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Experimental and numerical analyses of the texturisation process of a viscoelastic protein matrix in a cooling die after high moisture extrusion cooking

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Texturisation, residence time distribution, viscoelasticity, high moisture extrusion cooking, CFD

Abstract

A few years ago, meat substitutes made of plant proteins were still regarded as a niche product, nowadays the sales of these products are constantly increasing. Particularly flexitarians value the meat-like products (here: extrudate) in taste and texture to the traditional meat products. The products with the most meat-like texture are produced by high moisture extrusion cooking. During this process, a co-rotating twin-screw extruder is used, with the matrix being discharged through a long-drawn, double-walled cooling die.
The solidification by cooling and simultaneous texturing of the matrix takes place in the cooling die. The exact structuring process is not sufficiently understood and dependents on numerous factors, including the composition of the raw material and the processes in the extruder barrel as well as in the cooling die.
Experiments of coloring the matrix and determinations of the residence time distribution were done and allow an analysis of the flow fields. The experimental results are supported by numerical investigations. Thus, coloring tests can also be reproduced in numerical simulations and are consistent with the results of the experiments. As a result, shear rate in the cooling die can be identified as the main influencing factor of the structuring process.

1. Introduction

Extrusion cooking is a continuous thermomechanical process with multistep or multifunction operations. Thus, extrusion cooking sequentially leads to (1) mixing, (2) shear, (3) homogenization, (4) compression, (5) temperature and pressure build-up (Cheftel et al. 1992).
High moisture extrusion cooking is a subunit of extrusion cooking containing 40 % to 80 % water and represents a novel and useful technique in the field of texturisation to produce meat substitutes. Due to the growing demand for food proteins because of the growing world population and changes in dietary habits new food technology concepts are required. From an ecological and nutritional physiological point of view, the use of plant proteins represents a possible substitution of animal protein (Smil 2000). In addi-

tion to vegetarians, meat products are also referred to as so-called flexitarians who want to reduce the consumption of meat without renouncing this completely. They consider to be particularly important that meat-like products correspond in taste and texture to traditional meat products (Hoek et al. 2011). Therefore, extrusion cooking at high moisture contents offers considerable potential for the production of vegetable meat analogues that resemble the product properties of muscle meat. Due to mechanical and thermal stresses during the extrusion process proteins are altered in their native structure; denaturation and changes in their molecular structures occur (Liu and Hsieh 2007; Chen et al. 2011; Osen et al. 2015).

At the end of the extruder a long-drawn, double-walled cooling die is attached through which the protein matrix being discharged. In this cooling die an anisotropic fibre-like structure is developed (Arêas 1992; Cheftel et al. 1992). Additionally, the cooling die prevents expansion of the matrix. Until now the exact structuring process is not sufficiently understood and dependents on numerous factors, including the composition of the raw material and the processes in the extruder barrel as well as in the cooling die. Through the solidification in the cooling die, the extrudate preserves fluid-mechanical stresses in its layered structure and shows a clearly defined, three-dimensional orientation, which is partly due to shear stresses during cooling; thus, the thermomechanical treatments influence the texture quality of the final product, leading to a process window to produce meat substitutes with different texture characteristics (Ilo and Berghofer 2003).

Several studies have focused on the high moisture extrusion cooking process to understand the effect of barrel temperature and moisture content on final product properties using soy and pea protein (Chen et al. 2010; Chen et al. 2011; Fang et al. 2013; Lin et al. 2002 ; Liu und Hsieh 2007; Osen et al. 2014). However, the influence of the cooling die on the texturisation of a viscoelastic protein matrix has not yet been investigated. Therefore, we investigated the solidification in the cooling die on the formation of a meat-like product based on soy protein and linked experimental studies with numerical simulation. Numerical investigations can determine local distributions of shear rates as well as temperature distributions in the viscoelastic protein matrix. Due to systematical variation of non-determinable rheological parameters and iterative comparison with experimental data, (i) the role of the yield stress as well as (ii) the solidification models can be parameterized for practical application (Delgado et al. 2008; George et al. 2014).

2. Material and methods

2.1. Materials

Commercial soy protein concentrate (ALPHA® 8) was kindly provided by DuPont de Nemours (Germany) GmbH (Neu-Isenburg, Germany). Refill Ink (Light Cyan) for coloring the matrix was kindly supplied by vhbw - B & W Handelsgesellschaft mbH (Kamenz, Germany).

To simulate the solidification during high moisture extrusion cooking different material parameters have been used from the literature (see Table 1).

Table 1: Material parameters for the viscoelastic protein matrix

Parameter	Value	Unit	Comment
Moisture content X_W	0.60	-	Experimental studies
Molar mass	2516	kg/mol	Fang et al. 2013
Density ρ	1078.59	kg/m^3	Calculated based on mass flow rate of 13 kg/h and velocity of 0,0062 m/s:
Specific heat capacity c_v	3241,5	J/(kg K)	$c_v = 1480 + 2710 \cdot X_W$ (Heldman and Singh 1981)
Thermal conductivity λ	0.4296	W/(m K)	$\lambda = -0.228 + 0.000249\rho + 1.304X_W - 0.926X_W^2$ (Wallapapan 1984)

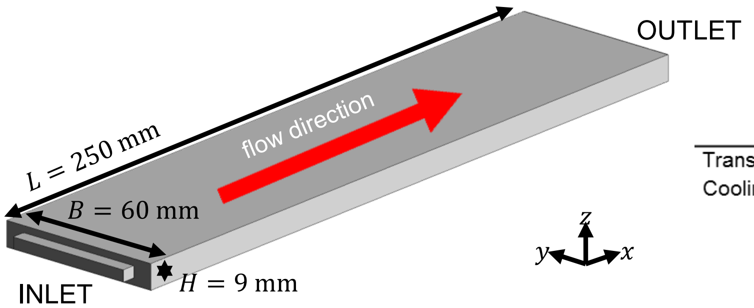
2.2. Experimental studies

2.2.1. High moisture extrusion cooking

Experiments of high moisture extrusion cooking were performed using a co-rotating twin screw extruder ZSK 25 (Coperion, Stuttgart, Deutschland) with a screw diameter of $25\ \mathrm{mm}$ and a length–diameter ratio of 28.8. The extruder barrel is segmented into six temperature-controlled zones which were heated at constant temperature by an oil-operated temperature control system (SINGLE Temperiertechnik GmbH, Hochdorf, Germany), except of the first two segments. These segments were tempered with tap water at ambient temperature. The dry protein concentrate was fed into the first section of the extruder by a gravimetrically controlled twin screw feeder K-Tron T22 (Coperion K-Tron GmbH, Niederlenz, Switzerland) and tap water was added into the second extruder segment by a diaphragm metering pump (LEWA ecoflow ®, LEWA GmbH, Leonberg, Germany). For the trails, a screw profile with low shear intensity was used.
The experimental studies were run at constant screw speed of $400\ \mathrm{U\ min^{-1}}$, feed rate of $13\ \mathrm{kg\ h^{-1}}$, barrel temperature of $140\ ℃$ and moisture content of $60\ \%$ (wet basis). Material temperature as well as extruder pressure were measured at extruder exit, directly before the inlet of the cooling die, using a thermocouple type J (Voltcraft, Wollerau, Switzerland) and a melt pressure sensor MDT422 (Dynisco Instruments, LLC, Franklin, Massachusetts, USA), respectively.

2.2.2. Cooling die

Texturisation of the protein matrix was conducted by attaching a transition plate and a long cooling die to the extruder (see Table 1).



	length [mm]	width [mm]	height [mm]
Transition plate	5	45.74	4.1
Cooling die	250	60	9

Figure 1: Schematic illustration of the transition plate and cooling die used to study the effect of texturisation on a viscoelastic protein matrix.

The cooling temperature was set to $40°C$ by an air-cooled thermostat (LAUDA DR. R. WOBSER GMBH & CO. KG, Lauda-Königshofen, Germany) and tap water functioned as coolant. The pressure gradient in the cooling die was measured using five melt pressure sensor MDT422 (Dynisco Instruments, LLC, Franklin, Massachusetts, USA) at a distance of $0.03\ \mathrm{m}$; $0.06\ \mathrm{m}$; $0.12\ \mathrm{m}$; $0.18\ \mathrm{m}$ and $0.24\ \mathrm{m}$ along the cooling die. Additionally, the temperature gradient was detected measuring the temperatures at a distance of $0.05\ \mathrm{m}$; $0.14\ \mathrm{m}$ and $0.23\ \mathrm{m}$ along the cooling die using thermocouples type J (Voltcraft, Wollerau, Switzerland). The positions of the pressure senores were in the middle of the die plate. However, the thermocouples were positioned $0.01\ \mathrm{m}$ outwards from the center.

2.2.3. Determination of velocity fields and residence time distribution

To analyse the velocity fields in the cooling die experiments of coloring the protein matrix and determinations of residence time distributions were done enabling an optical detection. For coloring the protein matrix and thus for measuring the residence time distribution (RTD) refill ink was used as colorant, since ink was determined to have no significant influence on texturisation. For the coloring tests ink was added for two seconds into the last extruder segment with a constant flow rate of $10\ \mathrm{ml\ min^{-1}}$ by a high-pressure pump (KNAUER Wissenschaftliche Geräte GmbH, Berlin, Germany). Dead stops were performed to determine the flow field in the cooling die.
Colored samples were collected and immediately stored in sealed airtight plastic bags at $-20\ °\mathrm{C}$. For analysis, the frozen samples were cut in the middle ($z = 0.0045\ m$) parallel to the flow direction by an electrical kitchen slicer (Gebr. Graef GmbH & Co. KG. Arnsberg, Germany).

2.3. Numerical studies on high moisture extrusion cooking

Inverse modeling is initially based on numerical simulations of the flow and temperature field of a single-phase, non-rigid viscoelastic protein matrix. By comparing the numerical and experimental results, a direct validation is carried out. The inverse modeling assumes the boundary conditions to be implemented by the experimental data. The commercial Software ANSYS CFX (ANSYS, Inc., Canonsburg, Pennsylvania, USA) is used for the numerical studies.
To simulate the solidification of a viscoelastic protein matrix, different boundary conditions are set and listed in Table 2.

Table 2: Type of applied boundary condition

	Velocity	**Pressure**	**Temperature**
Inlet	mass flow rate	zero gradient	fixed temperature
Outlet	zero gradient	fixed pressure	zero gradient
Wall	slip condition	zero gradient	heat transfer coefficient

The heat transfer coefficient describes the boundary condition at the die wall and is determined by measuring the temperature difference of the extrudate between die inlet and outlet as well as the heat flux during the experimental studies:

$$\alpha = \frac{\dot{Q}}{A\,\Delta T}$$

As the second boundary condition at the wall a slipping condition has been defined which allows a slipping of the extrudate along the die wall starting above a critical shear stress τ_c:

$$u_{ws} = U_s \left(\frac{\tau_w - \tau_c}{\tau_n} \right), \qquad \tau_w > \tau_c$$

Herschel-Bulkley Model with Arrhenius term (see Extrusion Cooking, p. 216) was used as the flow function with a yield stress τ_0:

$$\tau = \tau_0 + k\dot{\gamma}^n, \qquad k = k_0 \exp\left(\frac{A}{T}\right)$$

In total, four parameters (τ_0, n, k_0, A) from the flow function and another three parameters (U_s, τ_c, τ_n) from the wall slipping condition must be determined. Table 3 shows the assumed values for these seven parameters:

Table 3: Assumed values for the numerical studies

Parameter		Value	Unit
slip speed	U_s	0,007	mm $s^{(-1)}$
normalizing stress	τ_n	100	Pa
critical stress	τ_c	18000	Pa
yield stress	τ_0	1000	Pa
flow index	n	0.34	-
consistency parameter (adjusted)	k_0	0.79	-
Arrhenius parameter	A	4000	

3. Results and discussion

To assess non-measured material parameters, the numerical simulation is compared with experimental studies and adapted to experimental data. The numerical simulations shown below are based on the process variables (see Table 4) form the experimental studies.

Table 4: Process variables of the high moisture extrusion cooking

Process and material parameters		Value	Unit
moisture content	X_W	0.60	—
temperature at extruder output (direcly before die inlet)	T_{in}	116.4	℃
maximal temperature at die outlet	$T_{\mathrm{M,out}}$	102.1	℃
mass flow rate	$\dot{m}$	13	$\mathrm{kg\,h^{-1}}$
specific heat capacity	c_p	3106	$\mathrm{J\,kg^{-1}\,K^{-1}}$
thermal conductivity	λ	0.4296	$\mathrm{W\,m^{-1}\,K^{-1}}$
heat transfer coefficient	h	436.79	$\mathrm{W\,m^{-2}\,K^{-1}}$

The extrusion of a viscoelastic protein matrix shows that the solidification takes place from the walls of the cooling die towards the center. Thus, the extrudate becomes solid from the outside to the inside. The material temperature at extruder exit, directly before the die inlet, was measured at $116.4\,℃$. The maximal measured temperature in the core of the extrudate at die outlet was $102.1\,°C$. For the numerical studies, the above-mentioned measured input temperature was set as boundary condition at the inlet. The maximum material temperature at the outlet of the cooling die resulted in a temperature of $100.4\,°C$ and corresponds to the maximal measured material temperature of the experiment. During high moisture extrusion cooking temperature gradient as well as

pressure gradient along the cooling die was recorded. These experimental data were compared with the numerical studies and the results are shown in Figure 2.
Comparing the temperature gradients of the experiment with the simulation reveals slightly higher temperatures in the numerical studies. However for the pressure gradient, the numerical simulations and the experimental results were highly concordant. Therefore, the rheological data of the viscoelastical protein matrix must be determined and implemented to optimise the temperature gradient of the numerical simulation.

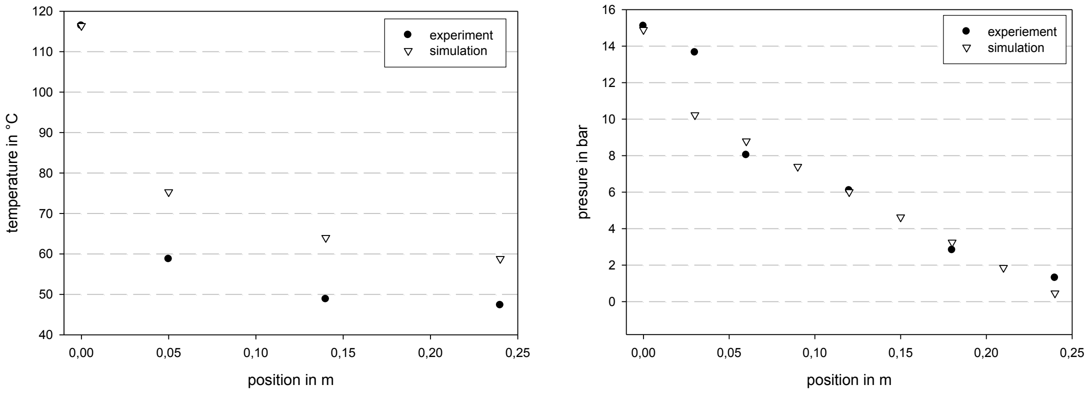


Figure 2: Measured temperature gradient along the cooling die during high moisture extrusion cooking compared to the calculated temperature gradient along the cooling die in numerical studies (left). Measured pressure gradient along the cooling die during high moisture extrusion cooking compared to the calculated pressure gradient along the cooling die in numerical studies (right).

Furthermore, Figure 3 illustrates the velocity profile perpendicular to the flow direction at different distances from the inlet.
The slipping velocity increases along the cooling die from $0.4\ \mathrm{mm\,s^{-1}}$ at $x = 0.01\ \mathrm{m}$ to $6.0\ \mathrm{mm\,s^{-1}}$ at $= 0.2\ \mathrm{m}$. The approach to the mean velocity leads to a reduction of the maximum speed in the center of the cooling die. Additionally, the influence of the yield stress and the increase of viscosity due to a decrease in temperature results in an almost constant speed. As the viscosity increases, the effect of the yield stress becomes smaller.

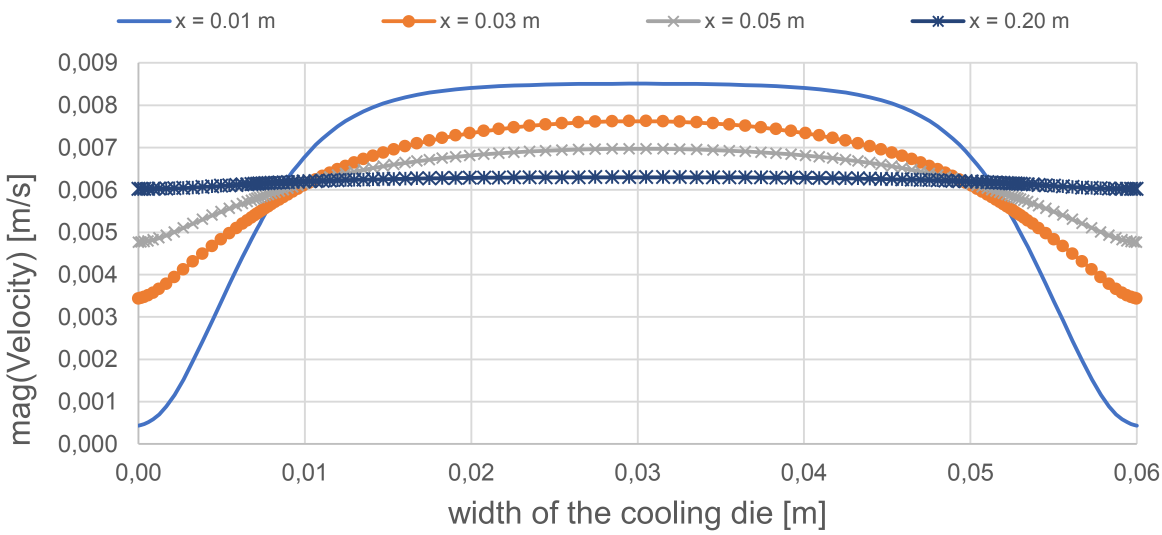


Figure 3: Velocity profile along the cooling die.

During experimental studies, the protein matrix in the cooling die was colored to determine the flow profile. Afterwards the colored extrudate was cut in half and a picture was taken. The colored sample that was taken out of cooling die after dead stop is shown in Figure 4.

Figure 4: Picture of the colored extrudate to determine the flow profile during high moisture extrusion cooking experiments. The sample was cut in the middle ($z = 0.0045\ m$) parallel to the flow direction (on top). The sample was cut lengthwise and perpendicular to the flow direction (below).

The blue color indicates the flow profile in the cooling die in Figure 4. The matrix was colored twice within 10 seconds. The dead stop was performed 35 seconds after the first staining. According to Figure 4, a change of the profile along the cooling die cannot be clearly derived.
The color tests can be simulated in numerical studies by calculating streamlines (in steady state flow equivalent to particle paths), see Figure 5. The stationary flow profile is taken as the basis and the streamlines are performed by calculating the distance a particle has traveled in a corresponding time. Additionally, the streamlines were plotted in xz-plane to determine the effect of the shear rate on the solidification.

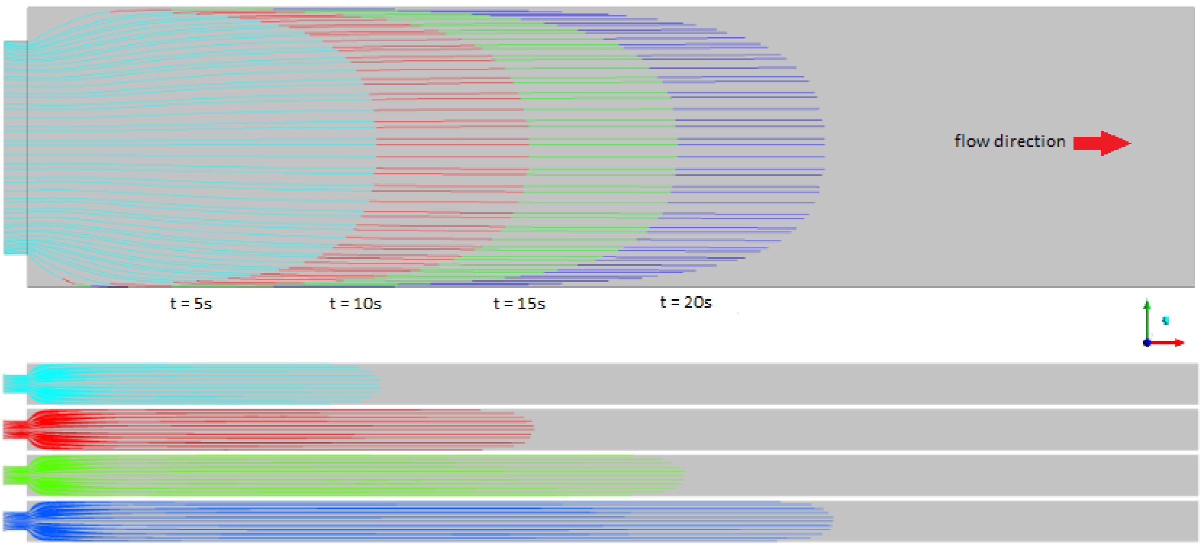


Figure 5: Streamlines after 5 s (light blue), 10 s (red), 15 s (green) and 20 s (blue) in x-direction on xy-plane (on top). Resultant streamlines after 5 s (light blue), 10 s (red), 15 s (green) and 20 s (blue) in x-direction on xz-plane (below).

By comparing the flow profiles of the experimental studies (see Figure 4) with the streamlines of the numerical investigation (see Figure 5), the simulation clearly shows a change of the profile along the cooling die. Thus, the tip of the profiles become tapered along the cooling die and the experiments of coloring the protein matrix are consistent with the results of numerical investigations. Regarding the flow profiles in x-direction on xy-plane as well as the streamlines in x-direction on xz-plane, the extrudate shows a clearly defined, three-dimensional orientation, which is partly due to shear stresses during cooling and the extrudate preserves fluid-mechanical stresses in its layered structure. Thus, conclusions can be drawn about fluid-mechanical fields even after the high moisture extrusion cooking process.

In the further approach of the inverse modelling, rheological variables (e.g. yield stress) should be implemented for describing temperature and shear dependent solidification within the cooling die.

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Literatur

Arêas, J. A. (1992): Extrusion of food proteins. In: *Critical reviews in food science and nutrition* 32 (4), S. 365–392. DOI: 10.1080/10408399209527604.

Cheftel, J. C.; Kitagawa, M.; Quéguiner, C. (1992): New protein texturization processes by extrusion cooking at high moisture levels. In: *Food Reviews International* 8 (2), S. 235–275. DOI: 10.1080/87559129209540940.

Chen, Feng Liang; Wei, Yi Min; Zhang, Bo (2011): Chemical cross-linking and molecular aggregation of soybean protein during extrusion cooking at low and high moisture content. In: *LWT - Food Science and Technology* 44 (4), S. 957–962. DOI: 10.1016/j.lwt.2010.12.008.

Chen, Feng Liang; Wei, Yi Min; Zhang, Bo; Ojokoh, Anthony Okhonlaye (2010): System parameters and product properties response of soybean protein extruded at wide moisture range. In: *Journal of Food Engineering* 96 (2), S. 208–213. DOI: 10.1016/j.jfoodeng.2009.07.014.

Delgado, A.; Rauh, C.; Benning, R. (2008): Thermodynamisches Modell zum Fest/flüssig-Phasenübergang von Substanzen hohen molaren Volumens. In: *Chemie Ingenieur Technik* 80 (8), S. 1185–1192. DOI: 10.1002/cite.200800075.

Fang, Yanqiang; Zhang, Bo; Wei, Yimin; Li, Shujing (2013): Effects of specific mechanical energy on soy protein aggregation during extrusion process studied by size exclusion chromatography coupled with multi-angle laser light scattering. In: *Journal of Food Engineering* 115 (2), S. 220–225. DOI: 10.1016/j.jfoodeng.2012.10.017.

George, Paul; Lundin, Leif; Kasapis, Stefan (2014): Effect of thermal denaturation on the mechanical glass transition temperature of globular protein/co-solute systems. In: *Food Hydrocolloids* 41, S. 156–163. DOI: 10.1016/j.foodhyd.2014.04.018.

Harper, J. M. (1989). Food extruders and their applications. In C. Mercier, P. Linko & J. M. Harper: Extrusion Cooking. American Association of Cerial Chemists, St. Paul.

Heldman and Singh 1987. In C. Mercier, P. Linko & J. M. Harper: Extrusion Cooking. American Association of Cerial Chemists, St. Paul.

Hoek, Annet C.; Luning, Pieternel A.; Weijzen, Pascalle; Engels, Wim; Kok, Frans J.; Graaf, Cees de (2011): Replacement of meat by meat substitutes. A survey on person- and product-related factors in consumer acceptance. In: *Appetite* 56 (3), S. 662–673. DOI: 10.1016/j.appet.2011.02.001.

Ilo, S.; Berghofer, E. (2003): Kinetics of Lysine and Other Amino Acids Loss During Extrusion Cooking of Maize Grits. In: *Journal of Food Science* 68 (2), S. 496–502. DOI: 10.1111/j.1365-2621.2003.tb05701.x.

Lin, S.; Huff, H. E.; Hsieh, F. (2002): Extrusion Process Parameters, Sensory Characteristics, and Structural Properties of a High Moisture Soy Protein Meat Analog. In: *J Food Science* 67 (3), S. 1066–1072. DOI: 10.1111/j.1365-2621.2002.tb09454.x.

Liu, Ke Shun; Hsieh, Fu-Hung (2007): Protein–Protein Interactions in High Moisture-Extruded Meat Analogs and Heat-Induced Soy Protein Gels. In: *J Amer Oil Chem Soc* 84 (8), S. 741–748. DOI: 10.1007/s11746-007-1095-8.

Osen, Raffael; Toelstede, Simone; Eisner, Peter; Schweiggert-Weisz, Ute (2015): Effect of high moisture extrusion cooking on protein-protein interactions of pea (Pisum sativum L.) protein isolates. In: *Int J Food Sci Technol* 50 (6), S. 1390–1396. DOI: 10.1111/ijfs.12783.

Osen, Raffael; Toelstede, Simone; Wild, Florian; Eisner, Peter; Schweiggert-Weisz, Ute (2014): High moisture extrusion cooking of pea protein isolates. Raw material characteristics, extruder responses, and texture properties. In: *Journal of Food Engineering* 127, S. 67–74. DOI: 10.1016/j.jfoodeng.2013.11.023.

Wallapapan 1984, In C. Mercier, P. Linko & J. M. Harper: Extrusion Cooking. American Association of Cerial Chemists, St. Paul.