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Influence of Injection Moulding Process Parameters on High-Density Polyethylene Surface Hardness

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Abstract. Commodity polymers are a common part of everyday life. They consist mainly of polyolefins such as polyethylene, polypropylene. They are primarily used for ease of processing, cost and especially chemical resistance. The disadvantages of these polymers are low mechanical properties as well as temperature resistance. Any improvement in the mechanical properties can extend the application possibilities of the commodity polymers to the areas reserved for the construction polymers. This paper deals with changing two injection moulding process parameters - melt and mould temperature to high-density polyethylene (HDPE) surface hardness. HDPE hardness was measured using the method of Depth-Sensing Indentation (DSI) on three different instruments (ultranano-, nano- and micro-hardness tester). It has been found that as the melt and mould temperature increases, the hardness slightly increases.

Introduction

Polymer materials are currently the fastest-growing material group and represent the most important production and consumption sector by volume. Their rapid expansion and popularity in the manufacturing industry is based on the relative simplicity and energy-saving of processing compared to other types of materials, as well as the specific properties of polymers that can be further modified. This creates a wealth of materials, some of which are tailor-made for particular applications. In addition to the advantages of polymer materials such as low density, low thermal conductivity, electro insulating properties and the like, these materials also have disadvantages. These are for example strong dependence of mechanical properties on temperature, creep or limited resistance to wear, chemicals etc. The resulting properties of the polymer product are therefore largely dependent not only on the type of polymer itself but also on the substances added to the base material (additives, fillers, pigments) that improve its properties or reduce its cost. Another factor affecting the end product is the polymer processing method and process manufacturing conditions. These can, for example, cause a local overheating of the material if the process is set up improperly, thereby causing degradation of the polymer, thus deteriorating the resulting properties and durability of the final product [1].

In recent years, a great deal of work has been done to study the morphology of polymers depending on the change in injection moulding process parameters. Optimal process conditions ensure the quality of injection moulded parts, which is dependent on the polymer structure and its thermokinetics [2]. The morphology of the polymer affects the behaviour of the resulting part, in particular its mechanical properties, which vary depending on morphology [3-8], which implies that the mechanical properties of the injection moulded part is dependent on the type of polymer used, as well as on process parameters such as injection velocity, pressure, melt temperature, etc. If any of the processing parameters of polymers are altered, the mechanical properties of the resulting product may also change without the apparent quality of appearance [9].

The extent to which process parameters can affect the resulting mechanical properties is the subject of many investigations. Especially the effects of injection velocity, pressure and holding

pressure (basic parameters of injection phase of polymer injection technology) and mould temperature are considered on mechanical properties and internal structure of the final product. These properties are determined by classical methods and then compared [10-11]. Tensile and bending tests, hardness tests are considered conventional methods; as well as differential scanning calorimetry (DSC) and Vicat softening temperature (VST) [12-15].

This work deals with the influence of process parameters change (melt temperature and mould temperature) in injection technology on mechanical properties of the surface layer of HDPE product characterised by its hardness.

Experimental

Material and sample preparation. High-density polyethylene (HDPE) with trade name 25055E from producer DOW was used as the basic polymer material. An ARBURG Allrounder 470C GE injection moulding machine with screw diameter 40 mm was used for specimen preparation according to ISO 294-1 with dimensions 80×10×4, with the processing conditional, as can be seen in Table 1. In the preparation of all test specimens, constant conditions were maintained while the melt and mould temperature were varied.

Table 1. Moulding machine set parameters.

Material	Melt Temperature [°C]	Injection Velocity [mm/s]	Holding Pressure [MPa]	Mould Temperature [°C]		
				T_{f1}	T_{f2}	T_{f3}
DOW HDPE 25055E	200	60	66	40	50	60
	225	60	66	40	50	60
	250	60	66	40	50	60

Tests and evaluation. Standard hardness tests allow you to determine the hardness number by evaluating the depth or impression image. DSI instrumented indentation hardness test is a method in which an instantaneous variation of the indenter indentation depth in the material under investigation is detected and at the same time, the course of the load is sensed during the entire measuring cycle.

The course of DSI can be divided into two basic phases. During the first loading phase, an increasing force is defined on the indenter at a defined speed. The load reduction to zero is then performed in the second phase. Often, a delay with the maximum load applied is included between these stages. This period of time makes it possible to investigate the cold flow of material under load - the so-called creep. The indentance curve - i.e. the dependence of the indentation force on the indentation depth and the time chart of the indentation test - i.e. the dependence of the indentation force on time can be constructed from the data thus obtained, see Fig. 1.

The DSI method allows evaluating the hardness, elastic modulus, creep and indentation work. Since the indentation curve, respectively, its shape expresses the reaction of the tested material to the loading force; it is possible to read from it, in addition to the calculation of the hardness and the modulus of elasticity, other important information. E.g. phase transformation, cracks and layer delamination manifest themselves in a discontinuous course of the indentation curve.

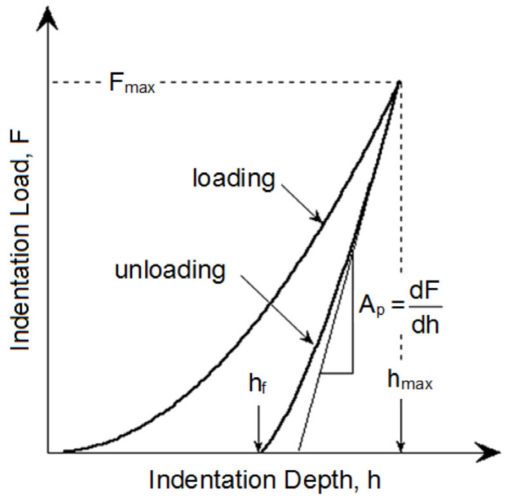

Fig. 1. Indentation curve.

Instrumentation hardness testing was performed according to ISO 14577 standard on three instruments:

1.) Micro hardness tester MICRO – COMBI TESTER (CSM Instruments) - The DSI hardness measurement was performed by a Vickers indenter (quadrilateral pyramid with a top angle of 136°). Mechanical properties were evaluated by the Oliver & Pharr method.

Table 2. Micro hardness test conditions.

	1. serie	2. serie	3. serie
Applied load [mN]	500	1 000	5 000
Delay on the max. load [s]	90	90	90
Loading/Unloading [mN/min]	1 000	2 000	10 000

2.) Nano hardness tester NHT2 (CSM Instruments) - The DSI hardness measurement was performed by a Berkovich indenter (triangular pyramid with a top angle of 142.3°). Mechanical properties were evaluated by the Oliver & Pharr method.

Table 3. Nano hardness test conditions.

	4. serie	5. serie	6. serie
Applied load [mN]	10	50	250
Delay on the max. load [s]	90	90	90
Loading/Unloading [mN/min]	20	100	500

3.) Ultranano hardness tester UNHT (Anton Paar) - The DSI hardness measurement was performed by a Berkovich indenter (triangular pyramid with a top angle of 142.3°). Mechanical properties were evaluated by the Oliver & Pharr method.

Table 4. Ultranano hardness test conditions.

	7. serie	8. serie	9. serie
Applied load [mN]	0,5	0,75	1
Delay on the max. load [s]	90	90	90
Loading/Unloading [mN/min]	1	1,5	2

Each series of measurements contained 10 measurements, which were performed on 10 different products under the same production conditions.

The structure of the fracture surfaces was examined using a JEOL 7500F scanning electron microscope. Fracture surfaces for SEM were prepared by breaking test specimens after cooling in liquid nitrogen. The specimens with fracture surfaces were glued to the targets by means of a dispersion adhesive and plated with gold in an argon atmosphere on a Balzars sputtering machine. SEM images were captured in * .bmp format by PC SEM. The accelerating voltage used was 15 kV and the working distance (WD) from 6 to 9 mm.

Differential scanning calorimetry was performed on a DSC 1 from METTLER TOLEDO. Test samples weighing 20 ± 1 mg were pressed into aluminium (Al) pans with lids. An empty Al pan was used as a reference. The measurements took place in the temperature range (80 - 160) ° C with heating and cooling rates of the first and second cycles of 10 ° C / min in an inert nitrogen (N_2) atmosphere with a medium flow rate of 20 ml/min. The results were evaluated using the STARe program, METTLER TOLEDO.

MS Excel 2016 was used for statistical evaluation. The evaluation was performed according to ISO 2602, which determines the statistical interpretation of measurement results. The measurement results are described by arithmetic mean and standard deviation.

Results and Discussion

HDPE is a semi-crystalline polymer that is commonly encountered in households during normal activities. E.g. this material is used to produce packaging and household items. An important prerequisite is the long durability of the surface against scratches and other adverse effects. The change in surface layer properties is dependent on the arrangement and amount of the amorphous portion.

Throughout the work, Vickers hardness is evaluated, which is not dependent on the applied indenter load, which allowed the measurement of very thin surface layers on several instrumented measuring instruments (ultranano, nano and micro hardness tester). The “Depth Sensing Indentation” (DSI) method has been developed for the instrumented measurement of surface hardness. While Oliver and Pharr have improved this method (especially the nature of the measured data evaluation), it is currently considered to be one of the most accurate for evaluating very thin surface layers.

Surface hardness. At the lowest applied load (0.5 - 10 mN) there was a very slight increase in hardness (approx. 3-5 %), see Fig. 2, 3 and 4, at increasing melt temperature and increasing mould temperature. A significant change in hardness (approx. 20 %) occurred at a load of 50 mN, where the indentation depth was up to 10 μm. A further increase in the indentation force brought an increase in the indentation depth, but the hardness varied only slightly with the indentation depth. A similar course of hardness was observed at all melt and mould temperatures.

Scanning electron microscopy. Two layers can be identified from microscopic images. Layer I is approximately 1 μm thick (Fig. 5 and 6). In terms of hardness measurement, it corresponds to indenter load in the range of 0.5 - 1 mN. The thickness of the said layer is within the range of measurement possibilities available with hardness tester, in particular case ultranano hardness tester. The hardness of the HV layer I is in the range of 2.7 - 3.0. The hardness of the layer II is in the range of 3.5 - 3.8 and the hardness of the "substrate" remains substantially the same at each selected indenter load, which is in accordance with the measured indentation depth.

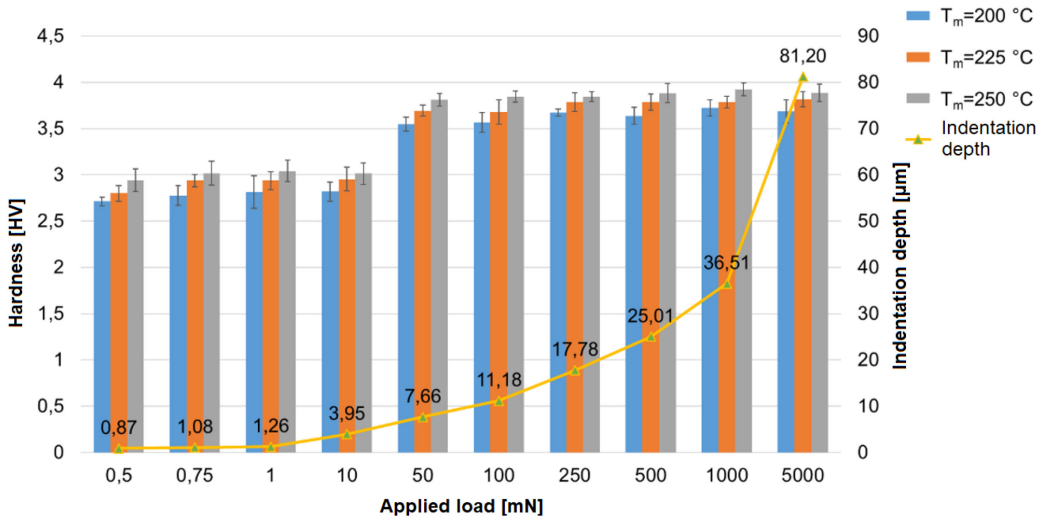

Fig. 2. Dependence of hardness and penetration depth on the load at three melt temperatures (T_m) and mould temperature T_f = 40 ° C.

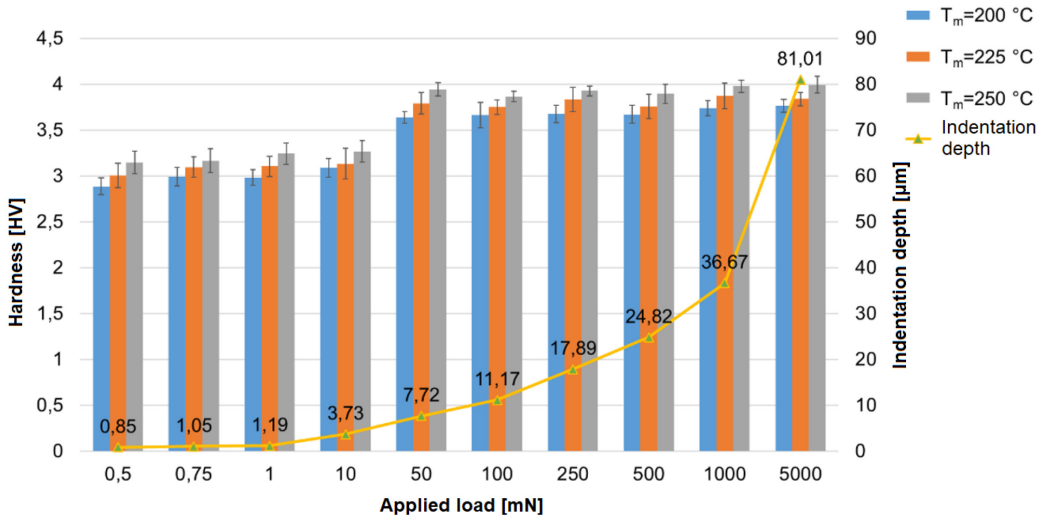

Fig. 3. Dependence of hardness and penetration depth on the load at three melt temperatures (T_m) and mould temperature T_f = 50 ° C.

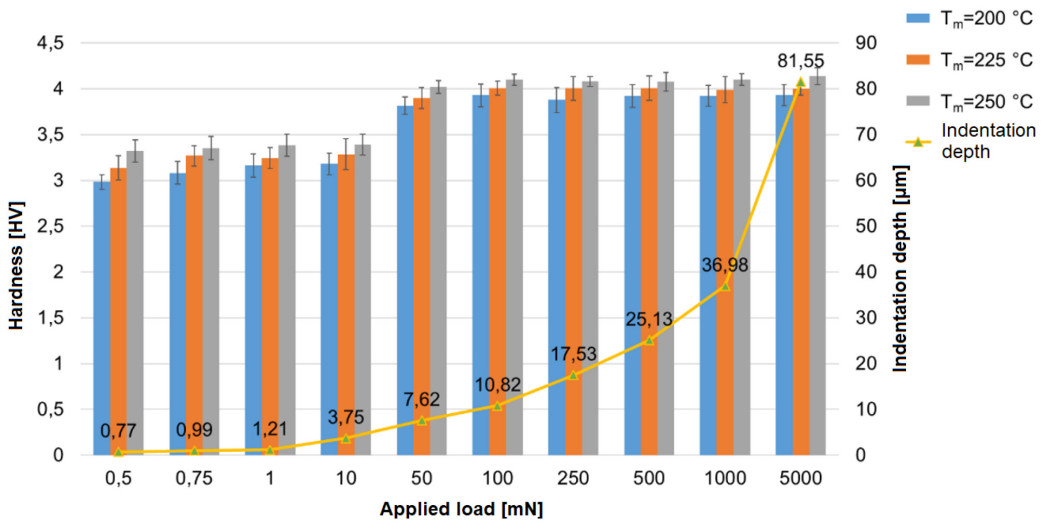

Fig. 4. Dependence of hardness and penetration depth on the load at three melt temperatures (T_m) and mould temperature T_f = 60 ° C.

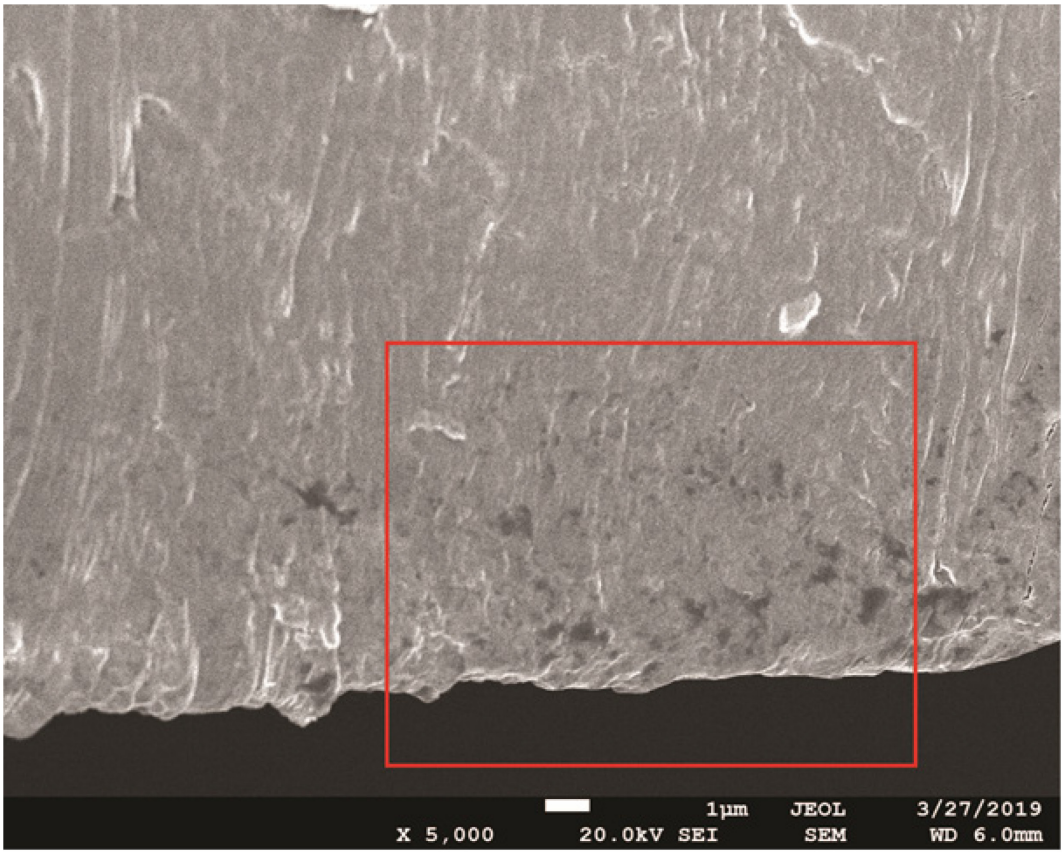

Fig. 5. Edge of fracture surface (magnified 5000x).

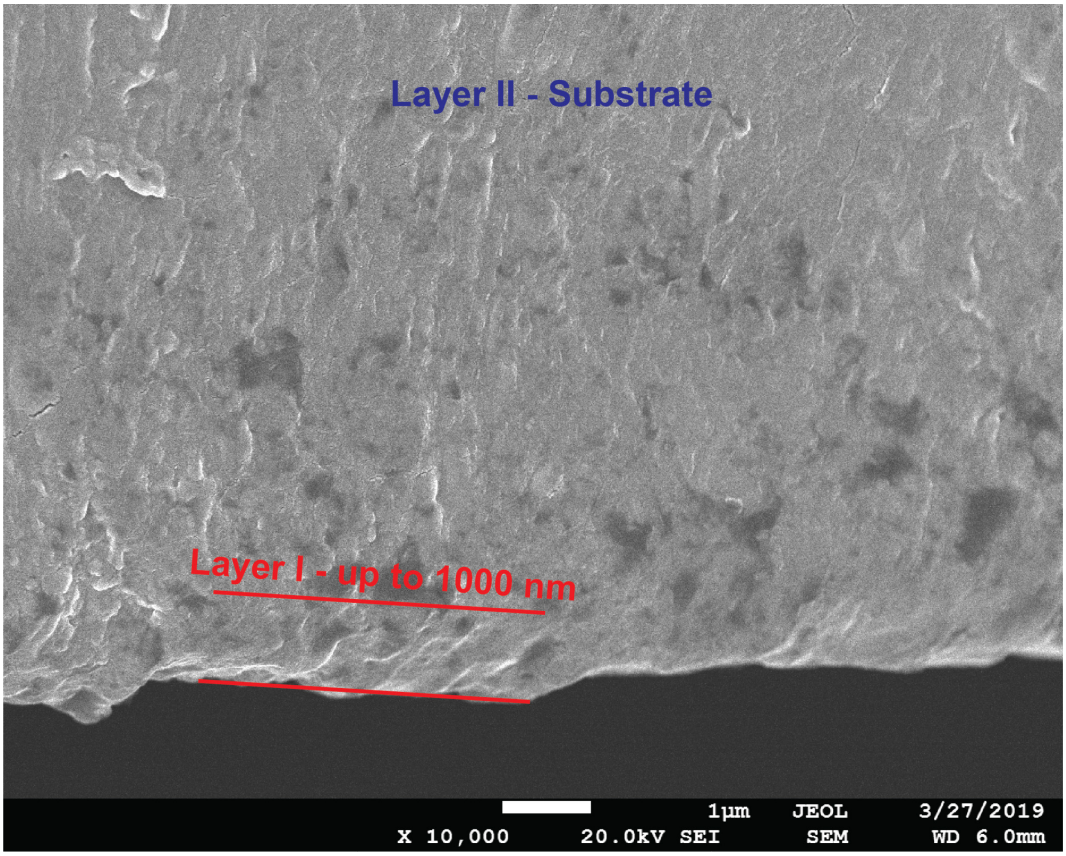

Fig. 6. Edge of fracture surface (magnified 10 000x).

Differential scanning calorimetry was used to evaluate the surface and inner layers of HDPE test specimens. Results from DSC measurements of the surface layer (microtome sections) and inner layers of test specimens are shown in Tab. 5. DSC measurements of HDPE test specimens with different melt temperatures show that the surface layer exhibits a lower crystallite melting point and a lower normalized crystallite melting heat than the subsurface layers, and an increase in the melting temperature and normalized crystalline melting heat in the surface layer (microtome sections from the test specimen surface). This means that the crystallization process is different at different melt temperatures and the nature of the crystallites formed is apparently different, which is reflected in the change in hardness of the surface layer.

Table 5. DSC measurement results

Specimen with melt temperature [°C]	Maximum melting point [° C]		Normalized heat of melting of crystallites [J/g]	
	Body surface layer	Body inner layer	Body surface layer	Body inner layer
200	131,69	138,31	99,32	137,15
250	133,12	137,21	134,64	141,37

Summary

When examining HDPE injection products, two layers can be identified from microscopic images. Layer I has a thickness of about 1 μm. In terms of hardness measurement, it corresponds to indenter load in the range of 0.5 - 1 mN. The thickness of the said layer is within the range of measurement possibilities of the available hardness testers, in particular, an ultranano hardness tester. The hardness of the HV layer I is in the range of 2.7 - 3.0. There is a relatively small change in hardness with a change in indentation depth. A significant change in hardness (approx. 20%) occurred at a load of 50 mN when the depth of indentation was up to 10 μm. The hardness of the layer II (substrate) ranges from 3.5 to 3.8 HV. After this step-change in hardness attributed to the second identified layer, the hardness does not change. The hardness of the "substrate" remains essentially the same at all indenter loads used, regardless of the depth of indentation. A similar course of hardness was observed at all melt and mould temperatures.

DSC calorimetry results from HDPE bodies with different temperatures indicate that the surface layer exhibits a lower crystallite melting point and at the same time a lower normalized crystallite melting heat than the subsurface layers. At the same time, as the temperature rises, the melting point and the normalized melting heat of the crystallites in the surface layer increase. This implies that the crystallization process is different at different temperatures and apparently there is also a different character of the crystallites formed, which correlates very well with the changes in nanohardness.

The results of the realized study brought new knowledge about the behaviour of polymers in the processing of injection technologies. At the same time, they have shown that the extent of these changes can be reliably identified by available methods, including the Instrumented Hardness Test. It is a highly accurate method of measuring hardness allowing accurate detection of indentation depth. An important contribution to further research in this area is also the verification of the importance of careful preparation of surfaces of examined materials.

Acknowledgement

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