Adhesives in Food Contact Articles
Current & Future European Regulation

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INTRODUCTION

Food contact articles and facilities, like cutlery, dishes, processing machines, containers and above all packaging [Fig. 1], are subject to special demands. This applies to both the processing of food and to its transport and warehousing. The term also includes materials and articles that are in contact with water intended for human consumption, but it does not cover fixed public or private water supply equipment. These articles are requested to not influence the food they contact, neither toxicologically ie, they should not cause a health threat for the end user, nor may they change the composition of the food or its properties like smell, taste and appearance. Therefore food contact articles shall not transfer their components into the foodstuff in unacceptable quantities. The first comprehensive regulatory activity for food contact materials and articles began in the late 1950s and early 1960s in the United States as well as in some European countries. Today, the regulation of food contact articles and especially packaging has become a global subject as the world's commercial channels have broadened in the last decades.

THE IMPORTANCE OF ADHESIVES IN FOOD CONTACT ARTICLES

Today’s variety of products would not be possible without modern adhesive technology. The same applies to food contact articles. There are applications like refrigerators, microwave ovens, kitchen furniture [Fig. 2], or corks for beverage bottles, but of special importance is the use of adhesives in the production of mass articles like packaging [Fig. 3]. Most of the food contact articles consist of different parts or components, which are often held together by adhesives. For the production of many of these food contact articles, today a vast number of adhesive systems are available, because there are so many different materials to bond. In addition, the demands on food contact articles during production process and use are very different.

While at the beginning of the last century adhesives were based on natural raw materials like protein, starch or cellulose, today many demands concerning different materials can only be met with the help of synthetic raw materials. In the first place, industrially processed adhesives [Fig. 4] are expected to bond the materials quickly and precisely and to allow trouble-free and cost-effective production, which is automated [Fig. 5] as far as possible. In addition they have to resist the stress of using the finished product. Today, ecological questions play an increasingly important role in the decision which adhesive system should be used. For instance it is increasingly demanded that adhesive systems can be processed in a way that produces as little waste or waste water in the production facilities as possible. Next to these demands also the question of the influence of adhesives on recycling/reuse – eg, returnable bottles – of the bonded products is becoming more and more important. Last but not least, the price should also not be underestimated for choosing an adhesive.

The evolution in materials and processes led to an intensive development of adhesives systems, like dispersions, hot melts or reactive systems in recent decades, so that today there are tailor-made
adhesives for all kinds of applications and performance demands. The adhesives that are used for the production of food contact articles are requested to fulfil all technical requirements as well as the requirements of national or European directives and regulations for materials and articles intended to come into contact with foodstuffs.

NATIONAL REGULATIONS OF FOOD CONTACT MATERIALS AND ARTICLES

In the past, food-safety demands -- that is, the protection of human health from any potentially harmful substance that might migrate from a food contact material or article into foodstuff -- have been described in national laws. In Germany, since 1958 there have been "Recommendations on the health assessment of plastics and other high polymers" like for instance paper and rubber. They are elaborated within the framework of the German Foods and Other Commodities Act (LMBG). The "Plastics Recommendations" were initially the responsibility of the Federal Health Office (BGA). Since November 1, 2002 the responsibility lies with the Federal Agency for Risk Assessment (BfR), which is advised by its Plastics Committee. Some of the "Plastics Recommendations" apply to adhesives [Fig. 6].

EUROPEAN REGULATIONS

On the European level two authorities deal with questions of food safety. One is the Council of Europe and the other is the European Union. While the decisions of the European Union are obligatory for national legislations, those of the Council of Europe are only a recommendation.

THE COUNCIL OF EUROPE

The Council of Europe is a political organisation of 43 European countries. In the “Partial Agreement in the social and public health field” 18 countries joined together to deal with health protection. The resolutions are prepared in the Committee of Experts on Materials Coming into Contact with Food and other groups related to specific subjects. Later these resolutions are accepted by the Public Health Committee (Steering Committee) and adopted by the Committee of Ministers. Even though there is no formal agreement between the European Union and the Council of Europe concerning food contact materials, there are important and numerous contacts between the services of the Health & Consumer Protection Directorate-General and the Council of Europe in this field. The Council of Europe has prepared a number of documents, which are used as basics for many national regulations in Europe [Fig. 7].

THE EUROPEAN UNI0N

The steady deepening of the integration of the European countries into the European Community in recent years has led to a beginning harmonisation in this field. A first Legislation on materials and articles intended to come into contact with foodstuffs was adopted in 1976. It was replaced in 1989. This legislation was a basis for a harmonisation of the European regulations regarding materials and articles intended to come into contact with foodstuffs.

FRAMEWORK REGULATIONS

The bases for the regulations of the European Union are the framework directives or regulations. The framework directives or regulation for food contact articles applies to all materials and articles intended to come into contact with foodstuff and set some general requirements. A first framework directive on materials and articles intended to come into contact with foodstuffs was adopted in 1976. It was replaced in 1989 by the Framework Directive 89/109/EEC. This basic law for food contact articles was laid down in the Council Directive of 21 December 1988, consolidating the laws of the member states relating to materials and articles intended to come into contact with foodstuffs.
FRAMEWORK REGULATION (EC)1935/2004

Due to technological development it was necessary to adjust this framework directive in 2004. Hence on 27 October 2004 the new Framework Regulation (EC)1935/2004 was enacted and published in the European Official Journal on 13 November 2004. In contrast to a directive, which has to be converted after publication by the European Union within a certain period by the individual member states into national law, a regulation is automatically converted into the national law of all EU countries twenty days after publication. The regulation (EC)1935/2004 set some general requirements that must be met by all food contact materials: All materials and articles must be manufactured according to Good Manufacturing Practice (GMP) so that they do not transfer their constituents to foodstuffs in quantities which could endanger human health or "bring an unacceptable change in the composition of the foodstuff or a deterioration in the organoleptic properties". In article 3 the following general requirements are mentioned.

"Materials and articles, including active and intelligent materials and articles, shall be manufactured in compliance with good manufacturing practice so that, under normal or foreseeable conditions of use, they do not transfer their constituents to food in quantities which could:

(a) endanger human health; or
(b) bring about an unacceptable change in the composition of the food; or
(c) bring about a deterioration in the organoleptic characteristics thereof."

Detailed explanations about how these principal demands should be transformed are described in specific regulations within the framework regulation, as they have already been described in the previous framework directives. In this new framework regulation also some new articles and groups of substances were included for which specific regulations should be prepared. In Annex 1 seventeen groups of materials or articles are listed for which special directives or regulations are planned [Fig. 8].

SPECIFIC DIRECTIVES OR REGULATIONS

Annex 2 in the Framework Directive 89/109/EEC lists 15 materials and articles for which regulations should be prepared to fulfil the demands of the Framework Directive. In these specific directives detailed explanations about the principal demands of the framework directive are described. However, specific directives have not been adopted yet for all groups of materials and articles. Currently specific directives exist for three groups of materials and articles: ceramics (84/500/EEC), regenerated cellulose (93/10/EEC) and plastic (2002/72/EC). Three groups of substances are regulated individually in specific directives, i.e. vinyl chloride monomer in plastic (78/142/EEC), nitrosamines in rubber teats and soothers (93/11/EEC) and BADGE, BFDGE and NOGE in plastics and coatings (2002/16/EC).

The most-mentioned specific directive in connection with adhesives is the plastic directive (2002/72/EC). An update of this directive is currently discussed. The draft is called the "Super Regulation".

"SUPER REGULATION"

The driving idea in writing the “Super Regulation” (former “Super Directive”) was to collect in one text the rules set out in all the directives related only to plastic materials and articles i.e.: Commission Directive 2002/72/EC and its amendments; consolidation of 82/711/EEC and its two amendments, 93/8/EEC and 97/48/EC; 85/572/EEC; and the three vinyl chloride monomer directives, 78/142/EEC, 80/766/EEC and 81/432/EEC. This should be considered as an attempt to prepare a future Commission Directive on all types of plastics. As usual, the regulation or directive contains only general principles of the technical rules such as modelling, functional barrier etc. Details and guidance will be introduced at CEN level or in the Practical Guide.
To give an idea of the repartition of the rules, some examples or references to existing documents are given in the draft of the "Super Regulation", such as the extension of the rules to multilayers composed of different materials and the introduction of the concept of "functional barrier". It has to be noted that the layer acting as "functional barrier" may be either authorised by the directive or regulations and listed together with the conditions of its validity, or used without any authorisation by the manufacturer provided that the written declaration of compliance mentions the substances not subject to an authorisation procedure.

The draft of the so-called "Super Regulation" intends also to include the migration contributions of all layers of a multilayer construction, such as adhesives or printing inks used in multilayer constructions, but without mentioning a specific regulation. A multilayer plastic material shall comply with the following rules: Any layer shall be manufactured from substances mentioned in a community list subject to their restrictions. In absence of a community list, substances which appear in a national list from a European country may be used provided they comply with article 3 of the Regulation (EC)1935/2004. In the absence of these lists from a European country, only substances in amounts which, in the practical use of the final materials or articles, can be proved to comply with article 3 of the Regulation (EC)1935/2004 may be used. These substances are hereinafter referred to as "Listed food contact substances". The finished articles shall comply with the overall migration limit (OML), the specific migration limit (SML), and the other specific restrictions of the substances contained in the different layers; each layer maintains its own specifications.

A "Functional Barrier" is a barrier consisting of one or more layers which ensures that migration of components does not exceed their SML, and which reduces the migration of these components, their reaction products, and impurities into food to a "non detectable" level when subjected to a validated test method. For the purpose of control, "non detectable" means that a substance cannot be detected at a concentration appropriate for the nature of the substance, but in no case exceeding 0.01 mg/kg. This detection limit applies to groups of structurally related compounds, e.g. isomers.

MIGRATION

The transfer of components from the food contact materials into food is called migration. To ensure the protection of the health of the consumer and to avoid alteration of the foodstuff, two types of migration limits have been established in European law. For the first time these numerical values appeared in the Commission Directive (Specific Regulation) of 23 February 1990 relating to plastic materials and articles intended to come into contact with foodstuffs:

- an overall migration limit (OML) of 60mg (of substances)/kg (of foodstuff or food simulants) that applies to all substances that can migrate from the food contact material into the foodstuff and

- a specific migration limit (SML) which applies to individual authorised substances and is fixed on the basis of the toxicological evaluation of the substance. The SML is generally established according to acceptable daily intake (ADI) or tolerable daily intake (TDI). These values formerly were set by the Scientific Committee on Food (SCF) and today are set by the European Food Safety Agency (EFSA). To set the limit, it is assumed that every day throughout his/her lifetime, a person of 60kg eats 1kg of food packed in plastics containing the relevant substance at the maximum permitted quantity.

The current approach for the authorisation and control of substances used in food contact materials is considered to be cautious in relation to the estimation of potential exposure of the consumer to these substances. Approaches are being discussed which take into better consideration the actual exposure of the consumer to food contact materials in risk assessment.
THE CURRENT EUROPEAN REGULATION OF FOOD CONTACT ADHESIVES

Adhesives are not yet treated as an individual regulated group, so that the European regulation of adhesives still is not harmonised. As is the case with all other materials, adhesives for food contact articles have to fulfil the requirements of the framework directive (EC)1935/2004. Adhesives are mentioned in Annex 1 of the framework directive (EC)1935/2004 for the first time, along with sixteen other groups of materials or articles listed for which special directives or regulations are planned. However, no specific regulation for adhesives has yet been published.

The draft of the so-called "Super Regulation" intends to include the migration contributions of the adhesives used in multilayer constructions, but without mentioning a specific regulation.

Since there are no regulated adhesive components, only conclusions of analogy can be drawn from the information in the positive lists of other material groups. This is easy when adhesives are based on synthetic polymers. Most of the monomers and other raw materials needed for the production of adhesive polymers are listed and evaluated in the plastic directive (2002/72/EC). To some of these substances specific migration values and/or quantity limits have been allocated. For substances not mentioned in a community list, evaluations which appear in a national list from a European country, like the German BfR regulations, may be used provided they comply with article 3 of the Regulation (EC)1935/2004. In the absence of these lists from a European country, only substances in amounts which, in the practical use of the final materials or articles, can be proved to comply with article 3 of the Regulation (EC)1935/2004 may be used. The finished articles shall comply with the overall migration limit (OML), the specific migration limit (SML) and the other specific restrictions of the substances contained in the different layers; each layer maintains its own specifications.

FUTURE SPECIFIC REGULATIONS FOR ADHESIVES

In discussions between the European Commission and the European Adhesive Association (FEICA) several different approaches were prepared. Regardless of which of these approaches will be incorporated into the regulations, in the future the European idea, which is characterised by consumer and environmental protection, will lead to a clear expansion of the necessary documentations and surveys of adhesives and adhesive raw materials used for food contact materials. For raw materials especially it will be necessary to disclose many ingredients, so that processors can execute corresponding surveys.

This is mentioned in article 16 “Declaration of Compliance” of the framework regulation (EC)1935/2004, which says: The specific measures referred to in article 5 shall require that materials and articles covered by those measures be accompanied by a written declaration stating that they comply with the rules applicable to them. Appropriate documentation shall be available to demonstrate such compliance. That documentation shall be available to the competent authorities on demand. Therefore, the adhesive industry, too, will have to give much more information to its customers so that customers can ensure the compliance of their articles with the European food contact material regulation.

The written compliance declaration shall permit an easy identification of the materials or articles for which it is issued and shall be reviewed periodically. It shall contain the following information: Identity of material or article; its range of application; and the confirmation that the material or article complies with the requirements of the European Directives and, when appropriate, of national law. When a functional barrier is used in a multilayer material, the following additional information shall be provided: The identity of the substances of the functional barrier; the date of latest use of the material or article; and the maximum heat treatment (temperature and time) for the article. At each stage of manufacture, processing, and distribution an appropriate technical documentation able to demonstrate the compliance of the material or article or substances with the relevant provisions shall be available. This documentation, hereinafter called "Supporting Documents", shall contain the description and the results of the analysis carried out to demonstrate the compliance of the material and article, and in particular the compliance with quantitative restrictions in the use of the substances such as OML, SML etc, plus

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the requirements of the layer(s) constituting a functional barrier, and the requirements set out in article 2 of Directive 89/109/EEC related to the substances migrating in detectable amounts and which are not listed in positive lists.

For a specific future adhesives regulation, from today's point of view the following possibilities can be imagined:

1. Inclusion in the positive-list system of the Plastic Directive 2002/72/EC or in the succession regulation of the Plastic Directive, called "Super Regulation", which is currently discussed.

2. A specific directive for adhesives with a special positive list for adhesives.

3. The industry works out its own concept which has to be acceptable by the enforcement, and which ensures the compliance of the products regarding article 3 of the Framework Regulation (EC)1935/2004, namely the general demands (not to endanger human health).

**POSITIVE LIST SYSTEM**

The positive list system means that all raw materials and additives used for production of an adhesive have to be listed in an EU guideline dealing with adhesives for food contact materials. This procedure offers the advantage that finally all substances used have to be evaluated by the European Food Safety Agency (EFSA) and officially authorised. If necessary, limits have to be set in the European adhesive regulation. Some of the raw materials and additives used for adhesives are also used for plastics and so are already evaluated. However it has to be considered that short-chain polymers, such as are used in those adhesives whose viscosity is controlled by the length of the polymer chain, are now defined as polymer additives and are not regulated by the list of monomers and raw materials, but must be authorised specifically. Also, most raw materials used for the production of adhesive formulations are not used in the already regulated production of plastic materials. These, as well as the short-chain versions of approved plastics, have to pass through an approval procedure before they are included in the list. For this, migration and toxicological studies based on the extent of migration are necessary. Depending on the knowledge of the raw materials these studies can require a lot of effort and be expensive. To get all necessary data for migration tests approximately 3,000–50,000 € and for the toxicological dossier approximately 50,000 – 500,000 € (range based on migration levels) are estimated. In addition, reactions and by-products have to be measured and taken into account. Depending on their amount and composition, migration toxicological data can also become necessary for them.

It is obvious that an inclusion in the positive list system of all substances used in adhesives would be an enormously time-consuming and expensive process. To illustrate this amount of time one should look at the development of the positive list for plastic monomers. When the Plastic Directive 90/128/EEC was established, monomers and other raw materials that were used before 1990 for the production of plastics for food contact, but which had not yet been evaluated by the Scientific Committee for Food (SCF), were included in a B list of temporarily-usable substances pending a decision on inclusion in the positive list (Section A). This B list will presumably be cancelled by 31 December 2007, meaning that it will have taken 17 years until the last ‘old’ monomers and other starting substances have been evaluated and included in the A list of the approved monomers, or deleted.

It is assumed that many of the raw materials used for adhesives (and especially the additives) will get specific migration limits based on toxicological evaluation. Compliance with migration limits has to be controlled on the finished food contact material. As adhesives consist of many components, migration tests by the old procedure would be very expensive and time-consuming. For many or for most of the components today there are no, or at least no validated, analytical measurement methods.

In contrast to the materials of which food contact articles principally consist, adhesives are usually only a minor component of food contact articles like packages. The tonnage of adhesives is but a minor

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percentage of the tonnage of, for example, paperboard and plastic. Thus the pressure for adhesive raw material producers to invest time and money in the approval of raw materials is limited, and because of this the adhesive industry will lose a substantial number of raw materials. The consequence could be that some of today’s adhesive formulations will not conform to the food regulations anymore and therefore be forbidden for these applications.

INDUSTRY’S OWN CONCEPT – PRACTICAL GUIDE

The alternative to the positive list system would be to prepare a praxis-orientated testing concept to guarantee the safe use of adhesives in food packaging. This test could be developed by the adhesives industry, including small and medium-sized enterprises, cost-efficiently and without compromise to consumer protection requirements and thus to their own product image. Such a concept finally should be given to the European authorities to be written in a legal format. Most likely, the general principles would be embedded into a European regulation. The necessary details would be laid down in the so-called Practical Guide for food contact materials of the Commission.

Such a practical guide takes into account the relatively small quantity of adhesive compared to the material that forms the food-contact article itself, eg, slim glue line of folding boxes [Fig. 9]. The practical guide is also based on the fact that adhesives are usually between two other materials: rarely is there direct contact of adhesive and foodstuff. Hence the components of the adhesives cannot migrate into the foodstuff directly. Evaluation of the migration of adhesive components into foodstuff could be based on the principle of the so-called ‘Functional Barrier’ (FB) [Fig. 10], which will presumably be considered within the regimentation of the “Super Regulation”. The basic idea behind the FB principle is that the food contacting layer(s) (of a multilayer) may act as a barrier for migration of adhesive components. The performance of a functional barrier depends on the properties of the food contacting layer and the migrating substance. As a consequence, substances that are immobilised in the adhesive or that cannot migrate into the food because of the functional barrier properties of the interior layers under the presumed use conditions, can be omitted from this concept of approval. Then a toxicological evaluation would only be necessary for those substances that can migrate in significant or large amounts.

MATHEMATICAL MODELLING

Tests of migration and migration kinetic tests for barrier materials are very time-consuming and expensive. Usually, the adhesive exists in a very thin layer, and the amount of components able to migrate is small, so that such sophisticated migration tests often are not necessary. The test concept hence should be a fast, cost-effective, and relatively easy method of determining compliance. This should be achieved by a combination of extraction tests to determine the potential of components able to migrate, mathematical modelling, and a few verifying migration tests of the finished food contact materials. Conclusions should be drawn from a tested system with defined layers and layer thicknesses about the compliance of other systems with the same adhesive, without causing the need for new tests.

The use of mathematical modelling for prediction of migration, which can reduce the amount of tests to be undertaken, has been recently introduced into the legislation. Practical examples for the application of this new concept are described in the Practical Guide.

Adhesives are considerably more complex systems than mono plastics, so that the modelling methods used for plastics until now cannot be simply transferred to the testing of adhesives. Because of this there is yet no scientifically stable basis for simplified testing of adhesives. Different research institutes have prepared the ground in this field of research and from these preparations such a concept can be developed. The mathematical modelling of migration from mono plastics and the estimation of diffusion coefficients as proof (Piringer model) [Fig. 11] is validated for many plastics [Fig. 12] and has been included as a possibility to test the compliance in the European Plastic Directive 2002/72/EC (article 8/4) and Practical Guide. The model has been published among others as a guide in the Practical Guide for Food Contact Materials of the European Commission and as a CEN Report. It has been extended to

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the applicability on multilayer materials in connection with a recently finished research project.

**SUMMARY**

Without multilayer food contact materials modern life would not be possible. While there are already European regulations for plastics, for other components like printing inks or adhesives a preparation is intensively discussed. The establishment of a positive-list system for those components will require very much time and money and this surely cannot be accomplished for all formulation components used today. Thus in the future a number of formulations may no longer be legal for food contact materials. A positive-list system absolutely requires an individual compliance check with specific migration limits. It has to be considered that conventional migration test processes are time-consuming and costly and that there are no analytical methods for many components yet.

As a consequence the only possible and useful way is a specific testing concept by a combination of extraction and migration tests and mathematical modelling, that guarantees the safe use of these components in multilayer food contact materials, and which can be utilised cost-efficiently by the industry, especially by small and medium-sized enterprises.

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Fig. 1 Food Contact Articles

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Fig. 2 Kitchen Furniture

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Fig. 4 Industrially Processed Adhesives
Fig. 5 Fully Automatic Production Process
VII. Polypropylene
X. Polyamides
XIV. Plastics Dispersions
XVI. Polyvinyl Ethers
XX. Polyisobutylene, Isobutylene Copolymers and Mixtures of Polyisobutylene with other Polymers
XXII. Polymers Based on Esters of Acrylic and Methacrylic Acids, their Copolymers, and Mixtures of these with other Polymers
XXV. Hard Paraffins, Microcrystalline Waxes and Mixtures of these with Waxes, Resins and Plastics
XXVIII. Cross-Linked Polyurethanes as Adhesive Layers for Food Packaging Materials

Fig. 6 BfR Recommendations Used for Adhesives
Resolution AP (89) 1 on the use of colourants in plastic materials coming into contact with food
Resolution AP (92) 2 on control of aids to polymerisation for plastic materials and articles
Resolution AP (96) 5 on surface coatings intended to come into contact with foodstuffs
Resolution AP (97) 1 on ion exchange and adsorbent resins used in the processing of foodstuffs
(superseding Resolution AP (89) 2)
Resolution AP (99) 3 on silicones used for food contact applications
Resolution AP(2002) 1 on paper and board materials and articles intended to come into contact
with foodstuffs
Framework Resolution AP (2004) 1 on coatings intended to come into contact with foodstuffs
Resolution AP (2004) 2 on cork stoppers and other cork materials and articles intended to come
into contact
with foodstuffs
Resolution AP (2004) 3 on ion exchange and adsorbant resins used in the processing of foodstuffs
Resolution AP (2004) 4 on rubber products intended to come into contact with foodstuffs
Resolution AP (2004) 5 on silicones used for food contact applications

Present activity programme of the Committee of experts:
List of substances on paper and board products intended to come into contact with foodstuffs
(concerning Resolution AP (2002) 1);
Draft guidelines on the safety evaluation of food contact paper and board substances;
Draft resolution on printing inks, primers, coloured lacquers and overprint varnishes applied to the
non-food contact surface of food packaging and articles intended to come into contact with foodstuffs;
Guidelines on lead leaching from glass tableware into foodstuffs;
Guidelines on tissue paper kitchen towels;

Fig. 7 Council of Europe Recommendations
Regulation of the European Parliament and of the Council on materials intended to come into contact with food: (EC)1935/2004

Annex 1: List of groups of materials and articles which may be covered by specific measures

<table>
<thead>
<tr>
<th>Active and intelligent materials and articles</th>
<th>Plastics</th>
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<tbody>
<tr>
<td>Adhesives</td>
<td>Printing ink</td>
</tr>
<tr>
<td>Ceramics</td>
<td>Regenerated cellulose</td>
</tr>
<tr>
<td>Cork</td>
<td>Silicones</td>
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<tr>
<td>Rubbers</td>
<td>Textiles</td>
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<td>Glass</td>
<td>Varnishes and coatings</td>
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<tr>
<td>Ion-exchange resins</td>
<td>Waxes</td>
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<tr>
<td>Metals and alloys</td>
<td>Wood</td>
</tr>
<tr>
<td>Paper and board</td>
<td></td>
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</tbody>
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Fig. 8 Specific Regulations
Fig. 9 Adhesive Application

Courtesy of Adhesives.org
Fig. 10  Migration in Multilayer Foodstuff / Simulant

Adhesives  Functional Barrier  Foodstuff / Simulant

$D_A$  $K_{A,FB}$  $D_{FB}$  $K_{FB,F}$  $\bullet$ Migration

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\[ -\left| \frac{\partial c}{\partial t} \right|_{\text{total}} = -D \left( \frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} + \frac{\partial^2 c}{\partial z^2} \right) + \left( v_x \frac{\partial c}{\partial x} + v_y \frac{\partial c}{\partial y} + v_z \frac{\partial c}{\partial z} \right) + k_r c^n + k_e \sigma \]

\[
\frac{m_{L,t}}{A} = c_{p,0} \rho_p d_p \left( \frac{\alpha}{1+\alpha} \right) \left[ 1 - \sum_{n=1}^{\infty} \frac{2\alpha(1+\alpha)}{1+\alpha + \alpha^2 q_n^2} \exp \left( -D_p t \frac{q_n^2}{d_p^2} \right) \right]
\]

\[
\alpha = \frac{V_L}{V_P} \frac{K_{P,L}}{K_{P,L}}
\]
Fig. 12  Migration Modelling (HDPE Single Layer)

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