Polymeric Materials Recycling / Processing Optimization

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The paper presents a brief overview of the polymeric materials recycling problems and especially those connected with polypropylene reprocessing. It is pointed out the manner in which reprocessing of polymeric materials determines the change of the polymeric melts viscous-elastic properties and, as a consequence, the change of rheological behaviour during the recycling process. It was chosen to do the study of the extrusion reprocessing, so, it was determined, after a significantly number of recycling, the modifying of the process parameters needed in order to obtain the best performances and the highest quality of the product. There were compared the screw extruder performances in the case of the first processing and after ten times recycling of the PP.

Keywords: recycling, rheological behavior, polymeric materials, polypropylene, screw extruder

The materials recycling, especially polymeric ones is one of the most challenging environmental protection problems in the present. This comprises the aspects of the collecting, transporting, sorting, washing and reprocessing in the optimum conditions to obtain the best quality products, as they can be obtained at the first processing. It is no question to lower the quality standards in reprocessing, because this will lead to the increasing of the wastes quantities.

Many papers deal separately with each of these problems (recycling methods and technologies, material behaviour changing caused by many times recycling, the influence on the quality of the products obtained from recycled materials, optimum reprocessing parameters/ high performances etc.) [1-13]. Function of the technology used and of the product processed, the quantity of the recycled polymer mixed with the new material in reprocessing varies, usually, between 3 and 60%.

The most spread type of recycling is that of the scraps produced in the factory and recycled immediately in the fabrication process (injection, extruding, blown film molding etc.). Using improved technologies and a better automation of the processes, the quantity of scraps is expected to decrease. These will be possible based, also, on the better knowledge of the polymeric materials rheological behaviour.

From the environment protection point of view the most important/critical recycling is after use and it is a rather complex problem, its correct solution needs an interdisciplinary effort – of organizing, researching, the best methods of recovery/treatment of wastes, up to applying in practice of the most recent recycling technical procedures/equipment.

The authors preoccupation for polymer recycling originates years ago, with the study of polymer recycling methods and modification of the rheological behavior due to many times recycling of the most commune used polymers LDPE, PP, biopolymers based on LDPE etc.[1, 2, 8 - 14]. Here the authors are trying to integrate the polymer recycling and reprocessing into new products in one research, having in this way an appropriate view of the whole. So, here there are presented, briefly, some of the experimental results for the polypropylene, virgin and many times recycled, and also their influence on its reprocessing by extrusion. Finally there are emphasized the main issues.

In the literature there are also presented some experimental results for recycled PP for example [15-18], that are to be used here to compare the authors results.

Simulations of the screw extruder performances

The sort of polypropylene used in experiments was MIDILENA (produced in Romania) and the experimental method applied and the results are detailed in [11, 12, 14].

To simulate the screw extruder performances one can use different models: deduced from the metering zone conditions with and without leaks [19] or from the feeding zone conditions [20-24]. Here it was chosen the first model. There were done simulations for an extruding machine with one screw, in the case of pressure generation $\left(\frac{\partial p}{\partial z} > 0\right)$ or pressure consumption $\left(\frac{\partial p}{\partial z} < 0\right)$ in the metering zone, for virgin polymeric materials and 10 times recycled.

In the figure 1 it is presented the functional feature for the screw, $Q(\Delta p)$, for PP at $200°$ C, function of the screw rotation speed and as parameter the number of the recyclings compared with the virgin material, in the case of the flow model with leaks between the screw flight tip and the cylinder and the pressure variation along the channel is $\partial p / \partial z > 0$ (pressure generation – the worst situation from flow rate point of view, when pressure flow rate opposes drag flow). The mathematical model used for the flow rate is very simple, for Newtonian fluids rheological behaviour, but corrected with the actual value of the shear viscosity, determined based on the nonNewtonian rheological behavior of the polymeric melt. The flow rate is obtained as a sum between drag flow (it was used the hypothesis of the flow between parallel palates – the upper plate – the cylinder, the lower plate- the screw; it was used the lubrication approximation, so the upper plate – the cylinder rotates and the screw is motionless) and the pressure flow; the reduction influence of the flights is introduced by the coefficients F_a and F_p [19],

$$Q = i\frac{v_{c,z} \cdot w \cdot h_{p,3}}{2} \cdot F_a + i\frac{w \cdot h_{p,3}^3}{12\eta_{ef}}\left(-\frac{\partial p}{\partial z}\right) \cdot F_p - E_s\frac{\pi \dot{D}_c \cdot \delta_s^3 \cdot \Delta P_s}{12\eta_{ef,g} \cdot e}, \qquad (1)$$

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where η_{ef} is the effective viscosity in the screw channel given by the following equation:

$$\eta_{ef} = m \cdot \nu \left(\frac{v_{c,z}}{h_{p,3}} \right)^{\nu-1} \qquad (2)$$

and $\eta_{ef,g}$ is the effective viscosity in the gap between screw tip flight and the barrel given by the following equation:

$$\eta_{ef,g} = m \cdot \nu \cdot \left(\frac{v_{c,x}}{\delta_s} \right)^{\nu-1} \qquad (3)$$

In the relations 1, 2 and 3 the symbols used are: m, ν - rheological constants; $\nu_{c,z}$ - the cylinder velocity in z direction (towards the discharge of the melt into the extrusion head), $\nu_{c,z} = \pi \cdot n \cdot D \cdot \cos\varphi_{ec}$ where n is the screw speed, $\nu_{c,x}$ is the velocity of the cylinder in x direction (perpendicular on z, backwards over the helical screw flight), $\nu_{cx} = \pi \cdot n \cdot D \cdot \sin\varphi_c$, φ_c - screw flight helix angle measured at the cylinder surface, i – number of flights in parallel; h_{p3} - the metering zone depth; e - width of the screw flight, δ_s - the gap between screw flight tip and the barrel; ΔP_s – the pressure drop along one pitch of the screw, $\Delta P_s = \frac{\Delta P}{n_p}$, ΔP - total pressure drop, n_p - number of turns, E_s – coefficient taking into account the screw eccentricity.

The simulations were done for a screw having the geometry: the diameter D = 110 mm, channel width W = 99mm (square pitch flight), $h_{p,3}$ = 3 mm, e = 11mm, δ_s varies with the screw wear (between 0.2 - 1.6 mm), ΔP_s = 2 MPa, E_s = 1.2; n = 30 – 150 rot/min, $\partial p / \partial z$ = 10 – 30 MPa.

The rheological constants of the PP, at 200°C, virgin and 10 times recycled are presented in table 1 [12].

Table 1
RHEOLOGICAL CONSTANTS (EXPERIMENTAL RESULTS)

Material	PP	
Consts. / Pass	m [MPa·s$^\nu$]	ν
The first	0.0011	0.8628
The 10 th	0.0009	0.6404

The results presented in table 1 are in good concordance with the other scientific researches, for example given in papers [15-18] - it was verified that high temperatures and multiple processing cycles conditions change rheological properties dramatically - a lowering of the melt viscosity (here a steep decrease of the rheological constant m – which reflects the polymer melt consistency, which is attributed to molecular weight decrease [16]. Other scientists observed, also, a great loss in mechanical properties caused by degradation during multiple extrusion processes by a combination of thermal, mechanical and chemical degradation [15-18].

The influence of the leaks is important as the screw wears [19] so this case is taken into account in simulations.

Here are presented only some of the simulation results. In figure 1 it can be seen the results of the simulations: influence of the leak over the flight on the flow rate-pressure variation correlation of a single screw extruder, for PP virgin and 10 times recycled, at 200°C, in the case of pressure generation. For the screw speed were considered, in both cases, 5 values: 30, 60, 90, 120 and 150 rpm.

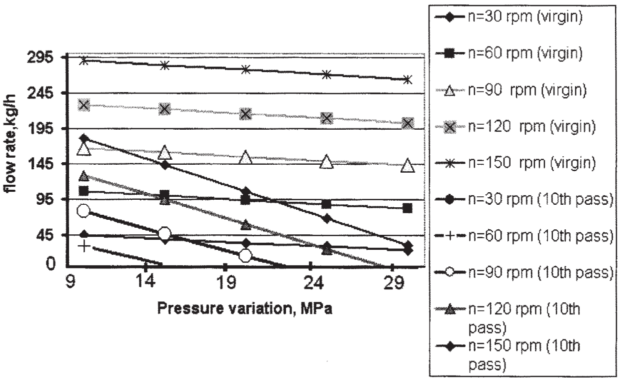

Fig. 1. Flow rate versus pressure variation for PP at 200°C (virgin,10th pass), model with leaks over the flight tip (δ_S = 0.6mm, in the case $\frac{\partial p}{\partial z} > 0$)

Due to recycling the inner polymeric structure is destroyed and as a consequence, the effective viscosity decreases so the amount of the polymer melt which is flowing backwards over the screw flight tip is growing, becoming greater as the pressure drop value increases.

From the simulations performed resulted that in the case of virgin PP the flow rate decreases slightly with the pressure drop. In the case of 10 times recycling PP, resulted a steep decrease of the flow rate with the pressure drop. From the limitation put for the lowest allowable output it can be seen that the screw speed, at high pressure drop values (~30MPa), must be high (over 150 rpm), otherwise the leak backwards/pressure effect could stop the polymer to flow towards the extruder head (the flow rate approach zero value). The differences between the flow rate in the case of virgin and 10 times recycled is about 38% at the lowest pressure drop value and reaches 85% at the highest pressure drop values. So the influence of the recycling on the screw characteristic is quite important, no matter the pressure profile is developed during the extruding process.

In the case of $\partial p / \partial z < 0$, the maximum pressure point has a metering effect, increasing the melt velocity in the screw channel and, as a consequence, the flow rate increases, as it can be seen in figure 2. In the case of 10 times recycled material (when the viscosity decreases) the flow rate is higher than in the case of virgin PP extruding – being the favorable situation.

In both cases presented in figure 1 and 2 the flow rate variation approaches a linear model. The worst situation, for the extruder performances point of view, is in the case of pressure generation in the metering zone $\left(\frac{\partial p}{\partial z} > 0 \right)$.

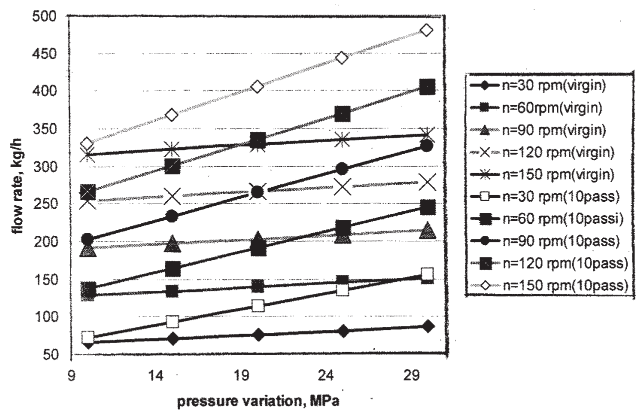

Fig. 2. Flow rate versus pressure variation for PP at 200°C (virgin, 10th pass), model with leaks over the flight tip (δ_S = 0.6mm), in the case $\frac{\partial p}{\partial z} < 0$

Table 2
INFLUENCE OF THE WEAR, REPRESENTED BY δ_s INCREASE, RESULTED FROM SIMULATIONS

Screw speed, n - rpm	Gravimetric leak flow rate, Q - kg/h Standard deviation	Number of recycling
150	$94{,}317\cdot\delta_s^2-66{,}42\cdot\delta_s+12{,}449$ $R^2=0{,}9984$	virgin
	$447{,}78\cdot\delta_s^2-267{,}75\cdot\delta_s+48{,}322$ $R^2=0{,}9991$	10 recycle
120	$91{,}937\cdot\delta_s^2-65{,}068\cdot\delta_s+12{,}241$ $R^2=0{,}9982$	virgin
	$413{,}26\cdot\delta_s^2-247{,}11\cdot\delta_s+44{,}597$ $R^2=0{,}9991$	10 recycle
90	$88{,}379\cdot\delta_s^2-62{,}55\cdot\delta_s+11{,}768$ $R^2=0{,}9982$	virgin
	$372{,}64\cdot\delta_s^2-222{,}82\cdot\delta_s+40{,}214$ $R^2=0{,}9991$	10 recycle
60	$83{,}597\cdot\delta_s^2-59{,}165\cdot\delta_s+11{,}131$ $R^2=0{,}9982$	virgin
	$322{,}08\cdot\delta_s^2-192{,}59\cdot\delta_s+34{,}758$ $R^2=0{,}9991$	10 recycle
30	$76{,}013\cdot\delta_s^2-53{,}798\cdot\delta_s+10{,}121$ $R^2=0{,}9982$	virgin
	$251{,}03\cdot\delta_s^2-150{,}1\cdot\delta_s+27{,}089$ $R^2=0{,}9991$	10 recycle

The extruder screw wears as the time went on. The influence of the screw wear, expressed here by the increase of the gap between the screw flight tip and the cylinder, respectively of the δ_S value, on the leak flow rate over the screw flight tip, is presented comparatively, for virgin and 10 times recycled PP at 200°C, at different screw speed values, in table 2.

The simulations showed that the total flow rate decreases with the increase of δ_S, a lot more pronounced with the increasing of the recycling number than in the case of virgin PP. A major decrease of the flow rate cannot be tolerated and on this basis it can be fixed its lowest limit. To compensate the decreasing effect, in the early stages of the screw wear the increasing of the screw speed could be an option, but this will lead to higher energy consumptions for the recycled PP extrusion, so the increasing of the products cost. The decrease of the flow rate must be limited at values that do not affect the productivity (only some percents). Previously the indication to change or to repair the screw was when the gap reaches 15% of the screw channel depth. In the case of the recycled material this could be unsatisfactory.

The leak flow rate, at the same value of δ_S, showed a relatively slightly increase with the screw speed and a very much increase with the number of recycling (for example the leak flow rate for δ_S=0.6 mm was 5 times bigger in the case of 10th pass than for the virgin material, for n=150 rot/min), leading to a dramatically decrease of the extruder performances.

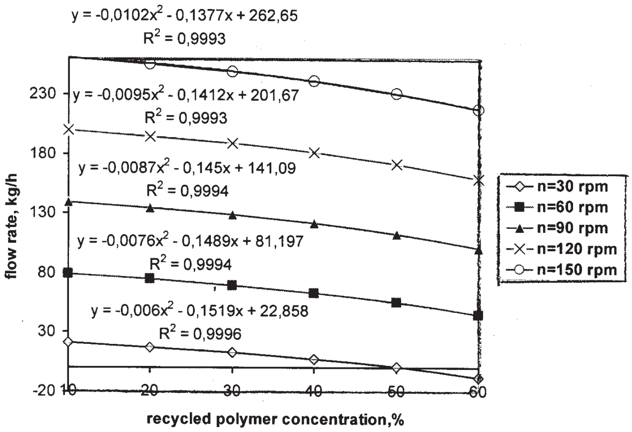

Fig. 3. Extruder flow rate variation versus the recycled material concentration (from the feedstock)

As it can be seen from the introduction is common to have as feeding material a mixing between new and recycled material. Now the trends are to increase the percentage of the recycled material to obtain new extruded products, up to 100%, even this is not yet often encountered in fabrication.

In figure 3 is shown the influence of the concentration of the recycled polymer from the feedstock on the flow rate, having as parameter the screw speed. It is obvious that the screw speed must increase to compensate the decrease of the flow rate caused by the increasing of the concentration of the recycled material (from the feedstock); the flow rate decrease is happening because of the decreasing of the melt viscosity and, consequently, of the backwards flow increase.

Conclusions

The rheological behaviour changing, due to recycling, is very important, generating in some circumstances, the ceases of the melt flow towards the extrusion head or obtaining lower performances (lower flow rates or higher energy consumption, higher production costs), so is very important when the polymer is many times recycled to know how much could affect the fabrication process. This means that experiments and simulations must be done.

The simulations performed showed that the screw speed increase is needed to compensate the material effective viscosity decreases due to recycling and because of this, the increase of the leaks backwards over the flight tip. The performances of the extruder are influenced by the pressure evolution along the screw channel, the worst situation being for $\left(\frac{\partial p}{\partial z}>0\right)$ in the metering zone of the screw. On the contrary, having the maximum pressure point before the metering zone, it was found to be favorable, for the case of using recycled material to produce extruded products. So the screw design must be done to create this favorable kind of pressure variation (correlated with the extruder head pressure drop). The use as feedstock 100% recycled polymer is possible but is compulsory to perform tests on the recycled polymer or at least to know the number of recycling (and to get information from literature about the changing in rheological properties), to know how to fix extruding parameters in order to obtain normal performances of the machine. This leads to the conclusion that is more efficient to process by extrusion on different machines the virgin and the many times recycled PP.

The simulations results showed, also, that a lower extruding temperature could be an option in reprocessing to lessen the thermo-modifications/destructions but this have to be specially studied in the coming time.

References

1.ŞTEFĂNESCU, M.F., SAUCIUC, R., DUMITRESCU, L., NIŢU, G., Equipment for recycling plastic food wraps, The 17th International Saloon HERVEX-2009, November 2009, p. 300-307.

2.ŞTEFĂNESCU, MARIANA-FLORENTINA, DURBACĂ, I., DUMBRAVĂ, VICTORIA, Recuperarea ambalajelor din polietilenterftalat, Simpozionului Naţional GEPROPOL 2009, Generarea, prevenirea şi procesarea emisiilor poluante în mediul industrial, 12 – 13 iunie 2009, Bucureşti, p. 342-352.

3.SCHUT, JAN H, Plastics Technology; May 2003; vol. 49, nr. 5, p. 57.

4.STTEIN, R.S., Polymer recycling: Opportunities and Limitations Proceedings of the National Academy of Sciences, Polymer Research Institute, University of Massachusetts, Industrial ecology, May, 1992.

5.AZAPAGIC, A., EMSLEY, A., HAMERTON, I., Polymers: the environment and sustainable development, John Wiley and Sons, 2003.

6.HUCKESTEIN, B., Plastics Recycling – Today and the Future, in the vol. The Polymer Processing Society, European Meeting, Stuttgart, 1995.

7.KHARE, A., DESHMUKH, S., Studies toward producing eco-friendly plastics, Journal of Plastic Film and Sheeting, **22**, July, 2006, p. 193.

8. TEODORESCU, N**.,** ŞTEFĂNESCU, M.F**.,** Ecological use of the solid polymeric wastes – Quality and economic limitations, in vol. The second Int. Conference on Polymers Processing in Engineering, Galati, Romania 22-23 Oct. 2009, p. 359-366.

9.TEODORESCU, N., PANDELESCU, (STROE) V.D., Study on the recycling of some polymeric materials characteristics. Experimental procedures, 4th International Conference on Advanced Materials and Systems, ICAMS 2012, p. 455-461.

10. TEODORESCU, N., PANDELESCU (STROE), V.D., Study on the recycling of some polymeric materials. Experimental analysis", 4th International Conference on Advanced Materials and Systems, ICAMS 2012, p. 449-455.

11.TEODORESCU, N., ŞTEFĂNESCU, M.F., Plastic wastes recycling-specific environmental properties Proceedings of the VIIth Edition of the International Conference The Academic Days of ATSR, Bucharest, 11-12 Oct. 2012, p. 349-355.

12. ŞTEFĂNESCU, M.F., TEODORESCU, N**.,** JUGANARU, M.R**.,** Ecological use of the solid polymeric wastes-Case study: Plastics properties modifications in the recycling process", in vol. The second Int. Conference on Polymers Processing in Engineering, Galati, Romania 22-23 Oct. 2009, p. 94 - 101.

13. TEODORESCU, N., STEFANESCU, M-F., RUSSU, M., TANASE, E-B., The influence of the recycled polymeric materials visco-elasticity on the pipe extrusion head design, Journal of Engineering Studies and Research, vol. 19, No. 2, 2013, p. 97-101.

14. TEODORESCU, N., STEFANESCU, M-F., PRODEA, I-M, DUMITRESCU, R., Extruder screw functional characteristic changing in polymeric materials reprocessing, Journal of Engineering Studies and Research, vol. 19, No. 3, 2013, p. 80-85.

15. SUSHAS, GHOSH, HORROCKS, A.R., The effect of waste polymer Inclusion on oriented PP, Geotextile Papers, Recycling of Fibrous Textile and Carpet Waste Conference, 1998.

16. HELSON, M., da COSTA, VALERIA, RAMOS, D., MARISA, C.G., ROCHA, Rheological properties of polypropylene during multiple extrusion, Polymer Testing, **24**, 2005, p. 86–93

17. J. AURREKOETXEA, M. A. SARRIONANDIA, I. URRUTIBEASCOA, Effects of recycling on the microstructure and the mechanical properties of isotactic polypropylene, Journal of Materials Science, **36,** 2001, p. 2607 – 2613

18. ANNA JANSSON, KENNETH MOLLER, THOMAS GEVERT, Degradation of post-consumer polypropylene materials exposed to simulated recycling-mechanical properties, Polymer Degradation and Stability, **82**, 2003, p.37–46

19.TADMOR, Z., KLEIN, I., Engineering principles of plasticating extrusion, Van Nostrand Reinhold Company, London, 1970.

20.JINESCU, V.V., A Model for The Extruder Feeding Zone, Polymer Processing Society, Strasbourg, August 1994, p. 195-196.

21.JINESCU, C.V., Mat. Plast., **40**, no. 2, 2003, p. 67

22.WOINAROSCHY, Al., JINESCU, C.V., POPESCU, I., Mat. Plast., **41**, no. 1, 2004, p. 11

23.JINESCU, C.V., Mat. Plast., **46**, no 3, 2009, p. 279

24.JINESCU, C.V., Rev. Chim. (Bucharest), **65,** no.7, 2014, p. 865

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