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Plastic chips as feedstock for 3D-printing-processes

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Zusammenfassung Das folgende Paper beschäftigt sich mit den Möglichkeiten Plastikspäne aus Zerspanungsprozessen von einem Abfallstoff mit hohem Mikroplastikanteil in einen wertvolles Ausgangsmaterial insbesondere für 3D-Druckprozesse umzuwandeln. Die Umwandlung von Abfall in eine Ressource erfordert die umfangreiche Untersuchung mehrerer Aspekte. Daher werden im Artikel sowohl Methoden zur Sortierung und Aufbereitung der Späne für eine effektive Handhabung beschrieben als auch die Effekte der Aufbereitung und ein Vergleich verschiedener Verwertungsketten. Eine ganzheitliche Sichtweise, welche die technischen, ökonomischen und ökologischen Aspekte der Materialien und Prozesse umfasst, ist der Schlüssel zur Umwandlung der Späne in ein wertvolles Ausgangsmaterial. Unter die technischen Aspekte fallen vor allem die Reinheit und Form der Späne sowie die Materialdegradation während des Aufbereitungsprozesses. Als ökonomische Faktoren wurden vor allem die verfügbare Menge der Späne und der Materialbedarf der untersuchten Prozesse betrachtet, während die Reduzierung von Abfallströmen und des Ressourcenverbrauchs durch die Herstellung von Neumaterial die ökologischen Aspekte darstellen.

Keywords Kunststoffe, 3D-Druck, Kreislaufwirtschaft, Späne, Fräsen

Abstract This paper is exploring the possibilities to turn plastic chips from milling processes from a waste containing a high share of micro-plastics into a valuable feedstock for plastic production processes especially 3D-Printing. Turning waste into a resource needs a complex investigation in several fields. Therefore, the paper describes the chips and methods of sorting, the treatment of the chips for an efficient handling, the effects of the treatment as well a comparison of different production chains, that use the chips. A holistic view including technical, economic and ecological aspects of the materials and processes is the key for a successful transformation of the chips. Technical aspects are the form and the pourability of the chips as well as the thermal degradation of the material while processing. Economic aspects are concerning the amount of chips that are available and can be processed by the tested methods and the effort that is necessary. The ecological aspects are the reduction of waste and the benefits of avoiding to produce new materials.

Keywords Plastics, 3D-Printing, circular economy, chips, milling

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Introduction and motivation

For single parts and small-scale-series milling is still a very common manufacturing process in addition to additive manufacturing processes. Because of high cutting-volume-ratio of up to 80% of the material is lost and huge amounts of waste are created. This is an ecological and economical problem, because expensive technical plastics like polycarbonate (PC), polyoxymethylene (POM), polyamide (PA) and ultra-high-molecular weight polyethylene (PE UHMW) and high-end plastics like polyether ether ketone (PEEK), that contain a lot of energy and resources from the production process, are turned into waste that can pollute the environment and it's disposal has to be paid. The high costs for native semi-finished PEEK-products, of actual about 200 €/kg [1] and for PEEK-3D-printing filaments starting actual about 320 €/kg [2], are the main economic reason for turning waste into feedstock. The high energy consumption for the synthesis [3] of polymers and avoiding the emission of microplastics are the most important ecological reasons for the research. The typical amount of plastic chips for a smaller company consists of 200 kg up to a ton of chips of three or four Materials per year. This is not enough as feedstock for conventional recycling processes to work economical. Therefore, the material is a good feedstock for processes with lower needs of material than extrusion or injection moulding, processes like 3D-printing and

micro-injection-moulding.

Their low weight-volume-ratio and the low pourability

are the biggest difficulties in processing the chips. Therefore, the aim of the treatment is it to improve these properties with as less effort and thermal damage as possible to gain high benefits for the environment and the companies.

Definitions

Chips occur in many different size and shapes, depending on the material, the milling process and the used tool. For the paper only the properties of PEEK-chips have been investigated, because of the high price of the material and the big sample, that could be supplied by the partners. Micro-injection-moulding of parts with a weight less than 1 g [3] with a DESMA formicaPlast 1K machine [4] and extrusion processes like

Fused-Deposition-Molding (FDM) or printing granulate with an extruder screw [5], [6], or a plunger printer [7], are used as reference processes.

Granulates for the investigations are produced by a twin-screw extruder (Noris Plastic ZSC 25/40D) and filaments by a single screw extruder and filament production line (Labtech LE30-30/C-HA, LFVW-100/L4, LCAT-25, & LTWU-25). The specimen for the tensile test is designed after the standard "Mikro Schulterstab" of Kunststoffzentrum in Leipzig (KUZ) [8].

Differential Scanning Calorimetry (DSC) with DSC 204 F1 Phoenix®, Netzsch and Thermogravimetric-Analysis (TGA) with TG 209 F1 Iris®, Netzsch are used for the identification of impurities and analysing the thermos-oxidative degradation of the material in the different recycling-processes. The main criteria are the time and the temperature, when the specimen starts to lose weight in the TGA because of the beginning of its oxidation with formation of volatile components. Therefore, the material to be tested is first heated under inert gas, kept at a constant temperature for a few minutes to achieve equilibrium and then exposed to an air atmosphere under isothermal conditions.

Literature review

The literature on which this paper is based is concerning the types of chips [10], [11], the handling and sorting of chips [12], [13], [14] and the processes of 3D-Printing [6], [7], [8] and micro-injection-moulding [4], [5], [15]. There is no literature available for using plastic chips as feedstock for direct recycling processes as they are used in standard mechanical, chemical or thermal recycling-processes [16]. The third topic of the research are the methods for comparing ecological and economical properties of the processes. A useful reference for this work is the life-cycle-assessment of 3D-printed-PEEK parts [17] Using 3D-printing as recycling process is for example described in [18], [19] and [20]

Properties of the Chips and mechanical Treatment

Material properties of PEEK

PEEK has outstanding mechanical, thermal and chemical properties (ref. Table 1). The high process temperatures of 350°C to 400°C [21] are a challenge for the most plastic processing machines. So, PEEK is a great reference material for this study, because materials like POM and PA can be processed easily with a technology capable of processing PEEK.

Table 1: Selected properties of PEEK-types for milling [22]

Tensile modulus	4000 MPa
Tensile strength	110 MPa
Elongation at break	15%
Melting Temperature	343°C

Shapes and properties of the chips

Chips as a waste material have a low quality concerning their purity, the regularity of their shapes and the resulting handling and processing properties. A sample of unprocessed chips is shown and classified in Figure 1. The sample was not randomized but pre-sorted to ensure, that all common types of chips and pollutions are included.

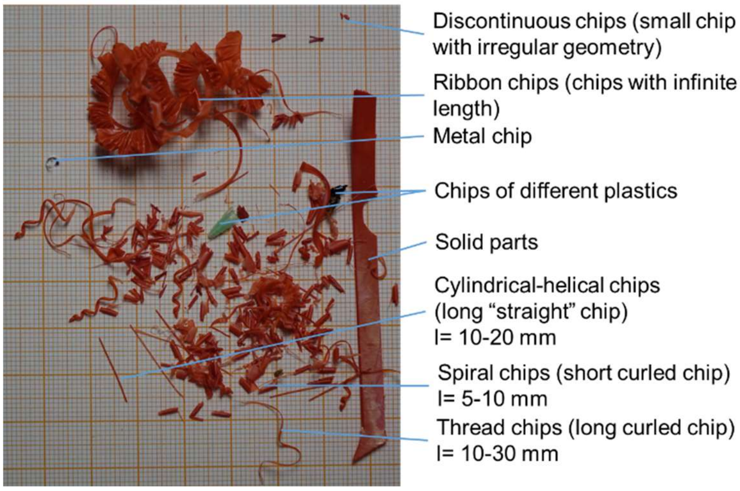

Figure 1: Pre-sorted sample of unprocessed chips

The handling of the plastic chips is one of the most crucial factors for stable processes. It guaranties a steady supply with feedstock material and thereby constant filling and pressures. Unfortunately, the chips are hard to handle their weight to volume ratio is about 0,17 kg/l and their repose angel of much more than 90° (ref. Figure 2) standard handling and dosing for example by vacuum conveyers or screws is not possible

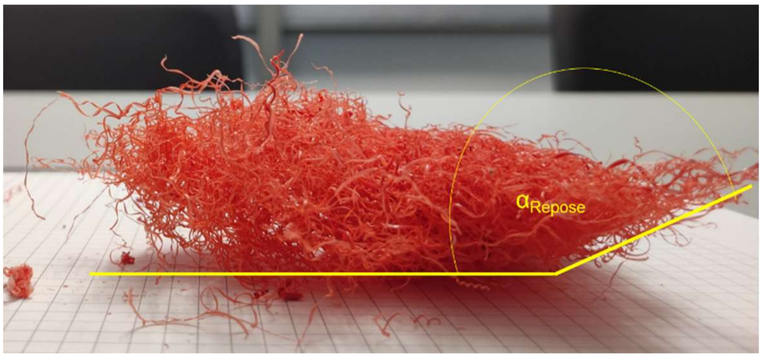

Figure 2: Repose angel of unprocessed PEEK-chips

The irregular curled and long shape of the chips is the reason for these properties. To increase the knowledge about the chip-geometry selected chips have been analysed by microscopy. The aim of the analysis is to measure the dimensions of the chips and to describe their surface and geometry in detail.

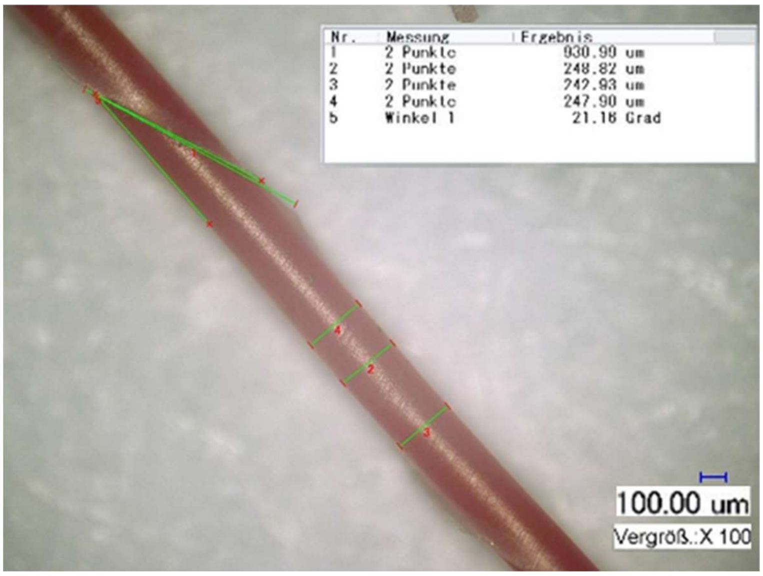

Figure 3: Microscopic picture of a cylindrical-helical chip

As shown in Figure 3 even the chips, that are straight on a macroscopic level are tightly curled and have a lined surface structure. The smallest particles of the sample were completely irregular fragments of chips with a size of less than a millimetre (ref. Figure 4)

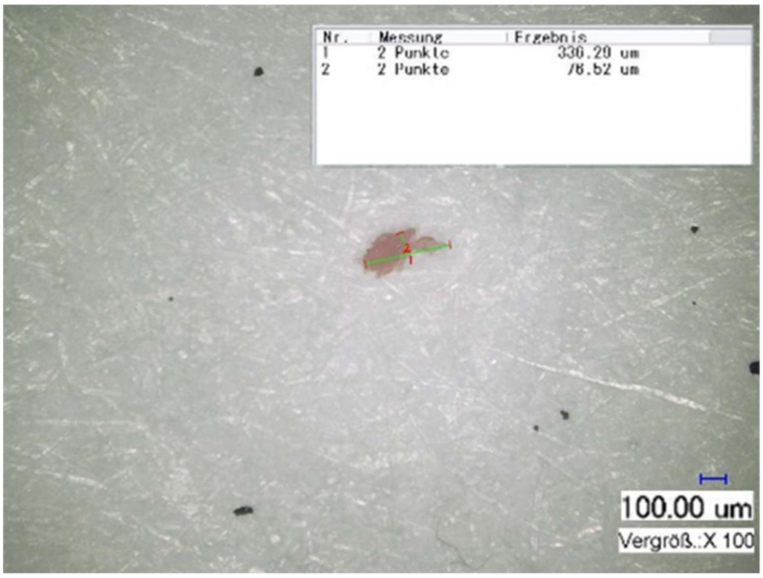

Figure 4: Microscopic picture of a discontinuous chip

So, the chips are a potential source of micro plastics [12]. A careful collection and recycling process is avoiding a release of microplastics in the environment and therefore a necessary part of a holistic recycling concept.

Because of those properties a sorting and mechanical treatment of the chips is necessary and special machinery as described in [23] should be considered.

Changing the shapes and properties of the chips by shredding

The reduction of the length is a promising way to improve the handling and processing properties of the chips. The ductile material behaviour (ref. Table 1) of the PEEK-chips and their low weight are complicating the treatment. The low weight leads to dominance of adhesive, aerodynamic and electrostatic forces over the gravity. The ductility of PEEK requires a cutting process instead of a simple breaking process to reduce the length of the chips. Anyways, a conventional shredding mill is already decreasing the length of the chips a lot and crushing bigger solid parts mixed in the chips. The handling of the chips was done manually for this study but can be supported strongly by an airflow. First tests prove this concept.

The sorting process is not part of this study but can be done by optical methods. Due to the handling properties of the chips and the shredding process it should be done in to steps. Before and after the shredding metal pieces should be separated by standard technologies to avoid damage at the machines. Different types of plastic should be detected by colour and be removed after the shredding process because of the easier handling. Optical detection methods and pneumatic removal methods, that are state of the art [13], are suitable for this task. Due to the low amount of plastic contamination in the samples no special methods are required to achieve a good quality.

The effect of the shredding is a significant length reduction of the chips, which can be seen on a macroscopic level and a microscopic level. On the microscopic level the edges of the chips are more rounded due to the cutting process. Therefore, their friction is probably reduced, what slightly increases the pourability.

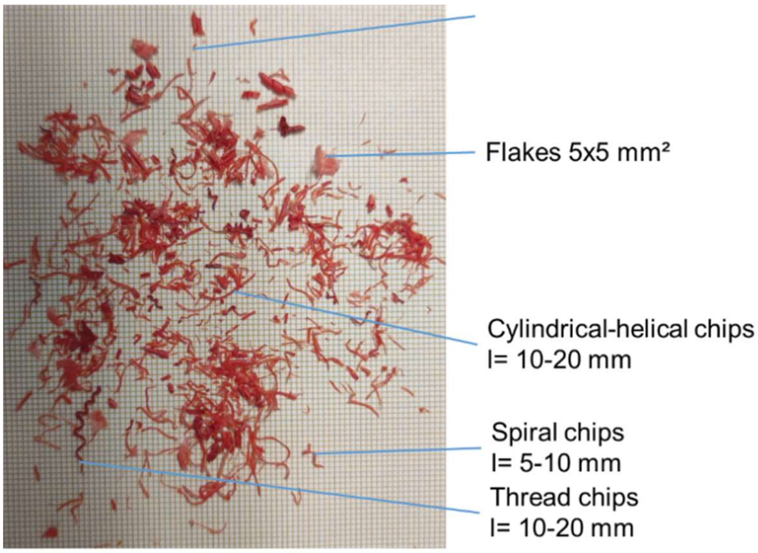

Figure 5: Randomized sample of shredded chips

The main effect is the length reduction. The reduction of the length reduces the possibility of the chips to interlock each other. The effect can be seen in Figure 6 and be measured by the weight-volume-ratio of 0,36 kg/l (+109%) and the repose angel of less than 90°.

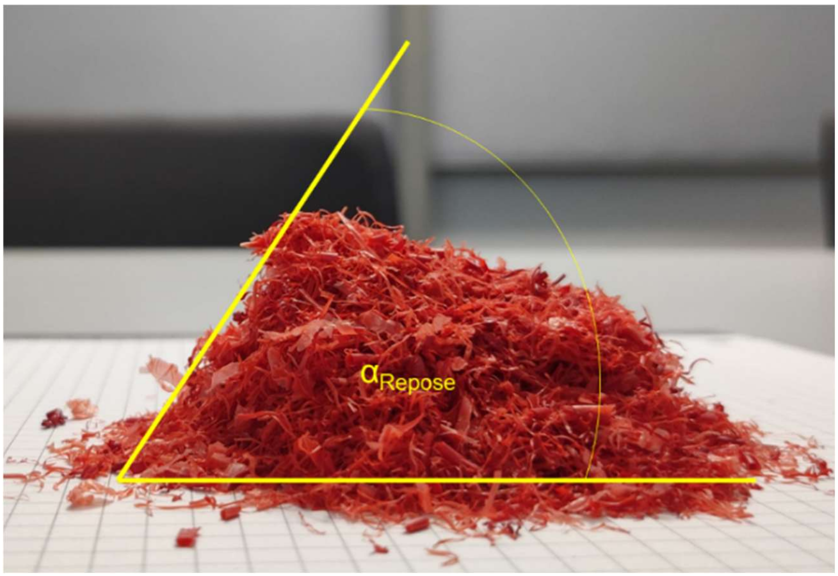

Figure 6: Repose angel of the shredded chips

The shredded chips can be processed in plastic-processes like extrusion, (micro)-injection-moulding and 3D-Printing with some extra effort. Compressing the chips to pellets is an option, that has to be explored. The expected challenges for pelletizing are the high tensile strength and the low friction of the chips. These properties complicate the forming of the pellets.

Possible Recycling process chains

Structure of the recycling processes

The aim of the recycling process is it to form the waste materials chips into new plastic parts. As seen in Figure 7.

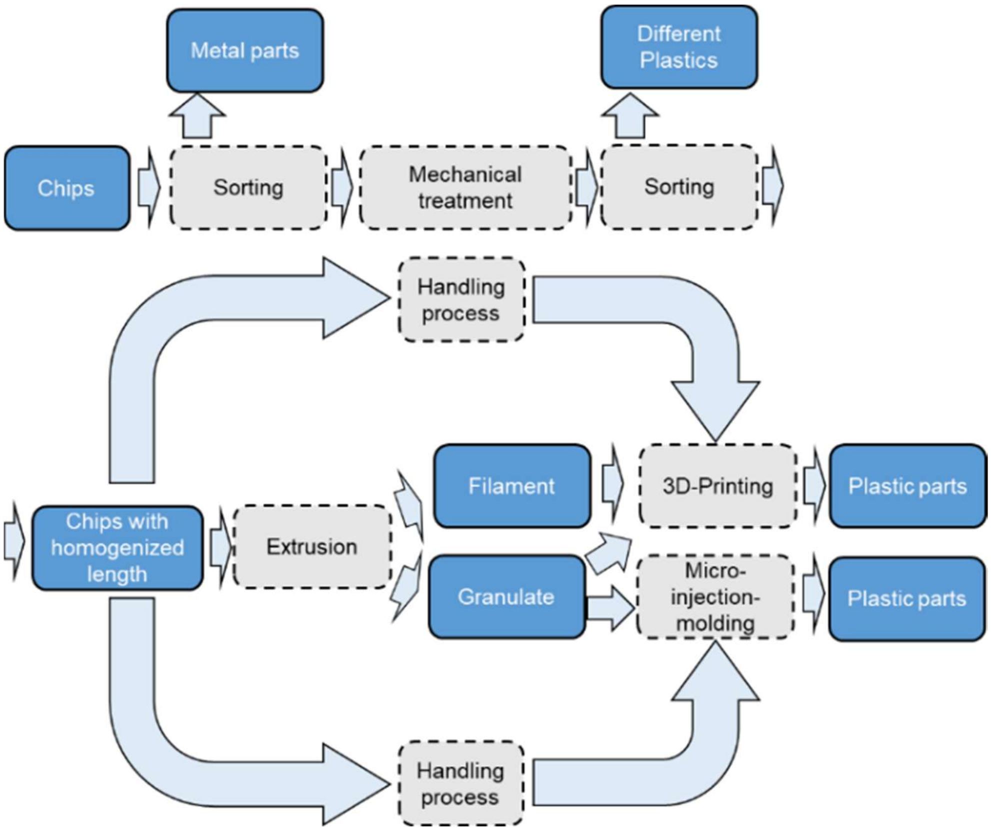

Figure 7: Process-scheme for the study

A direct recycling process from waste to part route is needed for a local recycling for economic and ecological reasons. It reduces the investment costs, the amount of energy that is needed for the recycling process and the loss of material during the process steps. Therefore, a characterization of each process steps is necessary. Companies, that produce parts by milling, are especially benefitting from a direct recycling by 3D-Printing. 3D-Printed parts are a nearly perfect addition to their current product portfolio.

Extrusion processes

The extrusion processes investigated in this study are not producing finished profiles but semi-finished products in form of granulates and filaments for the next steps in the recycling process.

The granulates were produced on a twin-screw-extruder, cooled down in a water bath and pelletized by cutting.

The result was a homogeneous and easy to handle granulate with a diameter of 2-3 mm and a length of 2-3°mm. As shown in Figure 8 the repose angle, as main criteria for the handling, is strongly reduced. The occurring fluctuations of the diameter are caused by the pulsation of the feedstock supply. This can also be seen at the fluctuation of the pressure between 35 bar and 45 bar.

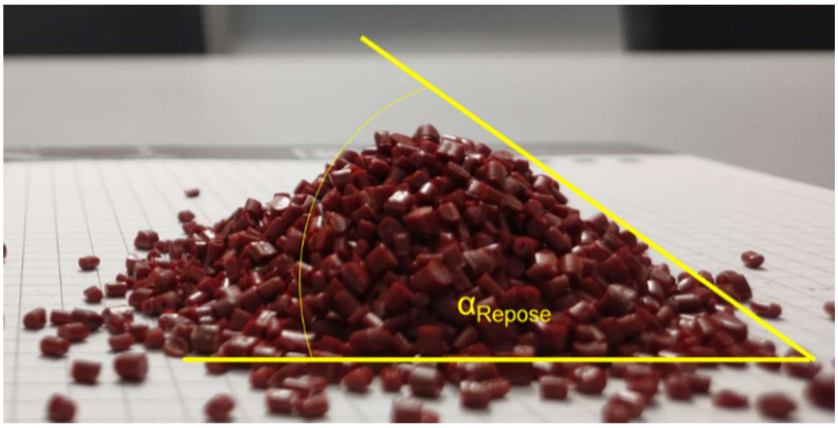

Figure 8: Repose angel of granulate

A constant supply of chips is crucial for the material quality. Therefore, the chips were distributed by weight with a dosing screw (Brabender DSR 67). A screw for fibres provided a higher and more continues feeding rate of up to 10 kg/h than a standard screw. The process temperatures were set between 300°C at the feeding zone up to 400°C at the nozzle.

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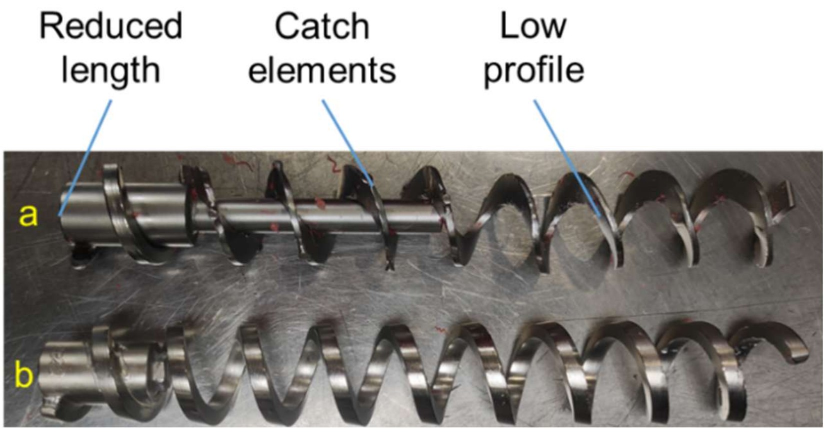

Figure 9: Comparison of a fibre feeding screw (a) with a standard feeding screw (b)

The single-screw-extruder, that is used to produce the filament for the FDM-printing, is not equipped with a feeding and dosing unit. So, the supply with the feedstock is done manually, which is resulting in fluctuating filament diameters, that can cause problems in the printing process.

The temperatures were set between 355°C and 385°C. Because up to 80% of the original material are turned into chips in the milling process, an extrusion process, that creates sheets or blocks for milling is not considered. The low ratio of fresh material and multiple recycling circles complicate the recycling process because of the expected thermal degradation. So, processes with a low amount of waste material are chosen as reference recycling processes.

3D-Printing setups

To evaluate the properties of the printing processes, especially the thermal-oxidative degradation two setups have been used. At first a conventional high FDM-printing-head attached to a self-designed portal-system [24] to print the filament. Because of the inconstant diameter of the filament only an extrusion process was tested with the filament printer, but not a complete printing process. The complete printing process and the investigation of the mechanical properties of the printed parts are the aim of further testing.

A prototype of a high temperature piston (ref. Figure 10) extruder for processing the granulate and the shredded chips directly. Chips and granulates could be extruded at 380°C with the used setup. The feedstock was supplied fully manually. The low weight-volume-ratio of the chips in combination with the simple setup has a big influence on the process. The printing head consists of heated and isolated plunger with up to three heating zones, that can be heated up to 440°Cand a piston driven by a spindle and a stepper motor. It applies pressures up to 10 bar. This pressure drives the melt out of the nozzle. The prototypic setup is actually not equipped with pressure sensor.

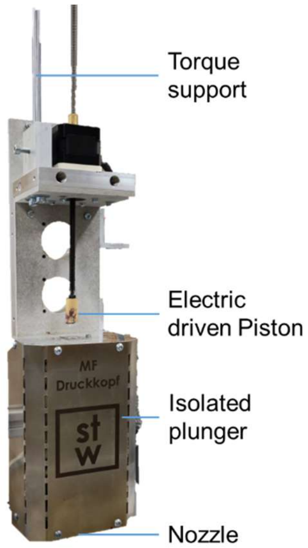

Figure 10: Piston-Printer-Prototype

The chips are compressed by the piston and the volume of the cylinder is strictly limited. Due to this, the amount of processable melt is only a few cubic centimeters. Therefore, a printing head based on screw, that ensures a constant material supply, is suggested for processing chips in 3D-Printing. But the direct processing is a prove of concept for a direct recycling process at the source of the chips.

For the current stage of the researches the focus was on the extrusion process and the behaviour and the properties of the melt. Other 3D-printing relevant processes like the adhesion to the building platform and former layers as well as the continuity and precision of the melt flow are part of further investigations.

Micro-Injection-Moulding-Process

Micro-injection-moulding has because of the low weight of the parts a much lower material need than conventional injection moulding, despite the extreme short cycle-times of 4 s [25] to 15 s .That is a good precondition for the use of the chips, which are only available in small amounts of a few hundred kg per year, as feedstock material.

The Desma formicaPlast 1K machine, that is used for this study has a two pistons system that separates the plasticisation - and the injection process [5]. The small amount of plastic melt in the machine reduces the thermal load of the plastic [15].The standard PEEK-granulate made by the twin-screw-extruder can easily be processed with standard settings. With processing temperatures of 390°C and mould-temperatures of 160°C and a max. pressure of about 1500 bar. The direct processing of the chips can be done with similar settings but requires a manual application of the feedstock in the first cylinder.

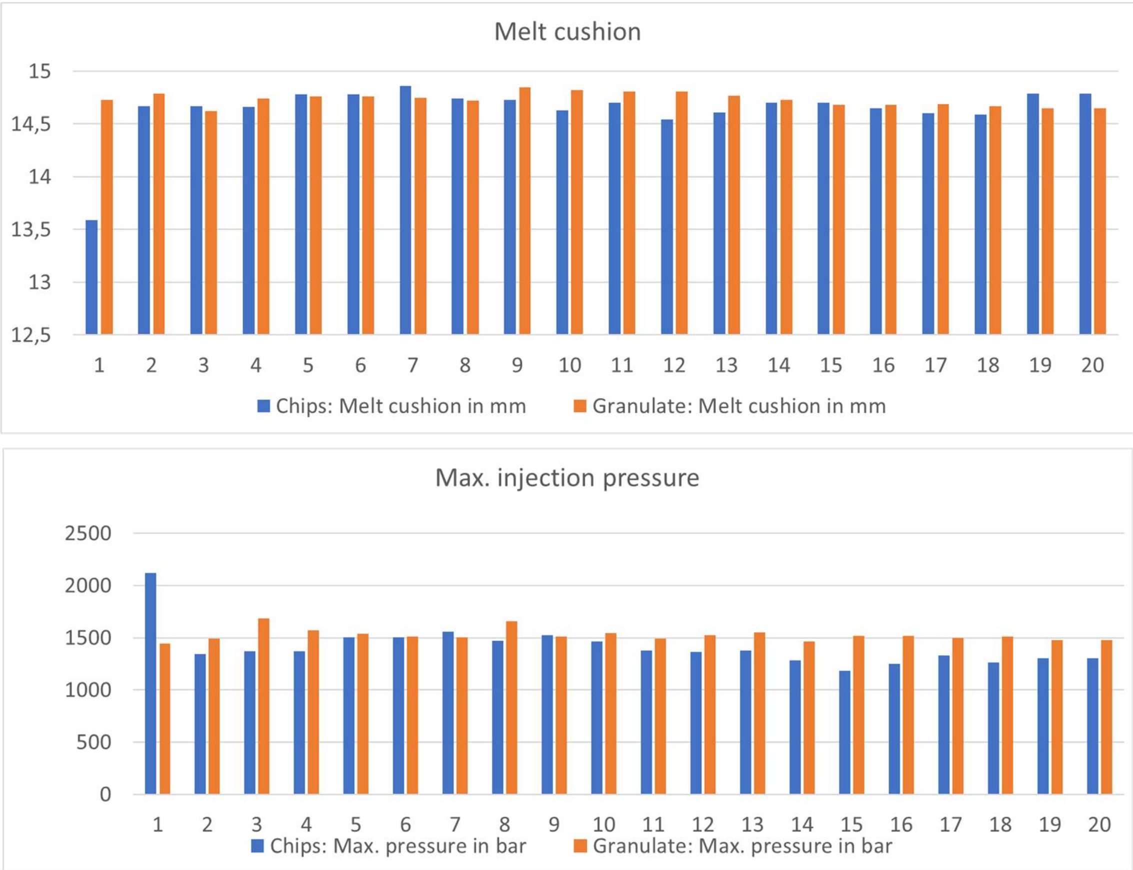

Figure 11: Melt cushion and max. injection of two micro-injection-moulding samples with chips and granulate

Figure 11 shows two main indicators, the melt cushion and maximum pressure, for a stable process on two randomized samples of 20 micro-injection-processes. Due to the more continues material supply the granulate process is a bit more constant, but both processes are in a stable state.

The main challenge of the direct recycling is to supply the chips to the plasticization unit. The standard vacuum transport to the supply box is working and the compression of the chips in the cylinder by the piston is also working, but because of the low weight at and low pourability of the chips they need a support for being put into the cylinder. For the study this was done manually but further developments can create a mechanical or pneumatic system for this task.

Thermal-oxidative degradation in the recycling processes

For detecting the thermal-oxidative degradation of the material, it was analysed by an isothermal TGA with synthetic air at 385°C. The loss of mass in the first 10 min is selected as criteria for the degradation. As higher the loss is, as more anti-oxygen-agent was consumed in the recycling process. Figure 12 shows the TGA-thermogram of the shredded unprocessed chips as example for the analysing process.

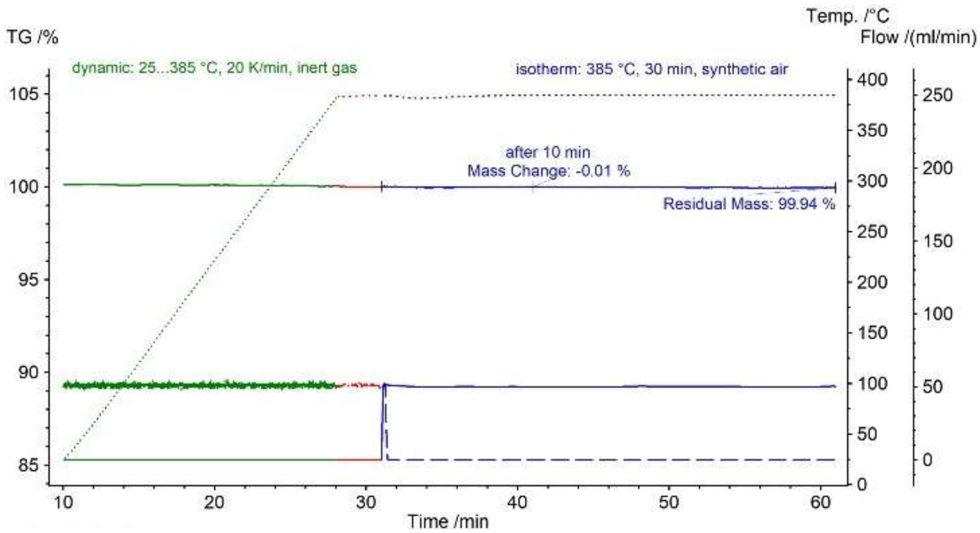

Figure 12: Isothermal TGA of the shredded PEEK-chips

The comparison between the weight losses, shown in Figure 13, demonstrate that there is a slight thermo-oxidative degradation during the recycling processes. This can be seen at the much lower weight loss of the unprocessed chips. The long heat up time of the 3D-Printing process with the prototypic printing head is visible at the increased weight loss.

The differences between the processes can be seen in Figure 13. The differences between the two micro-injection-moulding processes and between the two extrusion processes need further investigations. But due to the prototypic setup of the recycling processes the fluctuations in the thermal load and the resulting thermo-oxidative degradation are too big to rank the processes.

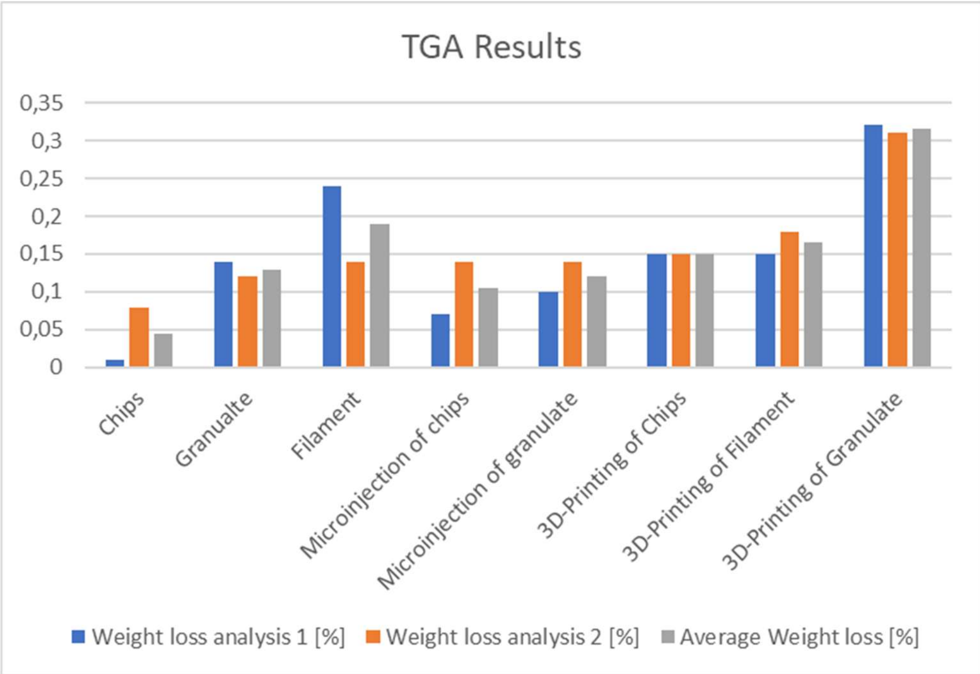

Figure 13: Results of the TGA

There is only a tendency, that micro-injection-moulding has only a small thermal load.

A side effect of the recycling process is the change in the crystallisation behaviour.

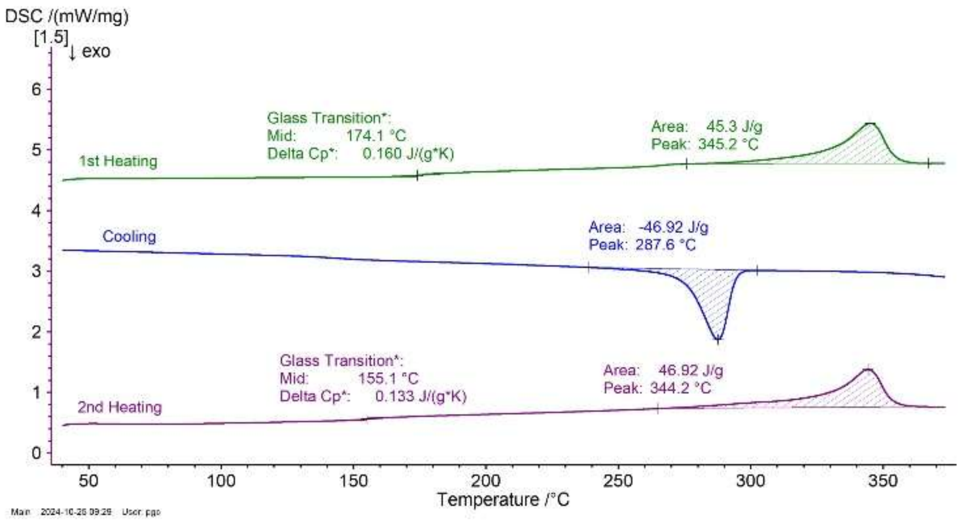

Figure 14: DSC analysis of the unprocessed chips

The unprocessed chips, as shown in Figure 14, show in the first and second heating of the DSC analysis only the typical glass transition temperature and melting peaks of PEEK. On cooling, the material crystallizes, as can be seen from the exothermic peak. Besides, impurities through the recycling process could not be detected. The second heating equals to the first one.

Figure 15 shows the different behaviour of the extruded PEEK in form of a filament. An exothermic effect between 170°C and 200°C with 178°C peak temperature is clearly visible at the first heat up. A so-called post-crystallization (cold crystallization) phase is setting heat free at this point. A regulated and slow cooling process of about 18 min (cooling rate 20 K/min) does not lead to a cold crystallization in the second heating again.

So, the first heating allows a statement to be made about the thermal history of the polymer. With the quick cooling of the extruded materials in a water bath the melt solidifies more in its amorphous form and freezes some of the amorph parts of the material. The polymer chains do not have enough time to organise themselves and form crystals. If heating is then applied above the glass transition, the chains present in the amorphous phase can move and rearrange themselves. This leads to cold crystallization. The enthalpy of this process is the higher the cooling rate during processing.

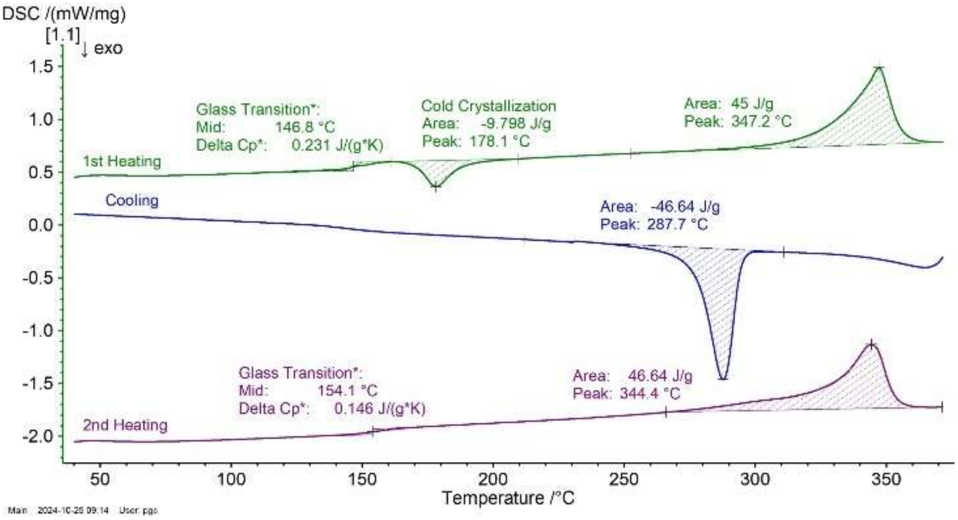

Figure 15: DSC analysis of the extruded filament

Therefore, the amount of enthalpy of cold crystallization, that is released, is a rough criterion to evaluate the cooling process. Furthermore, the finished components can be post crystallized through targeted heat treatment, thus optimizing the mechanical strength. Post shrinkage and distortion due to post crystallization are prevented.

Mechanical properties of the recycled materials

The specimens were tested in a tensile test setup on a Zwick/Roell Z010. For the micro test specimen special clamps with a geometric lock, are needed to avoid slipping. The test was done with a pre-load of 5 N and 1 mm/min testing speed for the young's module and 2 mm/min testing speed for the rest of the test. The 4 bar pressure of the clamps is not crucial because of the complete geometric lock of the specimens. Elongation is determined via crosshead travel.

The results show, that recycled PEEK chips still reach 81,6% of the strength and 1824% of the elongation of the original material. The material behaviour of the recycled PEEK, shown in Figure 16, is atypical for thermo-plastic-materials and it also differs from the standard behaviour of PEEK as shown in [26] and the native material also shown Figure 16. The clear peak between the elastic deformation and the plastic deformation is more common for metallic materials.

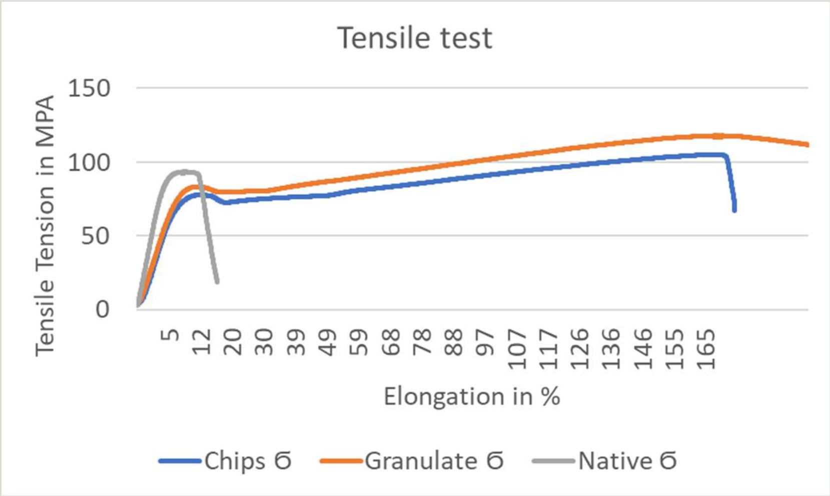

Figure 16: Tensile Test of a micro-injection-specimen of recycled chips, recycling granulates and a milled specimen of native material.

It is not clear if the difference between the native and the recycled material are related to the thermo-oxidative-degradation, the reduced crystallinity or specific effects of the micro-injection-moulding-process. A general comparison of the main mechanical properties of different recycling processes are shown in Figure 17.

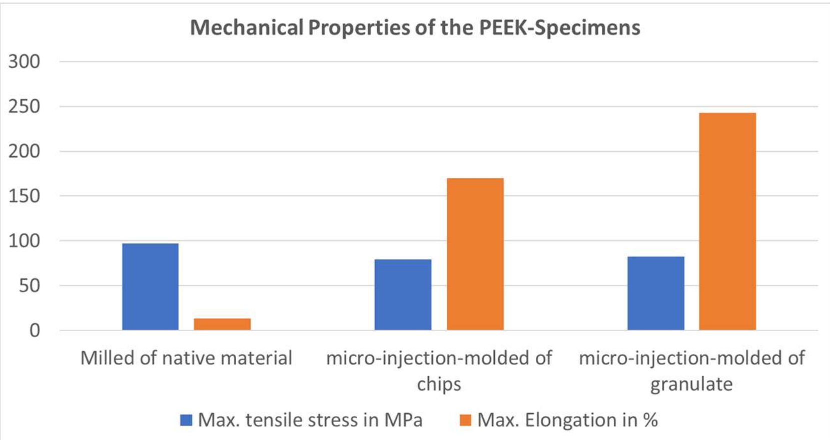

Figure 17: Tensile Test of a micro-injection-specimen of recycled chips, recycling granulates and a milled specimen of native material.

The cause of the complete different breaking mechanism especially the ductility must be investigated.

Economic and ecological evaluation of the printing process

The analysis of the recycling processes shows that the energy efficiency and cost-effectiveness of the processes depend largely on the number of process steps. There are clear differences in energy requirements, process costs and product usability between the production chains examined.

In terms of energy requirements, direct recycling using 3D printing, for example with a piston printer, is particularly resource efficient [27]. This process saves energy as preparatory steps such as granulation are eliminated. However, the advantage is partially offset by the low process stability and the manual and discontinuous processing of the chips, which leads to increased heat losses. In comparison, extrusion with pelletizing requires higher temperatures and a continuous supply of energy but delivers more consistent results. Micro-injection moulding proves to be the most energy-efficient process, as precise process parameters and short cycle times minimize the energy required per unit produced. Nevertheless, the challenge remains to further develop the technology of direct recycling through 3D printing in order to reduce material losses and inefficiencies.

The process costs also vary greatly. Piston 3D printers are associated with lower acquisition costs than extruders or micro injection moulding machines but are still in the prototype stage and are currently only of limited use. Micro injection moulding machines, such as the Desma formicaPlast 1K, offer proven reliability and reproducible quality, but are more cost intensive. Another criterion to evaluate the processes is the flexibility. Direct 3D-printing is the most flexible process, because of the low efforts for preparing the chips and the free forming of 3 dimensional shapes. Micro-injection-moulding has a massive, reduced flexibility, because of the needed mould and the strongly limited part weight.

Another advantage of direct recycling is the reduction in transportation distances, as the material can be processed on site. This reduces energy and costs compared to centralized processes with granulate. The direct processing of chips also requires fewer sorting and preparation steps. Although shredding significantly increases the bulk weight of the chips and thus improves their manageability, friction problems remain, which can make continuous feeding difficult.

The usability of recycled products depends crucially on their mechanical, thermal and aesthetic properties. Recycled PEEK achieves up to 81.6% of the original tensile strength and 1824% of the original ductility, which is sufficient for many technical applications. Micro-injection moulding provides the most consistent quality, while 3D printing processes show greater variation due to process variations. In terms of thermal properties, direct 3D printing leads to increased thermo-oxidative degradation due to long processing times, while micro-injection moulding and pellet extrusion minimize these degradations through more precise process control and shorter exposure times. Aesthetically and functionally, filaments and injection moulded parts offer higher surface quality and dimensional accuracy than 3D-printed chips but are suitable for applications where surface quality is less important.

The direct processing of milling chips into 3D printed parts on site opens up both economic and ecological advantages (ref. [18]). Up to 80% of the waste produced during milling can be recycled without any additional material input, which significantly reduces microplastic emissions. At the same time, energy-intensive transportation and intermediate processes are eliminated. Technical innovations, such as automated feed systems and screw-based extruders, could further increase energy efficiency in the future. For companies, this method offers cost benefits through material recycling, reduced waste disposal costs and the opportunity to include individualized 3D printed parts in their product range.

In the long term, the development of robust print heads and automated feeding systems will be crucial to improve process stability and manageability. A flexible combination of direct recycling and optional pelletizing could deliver the best results. On-site implementation promotes circular value creation, minimizes the ecological footprint and supports innovative manufacturing approaches, especially for small and medium-sized companies.

Conclusion and outlook

Chips from milling processes are suitable as feedstock material for plastic processes. For economic reasons a direct recycling close to the source is the best way and the local recycling avoids emissions by transport and material losses too. The most efficient way of recycling avoiding energy consumption and material losses is a direct recycling path from chips to 3D-Printing. But the handling of the chips is crucial. For constant printing processes the indirect path over granulates or filament to 3D-Printing is at the moment the best way but is too inefficient and the need of complex machinery like twin-screw-extruders is very high. Therefore, further developments are necessary. A major research field will be the opportunity to pelletizing mechanically.

The current recycling process is changing especially the mechanical properties a lot. The thermal behaviour seen via the DSC and TGA analyses, which shows only slight differences between native and recycled material. But in opposite to the slight decrease of the tensile strength strongly the increased ductility is huge improvement for the most applications.

Contributions

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