**Research Article**

# Migration Phenomena Analysis of Virgin PET /Recycled PET Multlayers Films

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## Abstract

PET is thermoplastic polyester prepared from terephthalic acid and ethylene glycol. It is used for the production of bottles, sheets, films, co extrusion and injection molded products. Presently, there is an increasing emphasis throughout the world to recycle PET. The use of recycled PET for food packaging applications however, has raised questions regarding its safety. Recycled PET presents a source of a wide range of potential migrants such as residues from the polymerization process and degradation compounds. When such materials are used to manufacture packaging materials intended to be in direct contact with food, these pollutants may migrate into food.

The aim of this work was the analysis of migrations in Multilayer film containing recycled PET from 3 layers plastic film. The film having the structures with recycled PET inner layers and virgin PET outer layers, Functional barriers form parts of multilayer packaging materials, deemed to protect the food from migration of a broad range of contaminants. The use of multilayer structures with a virgin polymer layer as a functional barrier may prevent such migration. Often, neither the presence nor the identity of the contaminants is known, so that safety assessment of the materials has to rely on predictive tools. Several complimentary freeware allow to model diffusion in multi-layer films. These tools require the input of parameters which are not easy to determine or to predict. However many other kinetic and thermodynamic parameters are needed to describe transport properties during processing of material at high temperature and during its shelf life all parameters needed for the calculations are discussed. In order to propose default values, the approach consists of

(i)     Reviewing available literature data

(ii)    Critical Analysis of Migration Model reports in literature

(iii)   Identification of default value from experimental validation conditions

The problem of models validation will be further discussed and, in order to make film samples available for the experimental evaluation of migration, multilayer cast film co-extrusion equipment was set up and multilayer films were produced for Migration experimental tests.

## ABBREVIATIONS

PET: PolyEthylene Terephthalate; SML: Specific Migration Limits; OML: Optimum Migration Limits; EVOH: Ethylene Vinyl Alcohol; HDPE: High-Density PolyEthylene; LDPE: Low-Density Poly Ethylene BOPP: Biaxially Oriented PolyPropylene; L/D: Length/Diameter; QM: Quantity Materials; GC: Gas Chromatography; MS: Mass Spectrometer; BP: Benzo-Phenone; PCH; Phenyl–Cyclo Hexane; MM: Methyl –Margarte; MW: Weight Average Molecular weight Mw; MN: Number Average Molecular Weight; MWD: Molecular Weight Distribution; FDA: Finite Differential Analysis; SM; Specific Migration; GPC: Gel Permeation Chromatography

## INTRODUCTION

The accumulation of plastic waste in landfill together with the increasing market share of plastic has encouraged recycling industries around the world to consider recycling post-consumer PET (polyethylene terephthalate) for food contact applications. Although recycling addresses the environmental concerns regarding waste disposal, there are serious health issues associated with the use of recycled polymers for food contact applications. A good approach for postconsumer food packaging materials is to make new packages from old packages. However, this raises food safety considerations, because post consumption collected packages may be polluted by common chemicals, available to households (detergents, petrol, garden herbicides, or

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pesticides, etc.). Numerous articles have studied the elimination of contaminants during processes such as washing, de-polymerization, post condensation, or extrusion under reduced pressure.

The evaluation of barrier performances of a material towards a given packaging application requires:

(i) Determination of whether contamination of the barrier occurs during processing of the multi-layer material [13,2] and

(ii) Determination as to whether the lag time for migration is lower than the shelf life of the packaging [2].

As the mass transport phenomena during processing and during food contact are mainly controlled by diffusion effects, the general strategy of the food packaging scientific community was to accumulate diffusion coefficient reference data:

(i) At molten state for processing conditions [13], and

(ii) At 40°C for food contact conditions [13]; these data, focused on low molecular weight migrants were used to complete the Piringer data bases and equations involving a large molecular weight range [4,5].

However migration is obviously not controlled only by diffusion, but also by partition coefficients, heat transfer, heat diffusion, mass transfer, diffusion coefficient activation, and factors describing the effect of plasticization on diffusion [19].

## BACKGROUND

### Definition of PET and its applications in the food industry

PET (polyethylene terephthalate) resin is a polyester polymer produced by the reaction of ethylene glycol with either terephthalic acid or dimethyl terephthalate and in the presence of catalysts including salts of manganese, cobalt, cadmium, calcium, lead, zinc, antimony, titanium and germanium. Polyesters, such as PET, are produced worldwide by companies such as Du Pont, Eastman, Monsanto, and Hoechst and are used in the manufacture of textile fibers, film, bottles and molding compounds [23]. PET was processed worldwide for these applications and the trend has been growing due to the polymer's superiority over glass for food packaging [5].

The use of PET as a film and bottle has been successfully incorporated into the food-packaging industry due to the polymer's strength, light-weight, flexibility, clarity, resistance to high temperature and its negligible permeability to carbon dioxide, which is especially important in the packaging of carbonated soft drinks [6,2]. PET bottles are also used in non-food contact applications, such as storage for toiletries, cosmetics and other household products [23].

### Migration considerations in food contact applications

One of the most important processes that occur as result of packaging food is migration, defined as the mass transfer of chemical substances (monomers, oligomers, additives and residues plasticizers) from the packaging material to the foodstuff. Any substance which migrates from the packaging into the food

is of concern because it could be harmful to the consumer. Even if the migrating substance is not potentially harmful it could have an adverse effect on the flavor and acceptability of the food. A series of chemical and biologic processes take place into the food itself and there is an interaction between the food and the packaging material, too [10]. The uptake (absorption) of food components by a plastic may lead to a considerable change of the initial properties of the plastic material (for example by swelling). This increases the rate of substance transfer from the plastic into the food and may also accelerate the degradation of the plastic packaging [3,5]. The other process which is the release of substances from the plastic packaging into the food is commonly named "Migration" and it is a quite complex process. The rate and dynamics of a migration process depend on a series of factors among which the concentration of the substance (migrant) and its mobility (diffusion coefficient) in the plastic matrix and the solubility of the migrant in the food play an important role [3,5]. Both phenomena occur due to the existence of substance concentration gradients between the plastic packaging material and food and due to the fact that the boundary between these two entities exhibits a non-zero mass permeability in both direction, i.e. substances can cross the boundary both from the food towards the plastic and vice versa [3,5]. Both processes can be investigated theoretically and experimentally with a high degree of accuracy and thus information can be obtained about the parameters and conditions which influence them as well as about the level of accumulation of migrants in a certain packaged food. Moreover, it has been shown in recent years that most of the migration processes taking place between plastic packaging and food can be estimated using theoretical models and adequate computer software [10].

The consumer protection problem with the migration process is that people consuming food packaged in plastic materials are inherently exposed to a series of substances, originating not from the food itself but from the plastic, which may become a health concern if they exceed certain limits. In other words this means that if a plastic material swells or even degrades when it is in contact with a certain food then this article is not suitable for manufacturing a packaging for the said food. From the point of view of interactions between plastic packaging and food the main aspect of concern for the consumer's health are the substances which migrate from the plastic into the food. In EU Directive there is a list of authorized substances and Specific Migration Limits (SML) for certain substances; the transfer of constituents which could endanger human health is addressed. The Optimum Migration Limits (OML) is understood as a guarantee for a sufficient inertness of the plastic material or article to avoid an unacceptable change in the composition of the foodstuffs [3].

### Multilayer, Flexible film Food Packaging

Flexible packaging has been the fastest growing segment of packaging industry since the 1970s. An important attribute of flexible packaging is its ability to form thinner, lighter, more compact packages. Regarding permeability, much effort has been devoted to develop new structures with barrier properties since oxygen is one of the most important components that determines quality and shelf life of foods. Over the last few years, a lot of development has been made in the area of new barrier

technologies. The innovations go along five major lines:

- Thin, transparent vacuum-deposited coatings,

- New barrier polymers,

- Blends of barrier polymers and standard polymers,

- Organic barrier coating or adhesives and

- Nan composite materials [13].

Multilayer coextruded or laminated flexible materials are a significant development in modern packaging technology. There are many possible combinations of single materials in a coextrudate. Most coextruded multilayer structures are based on polyolefin's because of their low cost, versatility, process-ability, chemical inertness and high moisture barrier properties, accompanied, however, by their poor ability to inhibit oxygen and aroma transitions. These multilayer structures are produced by processing of commodity plastics, such as virgin PET, Recycled PET and nanocomposite through their respective extruders and a feed block and then through a die to give a five layer films [19,11]. Polyamide resins are also an important class of polymers used in the packaging industry and owe their popularity to good barrier properties against oxygen, aromas, and organic solvents along with high tensile strength and toughness [10]. EVOH possesses excellent barrier properties to oxygen, aromatics and oils. Oxygen diffusion through EVOH is limited by high intermolecular and intermolecular cohesive energy. However, this polymer is hygroscopic and absorbs water at an elevated relative humidity and then, it loses much of its oxygen barrier performance. Therefore, the use of co extrusion to combine Virgin PET, recycled PET and nanocomposite in a multilayer structure is a very attractive process for many demanding applications, such as food, drugs and cosmetics. Co extrusion, however, is often a very expensive procedure and requires a complex system of control. Therefore, an alternative approach employing polymer blends appears more efficient and economical for the design of products consisting of a barrier material and polyester. The blend processing may be carried out in a single-step operation and offers versatility combined to low cost. DuPont Company has developed blends of high-density polyethylene (HDPE) and nylon for packaging agricultural chemicals or making organic solvent containers and fuel tanks. In addition, low-density polyethylene (LDPE)/EVOH and poly (ethylene terephthalate (PET)/EVOH blends for food packaging films. Poly (vinylidene chloride) has also been proposed as barrier polymeric film, but it is said to produce dioxins when it is burnt out at low temperatures for waste disposal [23]. Metalized films and/or structures using metalized films are an additional option that can greatly enhance barrier properties of BOPP film [22].

Improvement of fundamental food packaging characteristics, such as barrier properties, strength, antimicrobial properties and stability to heat and cold, could be achieved using nanocomposite materials [1]. Research on the use of nanocomposites for food packaging began in 1990s. Most of the research has involved the use of montmorillonite clay as nanocomponent in a wide range of polymers, such as polyethylene, polyvinyl chloride and starch. Amounts of nanoclay incorporated vary from 1% to 5% by weight. Nan components must have one dimension

less than 1 nm wide. The lateral dimensions, on the other hand, can be as large as several micrometers, leading to high aspect ratios of many of these materials. The high surface area results in unique properties when nanocomposites are incorporated into packages. There are three common methods to process nanocomposites: solution method, in situ polymerization and melt processing. Bayer Co produces transparent nanocomposite plastic films and coatings called Durethan, which contains clay nanoparticles dispersed throughout the plastic. Large amounts of silicate nanoparticles are dispersed in polyamide films. These nanoparticles block oxygen, carbon dioxide and moisture from reaching fresh meats and other foods. The nanoclay particles act as impermeable obstacles in the path of diffusion process, there by extending the shelf life of foods while improving their quality. The final package is also considerably lighter, stronger and more heat-resistant. A considerable amount of research is also running in the area of biodegradable nanocomposite food packages, since that kind of films can act as very effective moisture barriers by increasing the tortuosity of the path water must take to penetrate the films. Significant increases in film strength are also frequently achieved in these types of materials.

Traditional food packages are passive barriers designed to delay the adverse effects of the environment on food products. Active packaging, however, allows packages to interact with food and the environment and play a dynamic role in food preservation. Developments in active packaging have led to advances in many areas including delayed oxidation and controlled respiration rate, microbial growth and moisture migration. Other active packaging technologies include carbon dioxide absorbers/emitters, odor absorbers, ethylene removers and aroma emitters. Furthermore, active packaging technology can manipulate perm selectivity, which is the selective permeation of package materials to various gases [8].

## MATERIALS AND METHODS

### Extrusion process

The PET P60 and Recycled PET were dried under vacuum at 120°C for 24 hours before processing (Figure 1). The multilayer films were produced by cast co-extrusion, using three 20 mm screw extruders having 25:1 L/D ratio. Feed-block technology was used to assemble the extrudate layers into a three-layer film by way of a 200mm adjustable thickness slot die and temperature regulated cast film chill rolls. In particular, A and C were virgin PET and B was Recycled PET the Pattern "ABC" three layers structures. The temperature profiles set from the feed zone to melt zone were: 285, 285 285,280,280°C respectively for the extruder fed with pure PET and 280 ,280,280,275,280°C for the extruder fed with the Recycled PET. Temperature profile set for Die Head from zone 1 to zone 3 were 290, 290, 290°C respectively. The take-up system consisted in a couple of chill rolls thermally controlled by water circulation at 15°C and in a multi roll system to draw the produced films. The chill roll speed was 4 m/min, 8m/min, and 12m/min at screw speed of each extruder was 40 RPM with a thickness of the films were produced 150 μm, 100 μm, and 85 μm respectively. In order to realize structures with different relative mass ratios, the curve of mass rate vs. screw velocity have been determined for each material fed in the proper

**Figure 1** Laboratory set up of Cast film Co-Extrusion.

extruder. In particular the Recycled PET was fed in the extruder B, while the pure PET in the extruder A and extruder C. The nominal mass ratios of materials contained into each layer for a total extruded mass rate of 103g/min were reported and finally produced multilayer film for migration test.

**Migration test**

The study was conducted on a food grade PET (IV = 0.6 dl / g) supplied by MONTEFIBRE. The polymer was contaminated in laboratory holding the granules in contact (for 14 days at 25°C) with a mixture in toluene containing 0.1% by weight of three contaminants surrogate Benzophenone(BP), PhenylCycloHexane(PCH) and Methyl Margarato(MM). Subsequently, the PET was subjected to three cycles of processing by extrusion before each cycle; the PET was dried in a vacuum oven for 24 hours at 120°C (Table 1).

**Extraction of contaminants (QM):** The determination of the residual content (QM) of the contaminants in surrogate specimens of PET subjected to one, two and three cycles was performed by GC / MS / SIM subjecting to extraction of contaminants 0.5g of specimen ground.

**Determination of specific migration of contaminants:** The release tests were carried out according to the test conditions using the protocol adopted by the EU CEN (EN1186) for tests with simulants. Below are the solvents simulants and contact conditions used: isooctane (2 days at 20°C), acetic acid 3% (10 days at 40°C), 95% ethanol (10 days at 40°C); ethanol 50% (10 days at 40°C). The determination of migrants was performed using the same conditions GC / MS / SIM used for the determination of the amount of residual contaminants QM.

## RESULTS AND DISCUSSION

It was very difficult to adjust the melt flow of the polymer coming out from flat die. Several Types of instabilities and defects were occurred during extrusion process. The production of multilayer film was limited because interfacial instabilities of high frequency zig-zag or long wave type occurred in the pre-die region and film neck-in, edge beads, and draw resonance arisen in the post-die region. A three-layer film casting process, i.e., A-B-C type, has been selected to analyzed and adjusted draw resonance instability of the system toward various other process

conditions, After we did many experiments we adjusted the melt flow of the polymer ,rotation speed of the chill roller and the air flow rate and produced sample film. The rotation speed of that Chill roll was control the draw ratio and the final thickness of the film.

In order to determine the average thickness of each of the respective samples, four pieces of 10 x 10 cm were cut from each type of film. The thickness was measured on five locations, the four corners and the centre of each sample, using a hand-held micrometer with a resolution of + 0.005 mm the quoted sample thickness represents an average of the 5 x 5 measurements.

One of the most promising techniques to improve the properties of film for packaging is represented by the co-extrusion process. In general, these processes allow combining two or more polymeric materials at molten state, in order to obtain a single structure. Therefore, to guarantee a good adhesion among the different layers, the materials have to show a good chemical and rheological affinity.

Process parameters to be set for production of Multilayer structures such parameters are

- Screw speed of each extruder
- Temperature profile of each extruder
- Temperature of the cylinder , adaptor and extrusion head
- Water flow for cooling
- Speed and temperature of chill rolls
- Speed of the pulling roll and collecting roll
- Air flow for cooling

It is important to understand that the changes in screw speed during the trials were done in order to alter the thicknesses of the film layers. In doing this, the analysis of the sample films helped us understand how each individual layer affected the overall properties of the entire multilayer film and provided a method to design a film structure with properties for a particular application. The expected properties of the multilayer films were developed under the assumption that the individual polymer materials would be have similarly in a multilayer co-extruded structure as they would in a monolayer extruded film. This assumption is neglecting the effects of altered orientation and crystallinity in a multilayer structure compared to a monolayer film. The layer percentages of the film components were also

**Table 1**: Contaminants and surrogates.

| Surrogate Contaminate | Molecular Formula | Molecular weight | $T_m$(°C) | $T_b$(°C) | QM(mg/g) |
|---|---|---|---|---|---|
| Benzo Phenone (BP) | $C_{13}H_{10}O$ | 182 | - | 305 | 6.8 |
| Phenyl –Cyclo Hexane (PCH) | $C_{12}H_{16}$ | 160 | - | 238 | 4.5 |
| Methyl -Margarte (MM) | $C_{18}H_{36}O_2$ | 284 | 33 | - | 6.0 |

**Abbreviations:** BP**:** Benzo Phenone; PCH: Phenyl –Cyclo Hexane; MM: Methyl –Margarte; $T_m$: Melting Temperature; $T_b$: Boiling Temperature; QM: Quantity Materials
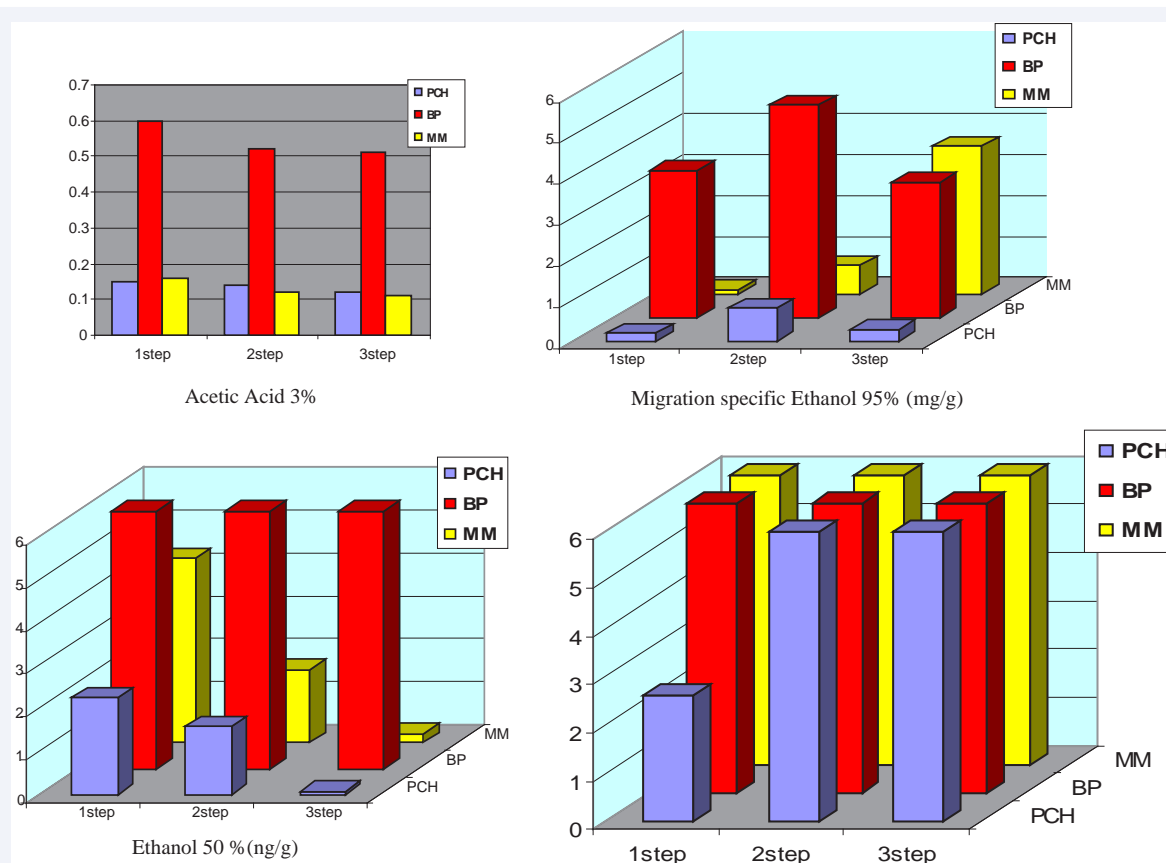
**Figure 2** Result of The determinations of QM.

varied by adjusting the screw speeds of the extruders. The screw speed of each extruder directly influenced material output and the corresponding layer thickness of the material in the multilayer film. The effect of layer thickness on film properties was examined so that the film and layer thicknesses needed to achieve a desired barrier property value could be estimated.

➢ After each processing cycle, there was a decrease in the values of QM of each contaminant. The decrease of the values of QM between the first and the third step is equal to 20% for the PCH, to 15% for the BP and to 31% for the MM (Figure 2).

**Factors that contribute to the decrease of QM were**

➢ Loss due to migration of contaminants

➢ Effects due to treatment of post consumer (washing, etc.) Loss of contaminants during the process of extrusion.

After each processing cycle, the values of SM in 95% ethanol were always higher than those measured in 3% acetic acid and 50% ethanol.

**Causes**

• The 95% ethanol interacts strongly with the PET polymer and penetrating into the structure swelling.

• The contaminants have a different solubility in the three solvents: when migrants are totally soluble in simulants (as in the case of ethanol 95%) the speed of the migration

process depends only on the rate of diffusion of the migrant in the polymer; instead, when migrants are poorly soluble in simulants (as in the case of acetic acid 3% and 50% ethanol), the speed of the phenomenon is limited by the partition coefficient of the migrant between polymer and simulants (Figure 2).

➢ The BP always presents the values of SM higher in all three simulants. This may be due both to the greater amount of residual BP present in the polymer, both to its higher mobility. The BP, in fact, has a planar structure that facilitates the spread.

➢ The differences in the Migration behavior of PET compared to the three contaminants and in the three simulants can be attributed to the change in migrant-polymer interactions and polymer- simulants as a result of recycling. In fact, PET, subject to subsequent recycling operations, undergoes significant changes in the parameters of molecular structure (Mw, Mn, MWD) (3-5) that modify the physical and Migration.

➢ The values of SM species migrants are strictly dependent on their concentration in the initial polymer. Therefore, the properties of PET Migration can be more correctly described by the analysis of the ratio SM / QM.

➢ GPC test on molded specimens of non contaminated PET, PET and PET contaminated and subjected to a recycling operation have shown that already after the first processing, and even more after the recycling, there is a remarkable decrease of

the weight average molecular weight Mw, and a very sharp drop in the number average molecular weight Mn, with consequent increase in the poly-dispersity (Mw / Mn) (Figure 3).

In conclusion of experimental result, The properties Migration of contaminant in PET recycling subjected to depend not only on the amount of residual contaminants present in the polymer but also by the type and number of processing cycles performed. The differences in the migration behavior of the polymer compared to the three contaminants and in the three simulants were attributed to the different chemical and physical characteristics of migrants and the change in the migrant-polymer interactions and polymer-simulant following the execution of repeated cycles. The PET fact, after thermo-mechanical degradation phenomena induced by recycling, undergoes significant changes of the parameters of molecular structure (decrease of average molecular weight, increase in the oligomeric fraction and the number of carboxyl terminal) that modify the physical properties and migration. Therefore, for the purposes of prevention of contamination of foods, to evaluate the potential contaminant of a recycled PET is indispensable knowledge of the composition of the starting material, the structural parameters of the polymer and the operations carried out for the recycling of the polymer.

## CONCLUSIONS

In conclusion, predicting migration is a promising cost and time saving alternative to migration experiments. Deterministic models do not give information on the variability or the uncertainty in the migration values. Stochastic and probabilistic approaches have proved to be excellent tools in a number of research areas and are receiving increased interest in packaging research dealing with migration. Stochastic models have the advantage of giving a more realistic prediction in combination with a probability distribution, which enables a better risk evaluation. However, the use of any migration model must be accompanied by knowledge on the scientific background and application area of the model. For regulatory evaluation, the uncertainty of overall migration is set to be 2 mg/dm2 or 12 mg/kg. However, for migration of specific compounds the variability of the diffusion and partition coefficients, for example, is not known. Therefore, probabilistic approaches in this field still require considerable research effort.

- In Europe, the Piringer model has been evaluated and a comparison of model predicted values with experimental results has shown that, in 95% of the cases, the model overestimates the migration values [14-16,7]. This means that the model can be used to support and verify compliance with the regulations, ensuring a margin for consumer safety. However, the need for models to be able to produce estimations close to the real values, reducing the overestimations [7] and reducing the risk of rejecting safe packaging systems has been recognized.

- Most of the work developed concerns deterministic modeling and the models were developed to assess compliance with regulatory limits. In this case, model
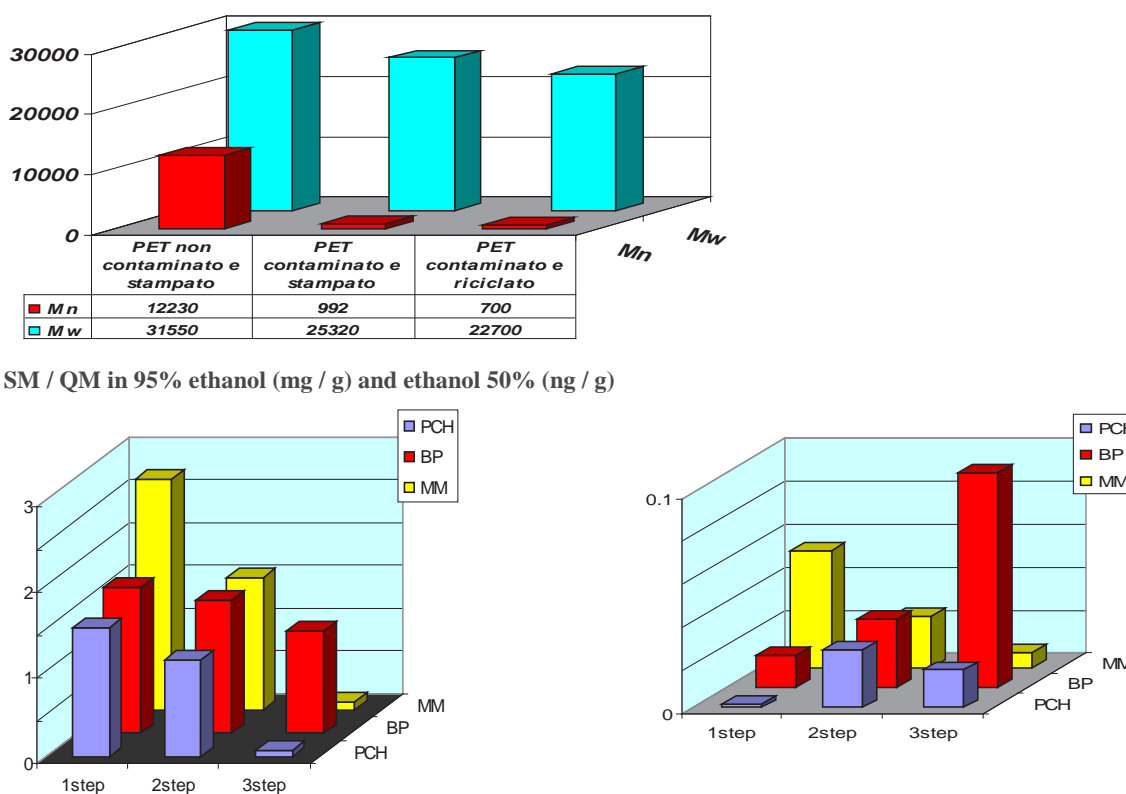


**Figure 3** Migration behavior of PET compared to the three contaminants and in the three stimulants and Changes the Parameters of Molecular structure.

coefficients are estimated, or approximated, with safe margins (and worst case assumptions) instead of precise determinations. The absolute worst-case migration model, verifies what would be the final concentration of the migrant in the food or simulants if all migrants are transferred from the polymer. If the specific migration limit is not exceeded, then no further model refinement or experimental determination is required.

- Isothermal rate predictions are approximate because factors such Sunlight, polymer length (number and weight average molecular weights), fluid substrates, etc., can affect migration rates.

- The modeling of migration is not restricted anymore to cases where analytical solutions of the Fick's equations can be calculated. Migration from polymeric multilayer structure can now be accurately simulated under real temperature variations, taking into account the diffusion coefficients of the migrant in the layers and its partition coefficient between the different layers.

- The absolute worst-case migration model, verifies what would be the final concentration of the migrant in the food or simulants if all migrants are transferred from the polymer. If the specific migration limit is not exceeded, then no further model refinement or experimental determination is required.

- The "FDA model" based on system assumptions leading to a simple solution valid for low migration times. FDA model was more accurate in most situations; it's important when dealing with highly irregular geometry of packaging or complex kinetics reactions but underestimated the results more frequently.

- The properties migration of contaminant in PET recycling subjected to depend not only on the amount of residual contaminants present in the polymer but also by the type and number of processing cycles performed. The differences in the migration behavior of the polymer compared to the three contaminants and in the three simulants were attributed to the different chemical and physical characteristics of migrants and the change in the migrant-polymer interactions and polymer-simulant following the execution of repeated cycles. The PET fact, after thermo-mechanical degradation phenomena induced by recycling, undergoes significant changes of the parameters of molecular structure (decrease of average molecular weight, increase in the oligomeric fraction and the number of carboxyl terminal) that modify the physical properties and migration. Therefore, for the purposes of prevention of contamination of foods, to evaluate the potential contaminant of a recycled PET is indispensable knowledge of the composition of the starting material, the structural parameters of the polymer and the operations carried out for the recycling of the polymer.

## RECOMMENDATIONS

In this study, the major research objectives were accomplished satisfactorily and additional information was generated. However, several questions remain unanswered due to limitation in the techniques and time constraints. Therefore the following studies are proposed to extend the current work. Further research is needed to develop migration model approaches and validate, both by fundamental research on diffusion in polymers and by well-considered applied research on real packaging and food or food simulants combinations.

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