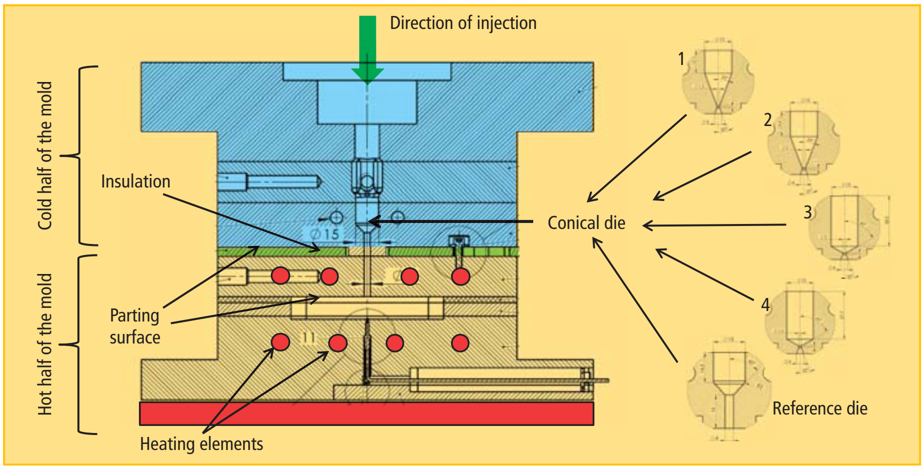


Fig. 3: Test mold for application of shear and elongational heating for cure time reduction [4]



basic principles for an application in cure time reduction within this project. Therefore, the compression heating as a function of the compression pressure had to be ascertained. This was done in two similar experiments using the capillary rheometer as well as the injection molding machine. A closed die featuring a thermocouple was used. The required compression pressure was applied using the piston of the capillary rheometer or the cylinder of the injection unit.

The compression heating was calculated according to the Joule-Thompson equation **(eq. 3):**

$$\Delta T = \frac{\alpha \cdot v}{c_p} \cdot \overline{T} \cdot \Delta p \qquad (3)$$

Figure 5 shows a good correlation of the measurements on the capillary rheometer and the injection molding machine to the calculation for one of the examined rubber compounds. The prediction of the temperature rises could therefore be classified as good. For a pressure of 1,800 bars the measured values for compression heating were around 10 °C.

3.2 Further tests and conclusions

Another specifically designed test mold was used for the evaluation of the potential for cure time reduction based on compression heating. For that purpose, a compression phase was integrated into the injection molding cycle with the purpose to reduce the incubation time of the material prior to the injection phase. Also for that purpose, a manually operated slider for closing the die was integrated into the mold.

After the determination of the optimal curing time for the respective compression conditions it was found that the compression phase in the cycle did neither improve the quality of the produced parts nor reduce the curing time. It was therefore concluded that the new processing strategy based on compression heating was not suitable for cure time reduction. This is caused by the relatively small temperature increase obtained which was only about 10 °C with 1,800 bars at the relatively low temperatures in the injection cylinder (approximately 70 to 100 °C). Furthermore, a high percentage of the heat generated gets lost to the walls of the injection unit which have a slightly lower temperature.

4. Calculation program for estimating the cure time reduction

A new calculation program model was developed for the estimation of the cure time and the potential reduction of the same. Within that program, the heat transfer from the mold wall to the rubber compound is determined using one-dimensional finite differences for the solution of the energy equation. For that purpose, the rubber part is divided into parts in the thickness-direction. Based on this, an estimation of the potential cure time reduction by increasing the bulk temperature during injection was made. The obtained results are shown in **figure 6.**

The curing time for molded parts at varying mold-wall temperatures (T_w) and varying starting temperatures (T_0) after injection (temperature of the rubber compound mass at the entry of the mold tool) is shown as a function of part thickness.

The starting temperature was first assumed at a theoretical minimum of 90 °C. This corresponds to a very slow injection speed with almost no temperature rise. The higher starting temperature of 150 °C was chosen according to the potential for temperature rise measured at the preceding tests **(fig. 2).** The results show that it is not possible to reduce curing times for parts with a thickness below 4 mm.

It can be concluded that the potential for cure time reduction (difference between the red curve and the blue curve in **figure 6)** for a given wall thickness increases with the mold wall temperature (T_w) and the thickness of the part.

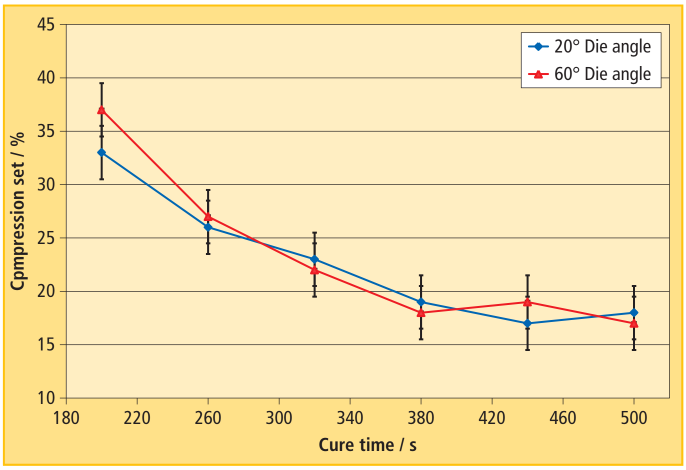


Fig. 4:
Results of injection molding experiments with constant injection work; SBR test compound [5].

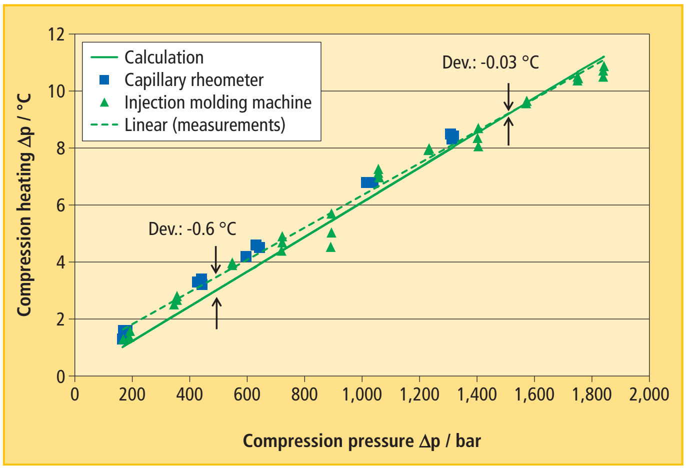


Fig. 5:
Measured compression heating on capillary rheometer and injection molding machine; comparison between measurement and calculation; EPDM test compound with initial temperature of 80 °C.

5. Additional findings

Within the framework of this project, new knowledge of practical importance for elastomer processing was gained in five different fields:

5.1 Temperature correction of shear viscosity data from capillary rheometry

Caused by the high viscosity of rubber compounds, significant temperature rises can occur in the capillary rheometer as a result of dissipative heating. As the rheological evaluation presumes stationary isothermal flow conditions it proved necessary to correct the ascertained viscosities for dissipative heating. A couple of methods for the correction of shear heating [6, 7] already exist which are not used systematically in practice however. The elongational heating in the inlet of the capillaries is usually not corrected. Within the framework of this project a new method of temperature correction for the measurement of improved viscosity data has been developed, validated and applied to solve practical problems.

5.2 Development of a new method for measuring the elongational viscosity

The required elongational viscosity data from the capillary rheometer test was planned to be determined using the established methods from Cogswell [1], Binding [2] or Obendrauf [8]. However, these methods have failed to provide a sufficient description of the flow phenomena occurring in the highly viscous rubber compounds. A new model for the evaluation of the capillary rheometer data for elongational viscosity was developed as well. This model provides a sound contribution to the understanding and the transparency of elongational flows which frequently occur during processing thus making these easier to calculate.

5.3 Method for visualizing converging flows

In order to visualize the flow geometries occurring at changes of cross-sections, a method for visualizing these converging flows was developed. This allowed a broader understanding of these phenomena which was necessary for developing the calculation models mentioned above improving the precision of predictions regarding temperature rises. This practical knowledge can be used to improve the design of injection molding machines dies and runners.

5.4 Program for estimating curing times and the potential for reducing the same

The injection molding experiments carried out proved the potential for cure time reduction using shear and elongational expansion heating. Furthermore a calculation program for estimating the cure time (cure time calculator) for simple part geometries was developed and tested. Using this tool it is possible to forecast the curing degree of rubber parts and to estimate the required cure time.

6. Summary and outlook

In order to achieve the goals set a model for calculating the flow in conical dies was developed based on knowledge derived from the scientific literature. Simultaneously the measurement of the corresponding material data was carried out and the state of the art regarding the methods for measuring shear and elongational viscosity on the capillary rheometer was improved. The verification of the bulk temperature calculation was carried out on a rubber injection molding machine type Maplan MTF750/160 edition. Furthermore the compression heat in rubber compounds was characterized. Results showed a high level of correspondence with the results of the calculations using the Joule-Thompson equation. Based on that knowledge a corresponding procedure for rubber injection molding using compression heating was developed and tested in practice.

In the field of shear and elongational heating, a prototype mold was developed and tested on a Maplan rubber injection molding machine. In doing this, new practically applicable knowledge regarding the injection molding process could be acquired. However, it was discovered that the achievable cure time reduction mainly depends on the injection work. This means that the die geometry, provided it has the required flow resistance to apply the necessary injection work, has little impact on reducing the cure time. For that reason it was decided to refrain from the time-consuming optimization of the geometry of the conical dies. It was decided instead to set focus on a substantial improvement of the calculation model for the bulk temperature prediction (under consideration of the axial, radial and tangential components of the converging flow).

7. Acknowledgments

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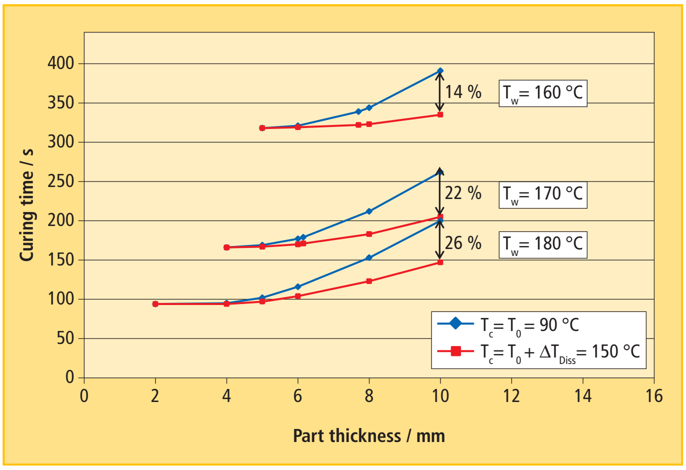

Fig. 6:
Estimation of the potential cure time reduction by increasing the injection speed

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New platform for European rubber research

National rubber associations and several benchmark rubber laboratories established **ERRLAB – European Research + Rubber Laboratories,** a new network of laboratories for R+D and testing in rubber applications. The involved national rubber associations are:

- **Assogomma,** Italy
- **Syndicat National du Caoutchouc et des Polymères (SNCP),** France,
- **Wirtschaftsverband der deutschen Kautschukindustrie e. V. (wdk),** Germany.

They are supported by the **European Tyre and Rubber Manufacturers' Association (ETRMA)** and the following rubber laboratories:

- **Deutsches Institut für Kautschuktechnologie e. V. (DIK),** Germany,
- **Laboratoire de Recherche et de Contrôle du Caoutchouc et des Plastiques (LRCCP),** France,
- **Laboratorio per la Certificazione e Ricerca sui Sistemi Elastomerici (Cerisie),** Italy.

Spokesperson of ERRLAB is Frenchman **Christian Caleca** (CFCP), and **Prof. Ulrich Giese** (DIK) from Germany is the acting scientific liaison. The role of scientific liaison rotates annually among the participating institutions.

"With the now established network of German, French and Italian rubber research institutions an important step toward strengthening of the European rubber industry has been completed", said **Boris Engelhardt,** Managing Director of the wdk, on occasion of the founding of the research platform ERRLAB in Brussels on 26 January 2015. "With this research network, companies in the European rubber industry now have access to the expertise of more than 100 researchers and scientists. This gives a good foundation for companies to meet future technical challenges and face global competition," said Engelhardt. He is convinced that the bundling of research with development capabilities and capacities for research and testing will lead to a significant improvement in the performance of the participating research institutions. Engelhardt added: "This multinational network also gives the opportunity to actively participate in EU research programmes."

"ERRLAB is also a good example and important signal of German-Franco-Italian cooperation of rubber associations – a collaboration with potential," Engelhardt concluded.

F. l. t. r.: Prof. Ulrich Giese (DIK), Fabio Bertolotti (Assogomma), Boris Engelhardt (wdk), Christian Caleca (SNCP), Fazilet Cinaralp (ETRMA), Claude Janin (LRCCP), and Fabio Negroni (Cerisie)