Recycling of Multilayer and Barrier Coated PET Containers

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Abstract

This paper describes the process for removing barrier layers and coatings (oxygen and carbon dioxide) from polyethylene terephthalate (PET) substrates through a conventional mechanical bottle recycling system. Varied wash chemistry and barrier medium have been examined and the effect on residual multilayer material or coating has been evaluated. Wash chemistry was found to be the controlling factor in improving the external coating removal efficiency. Delamination through mechanical working was found to be the controlling mechanism for separating multilayer materials. The conclusion drawn from our experiments is that the PPG Bairocade coatings were removed most efficiently. Internal deposition techniques may contribute fewer residues to the RPET, however substantiating this is difficult.

Introduction

PET resins are widely used in the food packaging industry, in such products as bottles and films. PET bottles have a large market share in the carbonated soft drink, fruit juice and bottled water markets. These products have a shelf life of 8-12 weeks, over this period the gas permeability properties of PET are sufficient. However, alcoholic beverages are much more sensitive to oxygen and carbon dioxide diffusion either into or out of the bottle. When this sensitivity to migrating gases is combined with the need for a longer shelf life, it is necessary to improve on the gas permeability properties of PET.

To overcome the excessive gas ingress the bottle industry manufacturers have several options. The bottles can be made from a polyester with lower gas permeability, such as polyethylene naphthalate (PEN). In most cases these materials do not reduce the gas permeability sufficiently.

A widely adopted technique is to utilise a multilayer bottle construction. This method can be used to incorporate thin layers of polymers that have low gas permeability, into a sandwich structure where the barrier layer or layers are in the centre of the bottle wall encapsulated by PET on either side. [1]

Other methods that have been employed are to coat the external or internal walls of the bottle with materials that lower the overall ingress of gas into the beverage. These techniques use materials such as graphite carbon, silica and epoxy resins to achieve the effect. [2]

This research was aimed at understanding how the introduction of barrier coated or multilayer bottles into the recycling stream will effect the final PET quality. This will help in developing modifications to plant, processing conditions and wash chemistry whilst processing these materials in order to minimise the percentage of residual material in the RPET flake.

Materials and Methods

Materials

Bottles for these trials were supplied PPG bairocade (functional coatings), Nylon & Oxygen scavenger multilayer, nylon multilayer, Krones (surface SiO_x deposition), Sidel ACTIS (internal carbon deposition).

Trial Procedure

The multilayer and surface deposition materials were all run with a constant wash chemistry of 2% NaOH and 0.02% Oakite RC-7A detergent. The PPG Bairocade functional coated material was processed with a modified wash chemistry of 2.5% NaOH, 1.5% Butyl Carbitol and 0.1% Cetostearyl Alcohol (Teric).

Firstly, a hot caustic, whole bottle wash was used, followed by wet granulation, flake wash and a centrifuge separation prior to drying. Whole bottle wash NaOH concentration and temperature was 1% and 70-80^{o}C respectively. Flake wash residence time and temperature was 15min and 85-90^{o}C.

Barrier bottles were added into a pure PET bottle stream at a 10-weight percent addition rate. This rate was chosen to simulate 10% of the test bottles in the waste stream. The flake produced was blended during processing and prior to testing to ensure even dispersion of barrier material in the flake.

Laboratory testing of the processed flake involved oven-aging 1.5 kg of flake at 230°C for 1 hour and then visual identifying and removing the barrier layer for quantitative parts per million (ppm) determination.

Results and Discussion

For the Bairocade functional coatings, a series of trials were conducted to optimise the wash chemistry composition. This work improved the delamination and break-up from the PET substrate. This allowed very effective separation through sink float techniques. The high delamination and separation resulted in an approximate 99.96% removal efficiency, the best result in this series of trials, as can be seen in table 1.

Changes to the wash chemistry in the laboratory made little difference with separating the multilayer nylon materials shown in table 1. It was observed that the abrasion during granulation, washing and centrifuge treatment were the main drivers for delamination. Furthermore the delamination of the multilayer materials was higher than the removal rate. This was due to by the density of the middle layer Nylon 1.02-1.15 being too high to float in the sink float separation tank. It was found that an addition of a hydrophobic frothing agent into the sink float tank could greatly improve separation of such contaminants with densities higher than that of water.

The Krones barrier bottles were clear in colour. It was not possible to identify the presence of coating using standard contamination identification methods using oven aging, due to the thermal stability of the SiO_x layer. Since the SiO_x barrier layer could not be detected through conventional techniques, the processed flake was subjected to Infrared (IR) spectroscopy. The samples did not display any Si peaks in the IR spectrum. This could be due to the low levels of SiO_x initially present in the bottles.

The Sidel carbon coating was visible, however removal was minimal. These coatings are very thin 0.1 micron or 3 milligrams to treat a 500ml bottle and do not contribute high levels of contamination into the RPET. [3]

Conclusion

It was found that with PPG Bairocade functional coatings the wash chemistry played a critical role in lifting and separating the epoxy-amine coating from the substrate. With multilayer barrier bottles mechanical delamination was the variable controlling the removal efficiency. To promote further removal efficiency with these bottle additional centrifuges would need to be added. Otherwise the adhesion between the layers could be reduced through material modification to promote delamination during the recycling processing. The Krones SiO_x coated bottles exhibited no visual or colour change during the standards testing. As the coating was clear it is not possible to comment on its removal.

References

1. R. J. Judson, "Latest Economics of High-Speed, In Line Barrier Coating of PET Bottles", Nova-Pack Americas – 98, Ohio, USA.
2. www.grm.org.miller3.html, 4/02/999.
3. Extract from Sidel, ACTIS Amorphous Carbon Treatment on Internal Surface, Industrial Application presentation, 1999.

Key Word Index

PET Recycling, Barrier Coated Bottles, Multilayer Bottles.

Material	Wash Treatment	Residual Barrier Layer (ppm)	Removal Efficiency (%)
PPG Bairocade Functional Coating	2.5% NaOH, 1.5% Butyl Carbitol, 0.1% (Teric)	4-8	99.96
Multilayer Nylon & Scavenger	2% NaOH, 0.02% Oakite RC-7A	700	86
Nylon	2% NaOH, 0.02% Oakite RC-7A	546	94.54
Krones SiO_x Surface Deposition	2% NaOH, 0.02% Oakite RC-7A	None detected	Unknown
Sidel Internal Carbon Deposition	2% NaOH, 0.02% Oakite RC-7A	None detected	Unknown

Table 1. Residual barrier materials remaining in RPET flake after processing and removal efficiencies.