TECHNO-ECONOMIC EVALUATION OF A POLYMERIZATION PROCESS: IMPACT OF DEGRADATION POTENTIAL ON PRODUCTION COSTS OF POLYPROPYLENE USING ALTERNATIVE ADDITIVES

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ABSTRACT

Simulation and economic evaluation for polypropylene process of Novolen® technology were developed by using Aspen Plus and Aspen Process Economic Analyzer (APEA). The simulation includes three fundamental parts of the polypropylene production process: polymerization, degassing and recovery. Process equipments were mapped by using technical information of existing equipments from a polypropylene plant. The influence of cost of raw materials on polypropylene production process with a production rate of 24.2 Tons PP/h and 3 wt.% of polyacetal (POM) as pro-oxidant promoter was evaluated. Economic evaluation indicates that propylene is the raw material with greatest influence on the production cost, followed by catalyst and POM. The net present value was positive after first years, and the payback period and net rate of return were 3.09 years and 110.41 mUSD respectively. POM concentration on PP/POM blends was varied from 1 wt.% to 5 wt.% to evaluate its economic impact. Studies indicated that the effect of the amount of POM on production cost is slight, however, the high amount of POM required for this process, may lead operational problems because existing infrastructure on polypropylene plants is not prepared to handle those level of additives.

Keywords: *Polypropylene; POM; Degradation; Economic Analysis; Simulation*

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1. INTRODUCTION

Polypropylene (PP) is a non-biodegradable and fossil-based synthetic polymer (Ahmed et al. 2018), which is obtained on an industrial scale from propylene and ethylene (used in copolymers) via Ziegler-Natta polymerization in gas phase, bulk and slurry technologies (Gahleitner and Paulik 2014). The global production of PP is calculated in around 55 MMT annually (Ceresana 2014) and is estimated to reach 75.22 MMT by 2022 (Research and Markets 2017). Its production on an industrial scale have experienced an increasing, starting with annual capacities of about 5 KT/yr in 1963 to more than 400 KT/yr for the newest plants being built now (Gahleitner and Paulik 2014).

Due to good chemical resistance, low density, attractive mechanical properties and good processability, PP is known as a versatile polymer (Abbas-Abadi et al. 2014). For instance, the extensive use of polyolefins in the daily life has led to PP to be one of the most commonly polyolefins used as packaging material (Shah et al. 2008). Last, even replacing paper and other cellulose-based products for their better physical and chemical properties (Shah et al. 2008); nevertheless concurrently it has become one of polyolefins most abundantly found in the plastic waste stream (Das and Tiwari 2018) with almost 20% of the spectrum (Saleem, Adil Riaz, and Gordon 2018).

Because of longevity and stability in the environment, the degradation rate of PP is too slow to be disintegrated completely (Ahmed et al. 2018) and its accumulation in the environment is a matter of great concern leading to long-term environment, economic and waste management problems. A wide variety of mechanisms of degradation of polymers has been assessed to led a deterioration of any chemical or physical property (Pospíšil et al. 1998). Degradation reflects changes of material properties such as mechanical, optical or electrical characteristics, in crazing, cracking, erosion, discoloration, phase separation or delamination (Shah et al. 2008) and it depends upon the nature of the causing agents (photo-oxidative degradation, thermal degradation, ozone-induced degradation, mechanochemical degradation, catalytic degradation and biodegradation). Molecular weight, size of the molecules, introduction of functional groups, additives and environmental conditions are some of important factors affecting the resistance of degradation (Singh and Sharma 2008). Synthetic polymers are considered to be non-biodegradable because of lack of conclusive evidences of degradation (Parthasarathy 2013).

Photodegradation processes of PP with different chemical structures (Tang, Wu, and Qu 2005) and PP nanocomposites (Mailhot et al. 2003) have been investigated without any significantly alteration on the rate of photooxidation of PP, but with an interesting reduction of efficiency of additives (antioxidants). Main advances to promote degradation have been obtained in polypropylene blends by incorporation of nanotubes of semiconductors like TiO_2 (Mailhot et al. 2003) or addition of metal-based pro-oxidants agents such as iron and cobalt stearates in low-density polyethylene (LDPE) and PP (Subramaniam et al. 2018). These pro-oxidants promote a controlled polymer degradation due to higher susceptibility to thermal degradation (Roy et al. 2007). Polyacetal (POM) has been studied as pro-oxidant agent, which promote the thermal degradation of PP (de Carvalho and Rosa 2014) at concentrations above 3% by weight of the POM (de Carvalho, Silveira, and Rosa 2013) limited only by the miscibility at the interface of the blend PP/POM (Paul, Sr., Forschirm, A., Glick, M., Jaffe 1990).

Notwithstanding, most of methods of degradation are not feasible due to the cost of manufacturing, the high sensitivity of the process to the cost of the raw materials, or simply lack of sufficient material to meet market demand. The aim of this study was to evaluate the impact on production costs by incorporation of POM in powder form to PP in a new polypropylene plant with a production rate of 24.2 Tons/h. Gas-phase polymerization process was simulated using Aspen Plus®. Production cost profiles were generated using the Aspen Process Economic Analyzer (APEA)®.

2. METHOLODOGY

2.1. SELECTION OF POLYPROPYLENE TECHNOLOGY

Polypropylene production process at industrial scale has a close relation to catalyst technology and product development. PP process technologies can be grouped into three main categories by the medium in which polymerizations take place: gas phase, bulk, and improved slurry as is shown in Table 1 (Gahleitner and Paulik 2014). Gas-phase processes are most versatile, because it can be used on the polymerization of ethylene, PP, and higher α-olefins. In the case of PP, polymer is formed around heterogeneous catalyst particles with very high melt flows (low viscosities) and high co-monomer contents. PP can be produced in both fluidized-bed reactor (FBRs) and stirred bed reactors (SBRs); in FBRs a gaseous stream of monomer and a gas fluidizes the polymer particles, while in mechanical stirring it is responsible for suspending the polymer particles in gas-phase SBRs (Gilbert 2016).

Table 1. List of polypropylene process technologies.

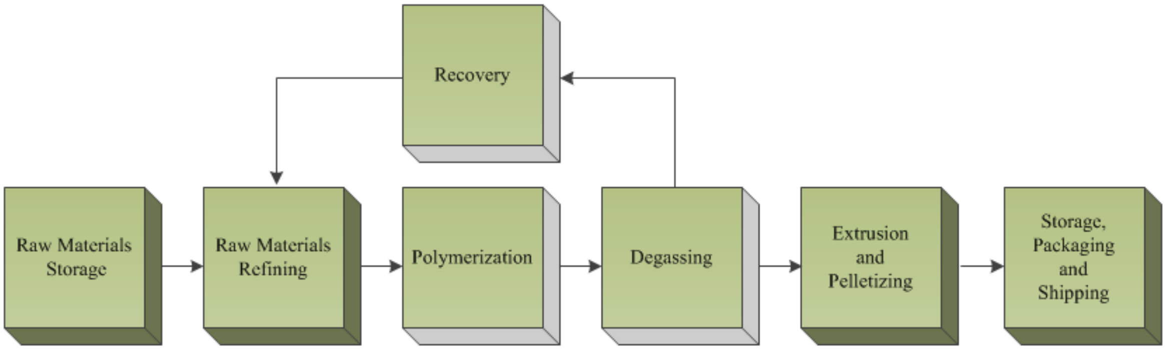
Regime	Technology
Gas phase	Unipol (Grace)
	Innovene PP (Ineos)
	Horizone (JPP)
	Spherizone (LyondellBasell)
	Novolen (CB&I´s)
Bulk	Spheripol (LyondellBasell)
	Borstar (Borealis)
	Hypol II (Mitsui)
	ExxonMobil
Slurry	Several

One of the most widely known technology for gas-phase polypropylene process is CB&I's Novolen®. CB&I's Novolen® licenses the gas-phase polypropylene process on the basis of Ziegler-Natta and metallocene catalysts for the production of a full range of polypropylene resins with highly reliable and versatile process (Novolen 2018). Novolen design consists of a vertical stirred reactor with a bottom-mounted helical stirrer maintaining uniform conditions in the polymer bed. This stirrer is designed to convey the particles up the reactor wall and let them fall down through the center of the bed. The heat of polymerization is removed to control the reaction temperature by circulating monomer gas through an external heat exchanger and pump (Asua 2007). This process has a simpler polypropylene powder discharge system downstream reactor, in which purge and desorption steps give the possibility of producing polypropylene with a very low level of odor. Polypropylene production capacity has led to an output of about 230.000 Tons/yr in a 75 m^3 reactor (Karger-Kocsis 1999). Novolen technology will be simulated in this paper, because is one of the polypropylene technologies installed in Colombia, South America in Esenttia (Polipropileno del Caribe S.A.) a 100% Colombian company dedicated to the production and commercialization of polypropylene, polyethylene and masterbatches (Esenttia 2018).

2.2. PROCESS DESCRIPTION

The process flow diagram for polypropylene production is basically made up of a raw materials storage process, raw materials refining process, polymerization process, degassing process, recovery process, extrusion and pelletizing process and storage, packaging and shipping processes, of which polymerization and extrusion processes are the core of the global process (Ross and Bowles 1985). A schematic process flow diagram of this process is shown in Figure 1.

Figure 1. Process flow diagram for polypropylene production.

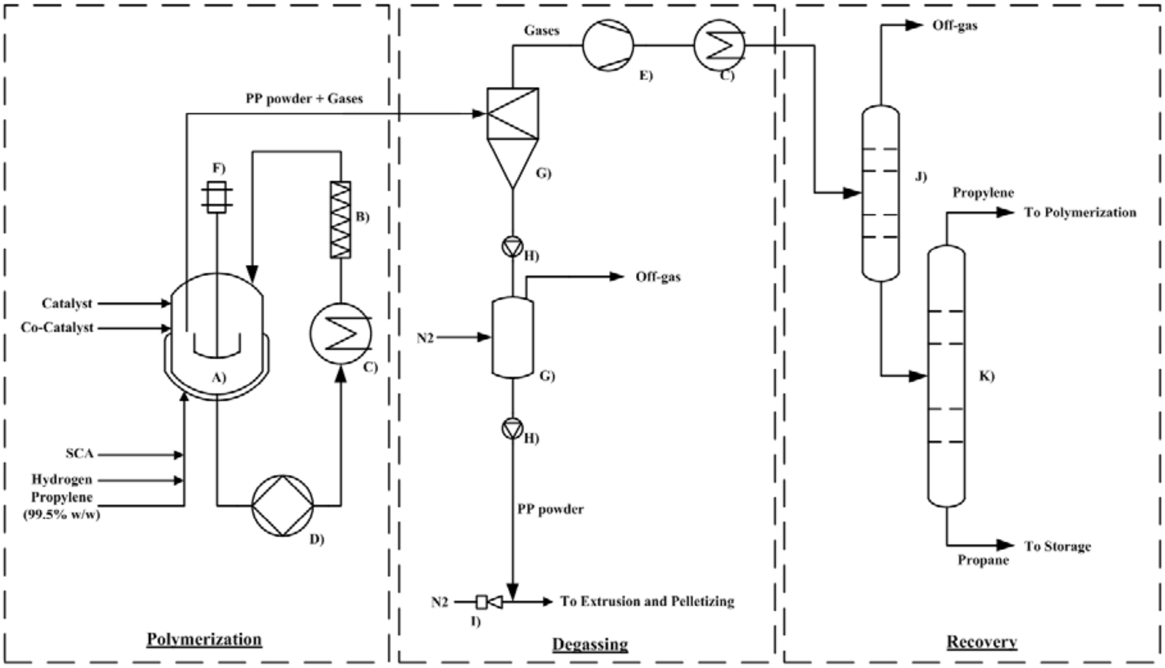


Often the raw materials are directly received from petroleum refinery by pipeline or are imported by ship and then stored at cryogenic temperature. The raw material process is a process for removing large amounts of impurities, such as water, oxygen, carbon monoxide, dioxide monoxide, sulfides and among others, that affect the catalyst activity in downstream processes. The polymerization process is the stage where polymerization reaction takes place. This stage brings the propylene monomers and other monomers into contact with a solid catalyst having polymerization activity. The polymerization is carried out at the active points of a catalyst where polypropylene powder is formed. Both polypropylene powder and unreacted monomers are extracted from reactor and separated in the degassing process. Unreacted monomer gases are returned to recovery area and subsequently to the end of refining process to be utilized in the reactor again (Fiscus, Doufas, and Datta 2017). The extrusion and pelletizing processes are the furthest downstream process, and they are a combined process for melting and kneading additives and fillers into the polypropylene particles that have undergone the after-processing and forming pellets. Of these processes, storage, packaging and shipping are indispensable further downstream in commercial plants, but since they are not unique to polypropylene manufacturing processes, they will not deeply touch in this paper, while polymerization, recovery and degassing processes will be subject of evaluation.

2.3. PROCESS SIMULATION

The scope of the simulation comprises the polymerization, degassing and recovery processes, which are important units of the polypropylene production process. The production rate of polypropylene used in the simulation is around 25 Tons/h. Figure 2 gives corresponding simplified polymerization diagram of the Novolen technology using one vertical stirred gas reactor (gas-phase) and the major equipments belonging to downstream processes.

Figure 2. Simplified Gas-Phase Novolen process for polypropylene production based on “Polypropylene” (Gahleitner and Paulik 2014); A) Reactor; B) Filter; C) Condenser; D) Pump; E) Compressor; F) Agitator; G) Receiver and Purge Tank; H) Rotary Valve; I) Blower; J) Deethanizer; K) Depropanizer.



In this process, propylene grade polymer (PGP) at 99.5% w/w and any other desired comonomers are fed into the reactor, like the hydrogen, catalyst, co-catalyst and selective control agent. Table 2 summarizes the inlet flows that were used for polymerization process.

Table 2. Inlet flow of components used for homopolymer gas-phase production

Species	**Function**	**Flow**	**Unit**
PGP (99.5%)	Monomer	24906	kg/h
Hydrogen (H_2)	Chain transfer agent	1450	gr/h
Titanium Tetrachloride ($TiCl_4$)	Catalyst	1650	gr/h
Triethyl-Aluminium (TEAl)	Co-Catalyst	1000	gr/h
SCA (Silane)	Co-Catalyst	900	gr/h

Polymerization conditions (temperature, pressure and reactant concentrations) are set by the homopolymer production. The reactor temperature ranges from 80°C to 85°C and the pressure ranges from 30 bara to 35 bara. Table 3 shows a comparison of different reactor operation conditions for gas-phase processes (Asua 2007). The reaction itself is exothermic and reactor cooling is achieved by flash heat exchange, where liquefied reactor is mixed with fresh feed and injected into the reactor. Flash evaporation of the liquid in the polymer bed ensures maximum heat exchange.

Table 3. Comparison of different reactor operation condition for homopolymer

Process	**Reactor type (Homopolymer)**	**Mode of operation**	**Temperature (°C)**	**Pressure (bar)**	**Residence time (h)**
Unipol (Grace)	Fluidized bed reactor	Condensate gas phase/gas	60-70	25-30	~1
Novolen (CB&I´s)	Vertical stirred gas	Gas/gas (non-condensed)	80-85	30-35	~1
Innovene (Ineos)	Horizontal stirred gas	Gas/gas	60-70	25-30	~1

Once polymerization process is carried out, polypropylene powder is discharged from reactor to degassing system, where is primarily separated from the unreacted monomer in a discharge vessel at atmospheric pressure and flushed with nitrogen in a purge tank to strip it of residual propylene. The remaining part is returned to the upstream recovery area to recover propylene and remove accumulated propane. Finally, powder is fed via gravity to the extruder, where it is then converted into pellets that incorporate a full range of well-dispersed additives.

Aspen Plus V9.0 was used for the simulation of the complete process. PC-SAFT equation of state (EOS) was employed to describe the thermodynamic properties of polymer system (Luo et al. 2009), while Peng Robinson was used to describe the thermodynamic properties of recovery area for separating a propane/propylene mixture (Fiscus, Doufas, and Datta 2017) (Lee et al. 2017). The reactor calculation was based on the kinetics reported by the technology provider Novolen® (with confidential agreement) until reaching 80% of conversion. For information on reaction rate constants used in this work and detailed information about the simulation process, the reader is encouraged to refer to Refs (Marin, Paula. Mogollón, Enrique. Álvarez, Juan. Cardona, Rodney. Giraldo, Oscar. Cardona 2018).

2.4. ECONOMIC EVALUATION

The economic evaluations were performed supplemented with data from datasheets of existing equipments in polymerization, degassing and recovery areas of polypropylene plant supplied by Esenttia (Polipropileno del Caribe S.A.). The energy and mass balances obtained from Aspen Plus were used to size the equipments. The cost of raw materials, products and utilities are given in Table 4 and Table 5. The costs are average values estimated reported by Esenttia during the year 2017.

Table 4. Raw materials and products price (Esenttia 2018)

Species	Price (USD/kg)
PGP (99.5%)	0.98
Hydrogen (H_2)	0.23
Titanium Tetrachloride ($TiCl_4$)	450 - 600
Triethyl-Aluminium (TEAl)	7 - 11
Polyacetal (POM)	3 - 5
Polypropylene	1.8 – 2.2

Table 5. Utilities cost (Esenttia 2018)

Species	Value	Unit
LP Steam	0.001 – 0.002	USD/kg
Nitrogen (N_2)	0.12 – 0.15	USD/Nm^3
Cooling water	0.009 – 0.0011	USD/kg
Electricity	0.08 – 0.12	USD/kWh

The costs for the following equipments were estimated with APEA based on dimensions, materials and specifications of existing equipments in the polypropylene plant: reactor, condenser, pump, tank, distillation column, filters and among others. The rest of process units were estimated based on information reported by Esenttia during commissioning and startup of the plant. The capital cost item (CAPEX) included the cost of equipment and installation, civil construction, electricity installation, instrumentation, land, engineering, and fees associated with the investment cost, while the operational cost item was based on Colombian conditions and separated into variable and fixed costs. Fixed costs included maintenance, insurance, working capital, and labor. The cost of working capital was assumed as 15%, the interest rate of 15% and the project lifetime of 10 years.

3. RESULTS AND DISCUSSION

3.1. CASE STUDY

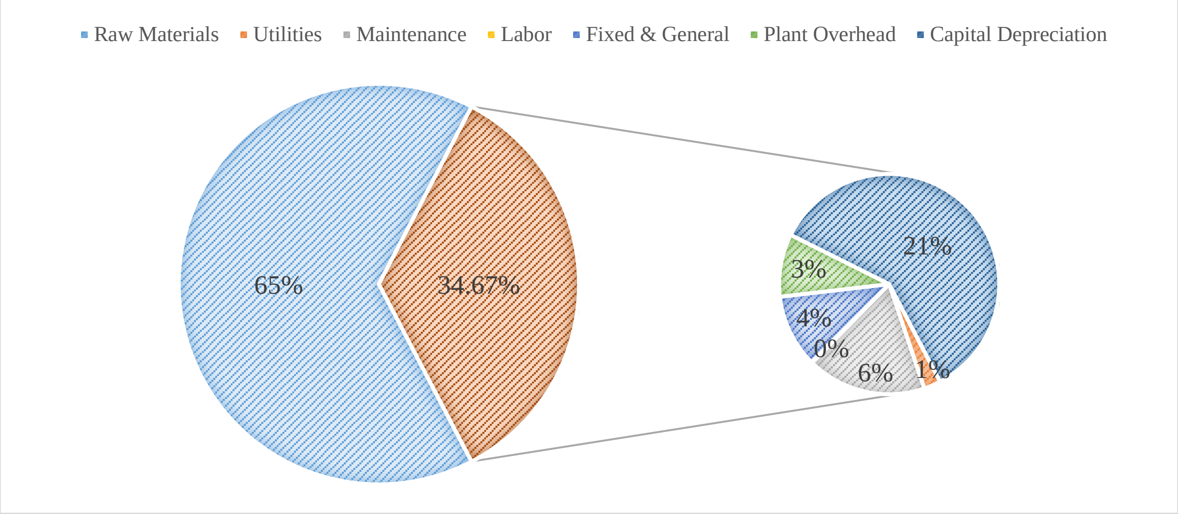
Three fundamental parts of the polypropylene production process were simulated in steady state by using Aspen Plus®: polymerization, degassing and recovery. Homopolymer production is conducted in a Novolen reactor by Ziegler-Natta polymerization, in which conversion is 80%. Propylene powder is discharged to degassing area, where gases are separated and sent to a recovery area, to get desired propylene with 99.5 mol% purity at the top, and propane at the bottom. Additionally, in this section the light gases (off-gas) are sent to the flare. In the downstream degassing area before extrusion and pelletizing, a new blend of polypropylene and polyacetal (POM) was evaluated with a mass concentration above 3 wt. % by weight of the POM. This additive promotes an early degradation in the polypropylene after consuming by end user. The impact in the production process of the amount of additive necessary to make polypropylene degradable is evaluated. The simulated rate of production of polypropylene in this work is around 24.2 Tons/h, if concentration of POM is 3 wt.%, the amount of POM required is 727 kg/h. Based on simulation of the process, Aspen Process Economic Analyzer (APEA) was used for calculating the net present value (NPV) and performing sensitivity analysis for new additive. Table 6 shows economic assessment of a polypropylene plant at 24.9 tons PP/h and 727 kg/h POM.

Table 6. Economic assessment of the polypropylene plant at 24.9 Tons/h

Unit	Value	Unit
Raw materials		
Raw materials cost	245.31	mUSD
Cost of utilities		
Utilities cost	3.24	mUSD
Interest rate	0.17	%
Lifetime	10	year
Maintenance cost		
Maintenance (6%)	19.83	mUSD/yr
Operating supplies (15%)	2.97	mUSD/yr
Labor cost		
Labor cost	0.18	mUSD/yr
Fixed charges		
Taxes (2%)	6.61	mUSD/yr
Insurance (1%)	3.31	mUSD/yr
Supervision (20%)	4.00	mUSD/yr
Total capital investment (CAPEX)	330.55	mUSD
Working capital (15%)	49.58	mUSD
Fixed capital investment (FCI)	280.96	mUSD

Figure 3 shows the distribution cost of the project represented mainly by raw material costs. It reflects the close relationship between the price of propylene and sale price of polypropylene. Any variation in the prices of raw materials (monomer) could impact strongly in the feasibly of the project because it represents almost the 65%. The remaining part is principally composed by capital depreciation and maintenance costs. Results show up on importance of recovery area for feasibility of the project, because is in this process where a large amount of propylene is recovered from unreacted mixture, about 5-6 Tons/h, almost 20% of monomer inlet flow to polymerization unit.

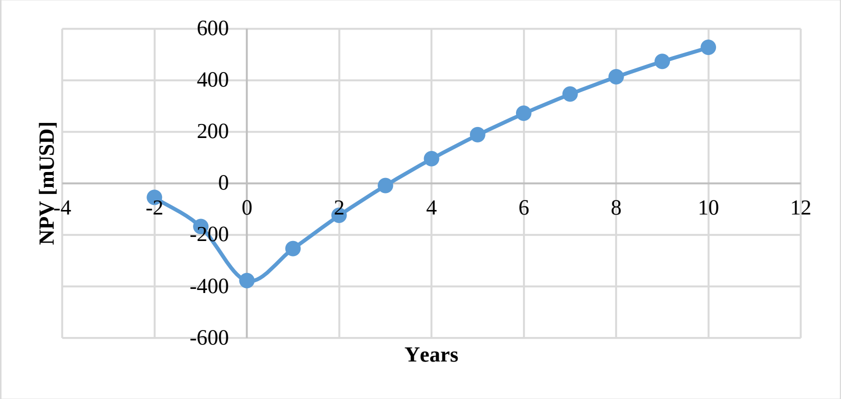
Figure 3. Distribution cost of the polypropylene plant at 24.9 tons PP/h and



3.2. ECONOMIC ANALYSIS

The net present value (NPV) is a capital budgeting technique used for evaluating projects, which include two financial streams: one is the cost stream and the other the income stream (Khatib 2003). Both streams contain all costs and incomes for polypropylene plant during lifetime of the project. For this process, the competitive operational life is ten years (10 years). The feasibility criteria of net present value (NPV), payback period (PBP) and net rate of return (ROR) have been estimated. The first step involved in the calculation of NPV is the estimation of net cash flows for the base case with a polypropylene production rate of 24.2 Ton/h. The second step is to discount those cash flows at operating and investment costs. The difference between the two streams is the cash flow along project lifetime. Figure 4 shows the net present values for the polypropylene production plant.

Figure 4. The values of net present value (NPV) over project lifetime.



The net present value is negative, particularly during engineering, construction and start-up and the early years of the project. When payback period is reached, initial cash outflow of the investment is expected to be recovered from the cash inflows generated by the investment. The project is accepted if at least two out of three criteria should be favorable for a feasible operation: NPV ≥ 0, PBP ≤ useful life of operation, and ROR ≥ interest rate. In this case, the NPN is greater than zero and the project is feasible, while payback period is 3.09 years and net rate of return is 110.41 mUSD. Those values are acceptable considering reported values in the literature (Anderson, Jhon., Fennell 2013).

3.3. SENSIVITY ANALYSYS

Sensitivity analyses were performed for variations from-100% to 100% in cost of raw materials and the selling prices of polypropylene. Figure 5 shows influence of cost of raw material such as propylene, hydrogen, catalyst ($TiCl_4$), co-catalyst (TEAl) and additives (POM) on feasibility of the project. This graph shows how variations in the cost of propylene move strongly the net present value to negative region. However, this result cannot be analyzed without context. The variance of polypropylene price is predominantly explained by variance of propylene price. Depending on price dynamic between propylene and polypropylene, a better understanding of how input costs and demand drive the product business This behavior explains the remarkable NPV tendency shown on Figure 5.

Figure 5. Influence of cost of raw materials.

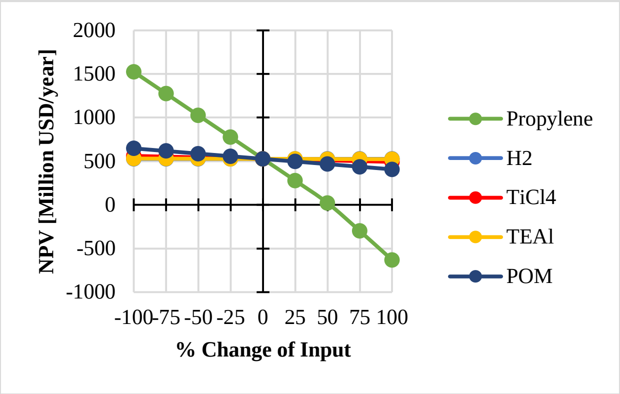


Figure 6. Influence of cost of products.

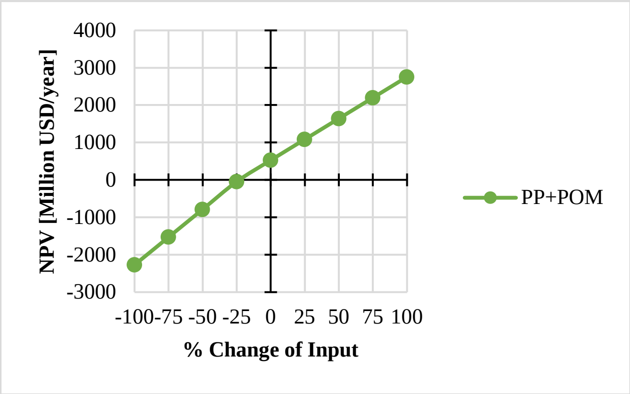


Figure 6 shows the influence of cost of polypropylene and POM on the profitability of the project. Nevertheless, this behavior must be analyzed carefully, because polypropylene price is highly influenced by propylene price (Naylor 2016) and neither inputs and nor outputs are fixed in this business. When the margin between polypropylene-propylene is less than break-even line, some polypropylene plants have to reduce operating rate (CCFGroup 2017). This entails to many contract are set based on propylene grade polymer (PGP) values plus an adder (Moore 2018). The other raw materials have a slight impact on the manufacturing process, however, POM and catalyst ($TiCl_4$) may affect the profit margin of the business if excessive amounts of them are fed during production process.

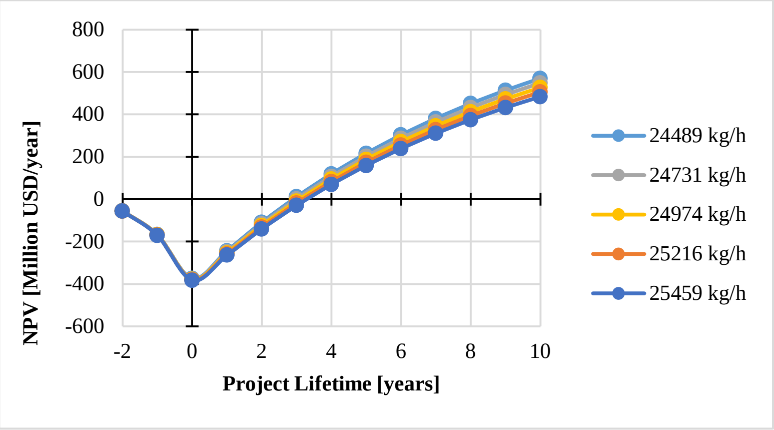
In addition, the effects of varying the concentration of POM in the blend PP/POM over NPV along project lifetime were investigated. The POM concentrations were varied from 1 wt.% to 5wt.% limited only by miscibility criterion of the blend. The amount of POM required for each case with a production rate of 24.2 Ton/h of polypropylene are presented in Table 7. Influence of variation of POM concentration on NPV over project lifetime is shown on Figure 7. The POM concentration has a slight influence on NPV due to high amount of POM in the product and its high cost.

Table 7. Amount of POM based on production rate of 24.2 Ton/h of PP.

PMO (wt.%)	POM (kg/h)	PP /POM (kg/h)
1%	242.4	24489
2%	484.9	24731
3%	727.4	24974
4%	969.8	25216
5%	1212.3	25459

The price of POM was added to product price, because high concentration of this additive in the blend with polypropylene (3 wt.%). For a high scale production rate of 24.2 Ton/h of polypropylene, 727 kg/h of POM are required to make degradable the polypropylene. Because as it is true that POM may reduce the lifetime of polypropylene, it is likewise so considerable amounts of POM entail operational implications related with preparation of blends and transport of additives. Just by the way of an example, dosage rate of antioxidant as Irganox® 1010 is added in an amount of 0.05-0.4 wt.% (BASF 2018), amounts much lower than amount required by POM. For instance, the ratio between Irganox® 1010 and POM is 7:1. This reflects that while in a day of 24 hours of operation, the required amount of Irganox® 1010 is 2.3 Tons, the amount of POM is 17 Tons. Currently, the polypropylene plants do not have equipments nor infrastructure for handling high flows of POM.

Figure 7. Influence of variation of POM (%) on NPV over project lifetime.



4. CONCLUSIONS

Gas-Phase polymerization process based on Novolen® technology for polypropylene production was simulated using Aspen Plus® with emphasis on polymerization, degassing and recovery units. Equipment costs involved in the simulation were rigorously evaluated using Aspen Process Economic Analyzer (APEA)® based on technical specification of equipments used in an existing polypropylene production plant. Economic analysis was performed using the net present value (NPV) as economic tool to calculate the feasibility of process along project lifetime with a production rate of 24.2 Tons/h of polypropylene and a POM concentration of 3 wt.%. The net present value was positive after first years. The payback period was 3.09 years and the net rate of return was 110.41 mUSD. The impact on production cost by the variation of raw materials cost, and especially by incorporation of a promising pro-oxidant like POM was also evaluated finding that propylene is the raw material with the greatest influence on the NPV, followed by the POM and catalyst. Minor changes on POM concentration (1 wt.% - 5 wt.%) affect slightly the NPV during project lifetime, due to considerable amount of POM required to make degradable the polypropylene. However, the major effects may be not related with affectation of product price but handling of tons of POM required in the process, because installed equipments are not currently designing for handling high quantities of additives. To do attractive the implementation of additives for degradation in the industry, many efforts must be done to reduce amount of additive to promote degradation. To improve the predictions of the simulator, specifically the economic mapping, it is recommended that the whole plant is modeled including raw material storage, refining, extrusion and pelletizing, conveying systems, product storage, among others.

5. ACKNOWLEDGMENTS

The authors express their gratitude to both COLCIENCIAS and Esenttia (Polipropileno del Caribe S.A.) for financial support and scholarships to develop the doctoral studies to which the present work belongs within the framework of the project of technological development CNBT-COLCIENCIAS code 665173454353 “Desarrollo del Producto y Proceso de Producción de Terpolímeros de Propileno-Etileno-Buteno”.

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