

## **Extended Summary**

“Untersuchungen zur Herstellung und Optimierung proteinogener Biowerkstoffe”  
 (“Studies on the production and optimization of protein-based biopolymers”)

### **1. Summary**

The aim of the present work “Studies on the production and optimization of protein-based biopolymers” is the development of a continuous manufacturing process for new bio-based products, the optimization of their functional properties and the addition of value as a contribution to a sustainable customer acceptance and market success. Therefore, a production design based on the extrusion technology was established, optimized and characterized. Furthermore, strategies for the modification of selective material properties via enzymatically catalyzed cross-linking of proteins and crystallization of additives were presented and evaluated. This study provides an integral process-based approach for a highly efficient production of natural polymers based on proteins that ensures economic and environmental sustainability. In conjunction with an innovative and tailor-made product design, exclusively adapted from renewable resources, the obtained results are an important contribution to a mass-market commercialization of environmentally friendly products.

### **2. Problem addressed**

The material use of biomass is of great importance due to their contribution to climate change mitigation, environmental protection and saving of fossil resources [1, 2]. Therefore, a new line of product have recently become of great interest in industry and society – bio-based polymers, respectively biopolymers.

In particular, protein-based biopolymers are considered as promising raw materials for a wide range of industrial products. Due to their unique set of properties and zero environmental impact they are highly applicable in market sectors such as packaging, agriculture, automotive, electronics, medicine and pharmacy [3, 4].

However, insufficient benefits in costs due to the absence of economies of scale, inadequate functional properties of the final products and a conclusive customer value in terms of an additional product benefit are limitative factors for a mass-market commercialization. To solve these problems, product and process design approaches were applied.

### **3. State of the Art**

Renewable resources as industrial feedstock for the production of chemical substances, such as oil, starch and cellulose received much interest, while the market growth of bio-based polymers, particularly protein-based biopolymers is comparatively low. This can be explained with unsatisfactory performances of the product, high prices, insignificant communication efforts and unconvincing additional

values [5, 6, 7]. Today, a strongly increasing interest in research activities concerning biodegradable polymers can be recognized with the aim to overcome the mentioned limitative factors. State of the art technologies, like extrusion and injection molding technologies were modified and optimized to meet the specific requirements of natural materials. Furthermore, the optimization and customization of natural polymers are in the focus of science and industry paired with the development of persuasive added values for market success. Several studies showed the potential of thermoplastic processing of proteins for the large scale-production of polymeric products [8, 9, 10, 11]. Coevally, present research dealing with enzymatic modification of the protein show remarkable results in terms of optimization the functional properties [12, 13, 14, 15].

The present work “Studies on the production and optimization of protein-based biopolymers” is successfully following up these research activities by focusing on the production technology (engineering), the product design (improvement of functional properties) and the addition of value for a high customer acceptance.

#### **4. Key innovations**

One of the key innovations of the present work is related to the development and implementation of a continuous manufacturing process including an inline enzymatically catalyzed reaction for the efficient large-scale production of highly customizable protein-based films and tapes. Furthermore, new protein-based polymers with a unique and controllable set of properties were developed by using 100 % renewable resources that are completely biodegradable, compostable, biocompatible and non-toxic. Finally, by incorporating crystallizing additives into the protein matrix during the manufacturing process, the functional properties (especially the mechanical properties) of the final products were enhanced significantly. In conjunction with the improvement of the material properties, a depot and release functionality was succeeded which allows the addition of functional additives (e.g. agrochemicals or pesticides in protein-based mulching films and active pharmaceutical ingredients in protein-based capsules, coatings and films). Thus, together with a time-controlled biodegradability and the resulting release of the inclosed additives lead to an unmatched functional product design with a desirable satisfaction of customer’s needs.

**The key innovations of the present work can be highlighted as follows:**

- Developing of new biopolymers (films, sheets, tapes and coatings) made of proteins with a unique set of properties which are completely biodegradable, compostable, biocompatible and non-toxic

- Pre-commercial development of protein-based mulching films and seed tapes
- Exclusive use of renewable resources (100 % BIO)
- Developing and implementation of a continuous manufacturing process based on the extrusion technology which is highly energy efficient due to optimized operation parameters
- Successful feasibility study and small-scale production of protein-based films based on the traditional extrusion technology
- Very successful integration of an enzymatically catalyzed cross-linking reaction in the extrusion process
- Inline process modification of selective properties via enzymatic cross-linking of proteins resulting in tailor-made products and unrivaled product design which meet the growing demand for environmentally-friendly products in market sectors such as packaging, agriculture, electronics, medicine, pharmacy and cosmetics
- Incorporation of crystallizing additives to enhance the mechanical properties of the protein materials significantly and to add additional benefit to the final products in terms of functional compliance and active ingredients
- Developing of innovative materials acting as depot (carrier) and release systems for functional additives like pharmaceutical ingredients, fertilizers, pesticides (e.g. protein-based mulching films which can be ploughed into the soil after plant cultivation and degrade rapidly in contrast to conventional plastics - they have to be discarded or recycled which needs big efforts in terms of costs and time. An additional benefit is offered by a controlled release due to added crystalline agrochemicals, fertilizers, pesticides from protein-based mulching film)
- Time-controlled biodegradability and resulting release functionality using product and process design approaches
- The combination of inner-film crystallized additives and enzymatic cross-linking showed a multitude of possibilities for optimizing material properties of protein-based films and coatings, e.g. improvement of the tensile strength and elongation (synergistic effect of crystalline additives and cross-linking enhances physicochemical properties)
- In summary, an integral process-based approach for a cost efficient production and optimization of customizable protein films that lead to higher throughput rates with a high standard of quality. This ensures economical and environmental sustainability with a reasonable satisfaction of needs

## **5. Applications, implementations and results**

The product and process innovations of the present work are an important basis for an economical large-scale production of bio-based polymers with excellent functional properties, high rate of customer satisfaction and unmatched sustainability.

There is a broad field of application for the designed products, ranging from agricultural and gardening use (mulching films, greenhouse cover, seed coatings and seed tapes), packaging solutions (films, foils, interlayers and plates) to medical and pharmaceutical products (absorbable implants, encapsulations, active ingredient depots) and beauty treatments by using protein materials as natural cosmetics (e.g. face mask). Primarily, short-term applications due to the biodegradability of the natural products are in the focus of interest. Selective self-developed products are shown in [Fig. 1](#).

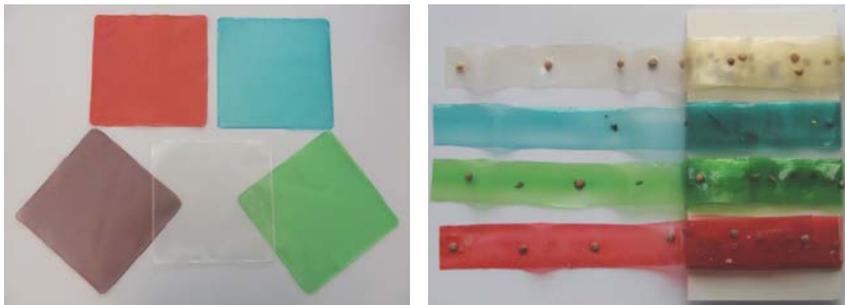


Fig. 1: Various protein films (left) and seed tapes

The implementation of the designed process was successfully realized by several small-scale production lines based on the extrusion technology and up-scaling efforts. The complete extrusion system is shown in [Fig. 2](#).



Fig. 2: Extrusion system ZE25A-UTXx48D-UG with power and control unit

Furthermore, an enzymatically catalyzed cross-linking reaction was directly integrated into the extrusion process for effective control of the functional properties of the final product. Subsequently, a functional process analysis was conducted with the aim of a quantitative description of the entire extrusion system for the processing of protein materials. This includes a detailed description of the main functions

feeding, conveying, plasticating, devolatilization, mixing and die forming as well as engineering approaches to the screw and die design following by troubleshooting aspects. The extrusion process was optimized for the reactive processing of proteins to films, sheets and tapes as the final product. In particular, the outstanding energy efficiency due to the processing at room temperature with no need of heating and cooling is unrivaled. Those preliminary implementation efforts led to the establishment of a manufacturing process for protein-based products with excellent optical, organoleptic and mechanical properties. Important process steps are illustrated in [Fig. 3](#).

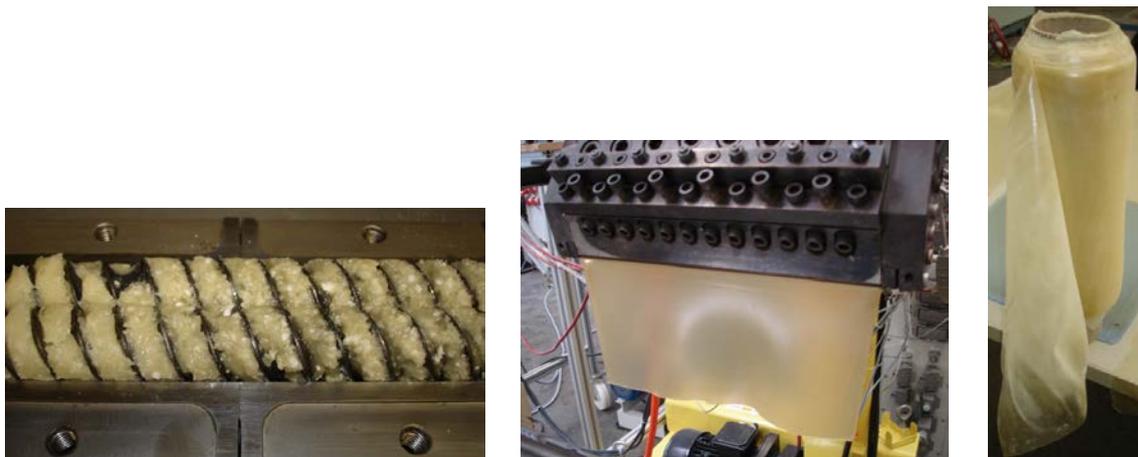


Fig. 3: Processing, forming and finishing of protein-based films

Thereafter, the functional properties of the products were determined according to standard procedures (European standards, EN). The results show clearly the potential of protein materials and of the manufacturing process to fabricate final products that meet manifold parameters essential for a commercialization. Compared to conventional discontinuous casting processes, the implemented process provides high throughput rates and a constant product quality which lead to remarkable functional properties.

The tensile strength of protein films was enhanced significantly up to 32 % by constant elongation values. With the introduction of the cross-linking reaction, the mechanical performance was further enhanced, numerical expressed by a considerable tensile strength increase of 73 % and elongation improvement of 72 %, respectively. Modification efforts by inner-film crystallization even showed elongations which could be increased by factor 3 up to elongations at break of 220 %. Morphological investigations showed homogenous smooth surfaces with no macroscopic visible defects. Furthermore, an appropriate control of the product properties was achieved due to selective adjustments of process parameters and material modifications leading to highly customizable products. All results were statistically ascertained and continuative optimized using the methods of DoE (Design of Experiments).

## 6. References

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