Development of functional barriers for the use of recycled materials in multilayer food packaging: the project BANUS

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BANUS, a project funded within European Union's Seventh Framework Programme for Research, finished last June after 24 months of research on the development of functional barriers allowing the use of non-authorized recycled materials in food packaging.

Thanks to the research carried out within this project, significant progress has been made on the optimisation of the functional-barrier assessment method and on the evaluation of the effectiveness of different materials on the market. Furthermore, the knowledge achieved opens the door to new working lines to develop new and completely safe products.

Regulation 10/2011 on food contact plastic materials, together with Regulation 282/2008 on food contact recycled plastics, set the stage for the use of post-consumer recycled plastic in food packaging applications. Their use is only permitted in two cases: if the recycled plastics come from an authorized recycling process, or if the recycled plastics are placed behind a functional barrier, with their corresponding specifications and restrictions. Moreover, there is currently no harmonised EU legislation on paper and board articles and materials for food contact applications beyond the general requirements laid out in Framework Regulation 1935/2004.

A functional barrier may be defined as a structure consisting of one or more layers, which either reduces the migration of authorised substances below their specific migration limit (SML) or reduces the migration of non-authorised substances into foods or food simulants to a non-detectable level.

The project has considered the substitution of a percentage of virgin material by recycled material (paper or plastic) in the selected structures in order to develop more eco-friendly food packaging structures.

Taking into account that the main objective of the project was to guarantee the suitability of the developed functional-barrier layers, it was necessary to check that, regardless of the quality of the recycled material used in the process, the functional barrier was able to prevent any migration of contaminants to food. The great advantage of the BANUS approach was to ensure highly efficient functional barriers to guarantee food safety when

using recycled materials (plastic and paper) coming from non-authorized recycling processes in food packaging structures.

The project BANUS has been centred on three different case studies, based on different materials, processed by different technologies and used for different applications, described as follows:

	Case Study 1: Semi-rigid multilayer plastic packaging	**Case Study 2**: Flexible multilayer multimaterial packaging	**Case Study 3**: Coated paperboard packaging
Current structure	Ext | **PP/EVOH/PP** | Int	Ext | **Paper/PET met/PE** | Int	Ext | **Paperboard/Coating** | Int
Current processing technology	Co-extrusion + Thermoforming	Lamination	Coating
Recycled material proposed	**Recycled PP**	**Recycled paper**	**Recycled paperboard**

The three cases are based on structures widely used in the market.

The aim was to replace a percentage of the virgin materials from the multilayer structure with a recycled one and then study the functional-barrier properties of the layers positioned between recycled layers and foodstuffs, taking into account that the effectiveness of the functional barrier depends on the chemical nature and thickness of the barrier and the conditions of use.

In the selection of the final structures, both migration and set-off were taken into account in order to guarantee food safety. Set-off is the phenomenon of the transfer of substances from outer layers of materials and articles to the food contact side, for example, during storage or transport. Functional barriers were applied to both sides of the recycled materials to avoid this set-off phenomenon.

The test method used in BANUS has been based on the 'Challenge Test' proposed in Regulation 282/2008 and by the U.S. Food and Drug Administration (FDA) to evaluate the recycling process efficiency. This test consists of contaminating virgin material with contaminants and then studying the behaviour of the whole structure regarding the migration of those contaminants. As specific compounds have been selected to cover a wide range of contaminants in a worst-case scenario, this approach guarantees that

functional barriers are initially evaluated in the worst situation, in contact with a highly contaminated material.

The project BANUS implies a great deal of innovation in the field of functional barriers because it has achieved technical advances in each one of the three cases studies proposed:

In every case, an improved and validated contamination methodology, including the selection of contaminants and their absorption in the virgin materials, has been developed and migration-testing methods have been optimized to perform a correct evaluation.

Case Study 1: Semi-rigid multilayer plastic packaging (PP/EVOH/PP)

In this case, the recycled material was PP and the proposed functional barrier was EVOH/PP. The intended use of this packaging taken into account for the evaluation of the functional-barrier performance is the contact with fatty liver (foie gras) at conditions of sterilization and subsequent long-term storage at room temperature or below.

Though quite interesting results were obtained after the evaluation of the selected structures, it was not proved that any of the structures tested in this case study act as functional barrier, as high levels of contaminants in the simulants after the exposure stage have been quantified, even after making some adjustments in the level of contamination to make it closer to the real conditions, taking always into account to cover all recycled grades that could be marketed nowadays.

These results could be due to a possible phenomenon of diffusion of the contaminants between contaminated and virgin layers in the molten state during the co-extrusion step due to high temperatures, the small size of the molecules of contaminants and the high quantity present in the contaminated material. If diffusion occurs in the processing step, results could not be representative of real migration kinetics as contaminants could be present in the food-contact layer before the migration exposure stage. The study of this diffusion phenomenon is a great advance regarding functional barrier considerations.

Taking into account the results obtained and considering that, even so, it is worthwhile to continue working in this issue due to the environmental and economic interest that the functional barrier concept generates among the industry, the work to find alternatives for the safe use of recycled PP in trays by introducing processing steps different from co-extrusion is being taken into account.

So, further trials and optimizations designated to achieve the functional barrier performance are needed beyond the end of the project. This further work will be based mainly on diffusion studies and on the modification of the processing technology. Thanks to both studies, alternative structures will be defined, thus fulfilling the requirements.

In terms of functionality, the differences obtained when testing prototypes manufactured from recycled PP show only minor differences that are considered as not relevant, taking into account that the values obtained with the new structure with recycled material are still appropriate parameters for their work and that there is still room for improvement of the prototypes tested by adjusting the processing parameters used for its manufacture.

Case Study 2: Flexible multilayer multimaterial packaging (Paper/PET met/PE)

In this case, the recycled material was paper, and the proposed functional barrier was PET met/PE. The intended use of this package, taken into account for the evaluation of the functional barrier performance, is the contact with preparations for soups in powder form (extracts, concentrates) at conditions of long-term storage at room temperature.

In the functional barrier evaluation, exaggerated case conditions were applied by means of four different spiking procedures. The results showed that PET met/PE could act as a functional barrier in real conditions.

The migration results of set-off samples showed that the set-off occurs rapidly, especially with the combination of heating in the lamination process. In the heating process, volatile compounds will most likely evaporate and will not be shown in the results. Migration after a long-time storage is mainly due to alkanes, mineral oils and some polyolefin oligomeric saturated hydrocarbons. Therefore, for full implementation, set-off phenomenon must be avoided by introducing an external coating in the structure. The best water-based coating selected for case study 3 was chosen also for the industrial scale-up of case study 2.

Regarding the scale-up, it is crucial to perform the industrial procedure in a one-step process without upwinding between the lamination of the metallised PET and the coating. Any deviation from the one-step procedure might result in potential set-off. Another aspect to be improved could be the excessive porosity and the worse optical quality of the recycled paper compared to the current standard virgin paper used for the application. The formulation of the water-based coating could be also adjusted to adapt it to the porosity of the paper. By controlling these aspects, the final product could give good results in the printing and coating stages, thus obtaining a product suitable for the clients' requirements. Adhesion tests showed that the PET/PE adhesion was excellent for both the pilot coated sample with recycled paper and the reference sample; other mechanical properties, such

as a tearing test, showed good overall values for both the reference sample and the pilot coated sample.

Case Study 3: Coated paperboard packaging (Paperboard/Coating)

In this case, the recycled material was paperboard, and the proposed functional barrier was a coating applied to this recycled paperboard in its internal layer (in contact with foodstuffs). Two intended uses of this packaging were taken into account for the evaluation of the functional barrier performance: the contact with fast food (high fat content) at conditions of short-term storage of hot foodstuffs and the contact with cereals at conditions of long-term storage at room temperature.

In the case of coatings, limited information was available about their functional barrier performance when positioned in the inner part of packaging, in direct contact with foodstuffs, so it was considered a priority to evaluate their performance as functional barriers.

A selective screening was performed in a laboratory setup to identify candidate coatings: The screening criteria to guide the selection of candidate coatings was the high penetration resistance to fats and oils, using the Tappi 454 test. In the screening of the dry coat weight determination and the Tappi test, about 120 applications of potential candidate barrier coatings were made. From the screening test, 9 candidate barrier coatings out of 120 were selected for further migration studies. Finally, one of the coatings was found to show the best performance as barrier and was, thus, selected for the scale-up, though the results obtained were not good enough to designate it as functional barrier for any substance in the evaluated conditions. However, a change of the environmental operational condition between the laboratory worst-case scenario for the migration tests to the real condition in the industrial coating and converting facility could still play a role for the coating as a functional barrier. Finally, the scale-up produced samples that showed properties as a barrier against migration of mineral oils from a secondary packaging of recycled fibre but they were not yet in compliance with the definition of a functional barrier and the barrier properties (OTR, WVTR). Additionally, the mechanical properties were not within the requirements. A further development of the coating recipe is required to make it persist and, thus, to get the proper barrier for a wider usage area.

The most promising barrier coating found on the market was shown not to be a functional barrier as tested on a board, per the requirements of the project. Thus, in the future, the search for a new development of a barrier coating is needed.

Concluding comments

Although none of the structures evaluated in BANUS was found to be 100 percent effective for their intended purposes at the end of the project, BANUS has finished providing relevant results that leave the partners in a better position to achieve the desired products in the near future. Now, the critical points of the current structures, the options that could properly work for certain applications and the developments needed to achieve 100 percent effective structures are known.

During BANUS development, major breakthroughs have been achieved on the existing methods for the evaluation of the functional-barrier performance of different structures, including the optimization of methodologies of contaminating materials and the evaluation of the migration of the substances used as contaminants. A considerable advance has also been made in knowledge regarding the characterization methods of the coatings evaluated and the behaviour of the contaminant substances inside the different structures evaluated.

It is also remarkable that BANUS has meant a major advance in the knowledge of the effectiveness of the coatings already existing in the market and the possible improvements applicable in order to get a coating that is able to satisfy the requirements of the project's participants. On the other hand, it has confirmed the existence of diffusion problems of the contaminant substances during the converting phase by co-extrusion, which limits the barrier effectiveness of the structures obtained by means of this processing technique.

From all this research and analysis of possible solutions, and given the major interest that this research line arises in the sector, BANUS opens the door to new works that allow the development of new fully efficient barriers that make possible the use of non-authorized recycled materials in food packaging.

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