

re  
 spectra  
 ok

## THE USE OF RADIATION FOR THE PRODUCTION OF HIGH MELT STRENGTH POLYPROPYLENE

A. B. LUGAO, E.C.L. CARDOSO  
 Institute for Energy and Nuclear Research – IPEN-CNEN/SP, Brazil

B. HUSTZLER  
 EMBRARAD – Jurubatuba – Sao Paulo, Brazil

S. TOKUMOTO, A.N.F. MENDES  
 OPP Petroquímica, III Pólo petroquímico, Triunfo, Brazil

PRODUÇÃO TÉCNICO CIENTÍFICA  
 DO IPEN  
 DEVOLVER NO BALCÃO DE  
 EMPRÉSTIMO

Controlled rheology PP or visbreaking PP was already introduced in the market and produced in some case by irradiation. For example, Williams [1], discloses a process for reducing the molecular weight of a propylene polymer by activating a first portion of the polymer by exposure to ionizing radiation, adding the irradiated polymer to a second portion of unirradiated polymer, adding a stabilizing amount of an antioxidant to the mixture, and visbreaking by shear mixing in an extruder.

PP have been extensively studied, as already shown, during many years, and even industrially used to produce PP grades of controlled rheology, nevertheless it was never reported the use of irradiation of PP under N<sub>2</sub> to promote long chain branching in order to improve the elongational viscosity before the clever patents of Scheve [2] and DeNicola [3] to Himont now Montell.

PP suffers from low melt strength, i.e., the melted PP does not exhibits an increase in resistance to stretching during elongation. It is well known that the melt-strength properties of a polymer increase with molecular weight and with long chain branching due to the increase in the entanglement level (high melt strength PP – HMSPP). In spite of been the most fast growing polymeric commodity nowadays those new grades of PP and its development have been barely studied and its general chemical characterization have been even less studied with few exceptions [4,5,6].

HMSPP as proposed by Montell patents are produced by low temperature and low dose irradiation of high molecular weight isotática PP in N<sub>2</sub> atmosphere. So the well-known reactions would be mostly degradation and crosslinking. Degradation however is supposed to be the first and more intense reaction at the initial steps as already shown. So, according to Montell patents, another reaction is likely to occur, branching or T links competing with crosslinkings or H links. Radical are likely to decay very fast in amorphous phase, but under annealing the radicals entrapped in the crystal phase is likely to move to the boundary and react. The group of professor Silverman [7] has already hypothesized in T links formation and also studied the radical decay of PP. It is easy to understand the difficulties in differentiate Hs from Ts links and even these links if in very small amount from the bulk or from virgin polymer, as the chemical groups and links are chemically speaking essentially the same.

This work proposes a new method for the synthesis of HMSPP based on the combination of degradation, branching and crosslinking of PP under reactive atmosphere of acetylene and irradiation.

This new process was based on the simultaneous irradiation of high molar mass PP and acetylene, followed by the same recombination step as proposed in previous patents. Table 1. shows standards resins of OPP company and one sample prepared according to the new process.

International Symposium on Utilization  
 of Accelerators, nov. 26-30, 2001, São Paulo

Table 1. Rheological characteristics of the irradiated samples

Sample N°	sample	FlowIndex [g/10 min]	Melt Strength [cN]	Extensibility [cm/s]
1	Virgin H603 PP	1,50	16,34	7,94
2	Virgin H503 PP	3.50	9,3	11,2
2	C <sub>2</sub> H <sub>2</sub> irradiated 20 kGy	3.33	96.2	10.90

Table 1 shows the relation of IF with MS from commercial OPP resins. It is possible to get acquainted with the direct relation of IF and MS, as both are related to molar mass and branching structure. Also table 1 shows the rheological features of one PP subjected to IPEN-OPP proprietary process. It is possible to observe the tremendous enhancement in the processing characteristics of PP. Even increasing the flow index after irradiation, the resin showed much higher values of MS and also higher values of extensibility, showing improved performance. It is important to observe that if we compare the results of MS with standard resins of the same level of IF, the HMS resin had numbers of MS 10 times higher than the standard ones. So our proprietary process proved itself as capable of producing a HMS resin with excellent processing characteristics.

#### ACKNOWLEDGMENT

FAPESP 97/023960; CNPq/RHAE; PADCT project 450, IAEA BRA8/023. The authors are indebted with the staff at EMBRARAD and OPP Petroquímica, mostly on the rheological and IR characterization.

#### REFERENCES

- [1] Williams S.D. et al, (1998) Radiation Visbroken Polypropylene and Fibers Made therefrom, United States Patent 5,820,981.
- [2] Scheve et al. (1990) High Melt Strength, Propylene Polymer, Process for Making it, and use thereof. U.S. Patent 4,916,198.
- [3] DeNicola et al (1995) High Melt Strength, Propylene Polymer, Process for Making it, and use thereof. U.S. Patent 5,414,027.
- [4] Yoshi, F., Makuuchi, K., Kikukawa, S., Tanaka, T., Saitoh, J. and Koyama, K., 1986a. High-melt-strength polypropylene with electron beam irradiation in the presence of polyfunctional monomers, J. Appl. Polym. Sci. 60, 617-623.
- [5] Lugao A.B., Hutzler B., Ojeda T., Tokumoto SS., Siemens R., Makuuchi K. (2000) Reaction mechanism and rheological properties of polypropylene irradiated under various atmospheres. Radiat. Phys. and Chem. 57, 389-392.
- [6] Jones R.A., Salmon G.A., Ward I.M. (1994) An infrared Spectroscopic Study of Changes in Unsaturation Resulting from Irradiation of Isotropic and Uniaxially Oriented Polyethylene Films in The Presence of Acetylene. Journal of Polymer Science. Part B: Polymer Physics, vol. 32, 469-479.
- [7] Wenxiu C., Goldman J.P., Silverman J. (1985). Radical Termination rate constants and Mechanical properties of  $\gamma$ -irradiated isotactic Polypropylene with some additives. Radit. Phys. Chem., 25, n°s 1-3, 317-321.