Volume 4 • Number 1 • January 2010
ISSN: 1557-7244

Journal of

APPLIED PACKAGING RESEARCH

Aim and Scope

The *Journal of Applied Packaging Research* is an international forum for the dissemination of research papers, review articles, tutorials and news about innovative or emerging technologies for the packaging industry. The journal is targeted towards the broad packaging community including packaging scientists and engineers in industry or academic research and development, food scientists and technologists, materials scientists, mechanical engineers, industrial and systems engineers, toxicologists, analytical chemists, environmental scientists, regulatory officers, and other professionals who are concerned with advances in the development and applications of packaging.

Editor

Jay Singh
Industrial Technology
Cal Poly State University
San Luis Obispo, CA 93407, USA
Phone: 805-756-2129, Email: singhjapr@gmail.edu

Editorial Advisory Board

JOURNAL OF APPLIED PACKAGING RESEARCH—Published quarterly—January, April, July and October by DEStech Publications, Inc., 439 North Duke Street, Lancaster, PA 17602-4967.

This journal is recommended by The National Institute of Packaging Handling and Logistics Engineers (www.niphle.org).

Indexed by Chemical Abstracts Service.

Indexed and abstracted by Pira International.

Subscriptions: Annual $319 (Print), $319 (Electronic) and $344 (Print and Electronic). Single copy price $95.00. Foreign subscriptions add $45 per year for postage.

(ISSN 1557-7244)

DES*tech* Publications, Inc.
439 North Duke Street, Lancaster, PA 17602-4967, U.S.A.

CONTENTS

An Evaluation Method for Quantifying Sustainability of Protective Cushioning

LIJIANG HUO[1,*] and KATSUHIKO SAITO[2]
[1]Department of Printing and Packaging Engineering, School of Textile and Light Industry, Dalian Polytechnic University, Dalian 116034, China
[2]Transport Packaging Laboratory, Graduate School of Maritime Sciences, Kobe University, Japan

ABSTRACT: Protective cushioning reduces damage for contained contents throughout product supply chains. The packaging/product system not only creates value but also brings positive benefits. Promoting more sustainable protective cushioning is challenging. The research aims at comparing sustainability of protective cushioning within the product supply chain by a sustainable assessment (SA) method with transparent analysis. The SA measures the performance of the protective cushioning with regard to social, economic and environmental aspects and produces quantitative results (in monetary units), and then indicates the sustainability of the protective cushioning using a sustainability indicator (SI). In the SA, a newly developed indicator representing social performance of the protective cushioning, named real value added (RVA), is based upon quality function deployment (QFD) method; life-cycle impact assessment method based on endpoint modeling (LIME) is mainly adopted for measuring environmental impacts. Two new protective cushioning designs for a CD receiver (for car use only) were assessed in this study. The optimum selection for the cases is identified by calculating the SI, and the potential improvements are indicated by quantifying the RVA, economic benefits and environmental impacts of the protective cushioning. The proposed SA approach can be used to assist in identifying practical designs and guiding stakeholders in the packaging/product system.

1.0 INTRODUCTION

OVER the last few years, some leading organizations and companies have worked to create evaluation tools to support decision-making for promoting more sustainable packaging, such as the Packaging Scorecard launched by Wal-Mart [1], the PIQET developed by Sustainable Packaging Alliance [2], to name a few. These tools apply performance parameters in their models to evaluate environmental impacts

*Author to whom correspondence should be addressed. Email: huolijiang@gmail.com

1557-7244/10/01 001-14

and economical benefits of packaging. But few initiatives incorporate parameters reflecting social value, or social functionality, of the packaging in their approach. Actually, social performance of the packaging materials, products and systems play an important role in advancing sustainability throughout product supply chain. It has to be indentified and presented in an evaluation tool for enhancing sustainability of the packaging [3]. In view of this, we established the sustainable assessment (SA) and tried to assess the sustainability attributes of protective cushioning by the SA approach. In this study, we focused on (1) identifying the sustainability attributes of the protective cushioning involving three pillars of sustainability; (2) characterizing input-output flows of the protective cushioning in its lift cycle by viewpoint of the product supply chain; and (3) quantitatively measuring the sustainability attributes and presenting sustainability of the protective cushioning using a single sustainability indicator (SI).

SA APPROACH

In this study, sustainable protective cushioning is considered within the context of product supply chain. Based upon life-cycle thinking [4], basic input-output logic regarding the protective cushioning itself is set up for describing the value and impacts brought by the protective cushioning, as diagramed in Figure 1. The processes contained by the dotted line in Figure 1, is considered as the system boundary in this study. The system under study commenced with raw material collection and ended with protective cushioning disposal. Inputs and outputs of the system throughout the system boundary are also indicated in Figure 1. Total inputs are regarded as physical consumptions and intangible consumptions for obtaining the protective cushioning. Total outputs are considered as positive outputs (creating value added for society) and negative outputs (resulting in environmental damage) [5].

The total inputs and outputs of the protective cushioning are identified and quantified on the basis of a monetary unit for communicating with stakeholders in more direct way. Further, the SI is expressed through an equation of more-is-better elements as opposed to less-is-better elements [6]. The total inputs are presented by direct costs of consumed inputs (DCC), as expressed in Equation (1). The value added (VA) produced by the positive outputs is presented by economic gains of the protective cushioning in the product supply chain related to market, as

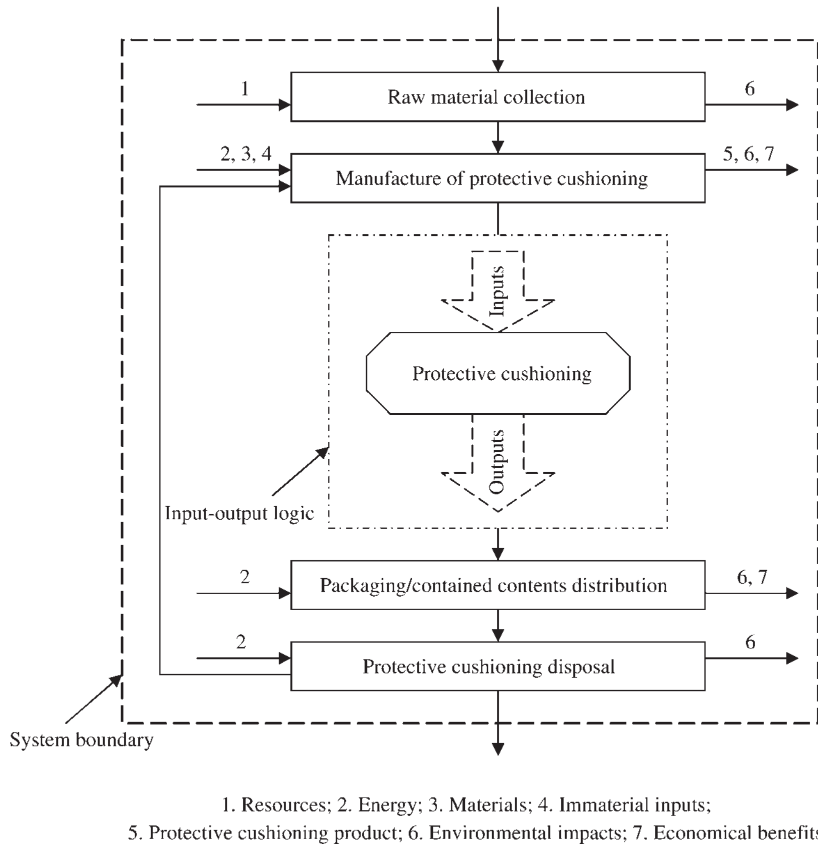


Figure 1. *Input-output flows of protective cushioning within lift cycle and system boundary in this study.*

expressed in Equation (2). The negative outputs resulting in environmental damage are considered to be hidden costs of consumed inputs (HCC). In this study, the HCC is calculated by life-cycle impact assessment method based on endpoint modeling (LIME) [7,8]. Eco-indicator' 95 and Ecopoint models are also used for validating the results of environmental damage [7].

$$DCC = C_{materials} + C_{resource} + C_{energy} + C_{other} \quad (1)$$

where

$C_{materials}$ = costs of materials needed during the protective cushioning production;

$C_{resources}$ = costs of consumed natural resources during the protective cushioning production;
C_{energy} = costs of consumed energy during the protective cushioning production;
C_{other} = costs of equipment depreciation, maintenance, salary and taxes related to the protective cushioning design and production.

$$VA = (SV_{contained\ contents} - C_{contained\ contents}) \times \frac{AC_{protective\ cushioning}}{C_{contained\ elements}} \quad (2)$$

where

$SV_{contained\ contents}$ = sale value of contained contents using the protective cushioning;
$C_{contained\ contents}$ = costs of contained contents using the protective cushioning;
$AC_{protective\ cushioning}$ = allowed costs of the protective cushioning, it depends on local regulations.

The VA is created by technical performance of the protective cushioning within the framework of related social laws and regulations. The VA calculation works on the premise that the technical indicators of each protective cushioning meet requirements of customer well. But differences in the social functionality of the available protective cushioning still remain. In this study, an indicator named real value added (RVA) is developed based on quality function deployment (QFD) method [9] for further distinguishing the differences in the evaluation. Quantitatively, the RVA presents the social performance of the protective cushioning.

According to sustainable packaging principles [10,11], the social functionality of the protective cushioning incorporates safety, convenience and health, and the related elements can be compared by a QFD matrix, as shown in Table 1.

Customer requirements and their importance (demand-side parameters) are derived from market survey. Relationship values between the customer requirements and quality characteristics (supply-side parameters) are commonly chosen from among 0, 1, 3, and 9. Relative importance of the quality characteristics is then calculated by Equation (3) [9].

Table 1. *QFD Matrix for Comparing Social Functionality of Protective Cushioning.*

Customer Requirements	Importance	Quality characteristics										
		Material Type	**Weight (kg)**	**Fragility (G)**	**Packing**	**Water Absorption**	**Speckiness**	**Causticity**	**Mildew Resistance**	**Weather Resisting**	**Flammability**	**Disposal**
Safety	9	3		9		3		1		1	1	
Convenience	3	3	1		9	1		1	1	1	1	3
Health	1	9					1	3	3			9
Relative importance (%)		**18.0**	**1.2**	**32.4**	**10.8**	**12.0**	**0.4**	**6.0**	**2.4**	**4.8**	**4.8**	**7.2**

Notes: Relationship: 9 = strong relation; 3 = normal relation; 1 = weak relation. Importance: 9 = most; 3 = normal; 1 = less.

$$w_j = \frac{\sum_i (p_i \times \alpha_{ij})}{\sum_j \sum_i (p_i \times \alpha_{ij})} \tag{3}$$

where

p = importance of customer requirements;
α = relationship value in a QFD matrix;
w = relative importance of quality characteristics;
i = customer requirements ($i = 1, \ldots, I$);
j = quality characteristics ($j = 1, \ldots, J$).

In the evaluation, actual data for quality characteristics are modified based on an improvement direction, given by Equation (4) [9]. This modification makes it possible to equalize the desirable direction of the quality characteristics. Then, improvement ratios of the modified actual data are calculated by normalization on the basis of the maximum, given by Equation (5) [9]. This normalization method makes it possible to evaluate new functions. Finally, the improvement ratios are multiplied by the relative importance of the quality characteristics derived from the QFD matrix and the social functionality value of the protective cushioning is calculated by their sum, given by Equation (6) [9].

$$\begin{aligned}&\text{If higher is desirable, } MF_j^n = F_j^n;\\ &\text{if lower is desirable, } MF_j^n = \frac{1}{F_j^n}\end{aligned} \tag{4}$$

$$RF_j^n = \frac{MF_j^n}{\underset{n=1,\ldots,N}{MAX}\{MF_j^n\}} \tag{5}$$

$$V^n = \sum_j (w_j \times RF_j^n) \tag{6}$$

where

F = data of quality characteristics;
MF = revised ratio of quality characteristics based on improvement direction;
RF = ratio of quality characteristics;
V = social functionality value of the protective cushioning;
n = protective cushioning products ($n = 1, \ldots, N$).

A modified coefficient *K* is further set up in this study, given by Equation (7). We presumed that the *VA* [expressed as Equation (2)] is created by the available protective cushioning with average social functionality value. Therefore, the *RVA* created by certain protective cushioning is the *VA* multiplied by the *K*, as expressed in Equation (8). Finally, the *SI* incorporating social, economical and environmental aspects is expressed as Equation (9). The bigger the *SI* is, the better the sustainability of the protective cushioning.

$$K = V^n / \overline{V}^n \tag{7}$$

$$RVA = K \times VA \tag{8}$$

$$SI = \frac{f(RVA)}{f(DCC,\ HCC)} \tag{9}$$

where

K = modified coefficient;
V = social functionality value of the protective cushioning;
$\overline{V}$ = average social functionality value of the protective cushioning in evaluation;
n = protective cushioning products ($n = 1, \ldots, N$);
VA = economic gains of the protective cushioning related to market;
RVA = social performance of the protective cushioning;
DCC = total consumptions for obtaining the protective cushioning;
HCC = environmental damage brought by the protective cushioning.

The key steps of the SA were diagramed by Figure 2.

CASE STUDIES

Protective Cushioning for CD Receiver

In this study, various protective cushioning for a CD receiver (182 × 180 × 50 mm, 1.35 kg, for car use only) were evaluated using the SA. EPS protective cushioning for the CD receiver is traditional design. Customer wants to replace it with corrugated board protection cushioning or moulded pulp protective cushioning. We carried out the overall evaluation for identifying optimum selection and potential improvements in

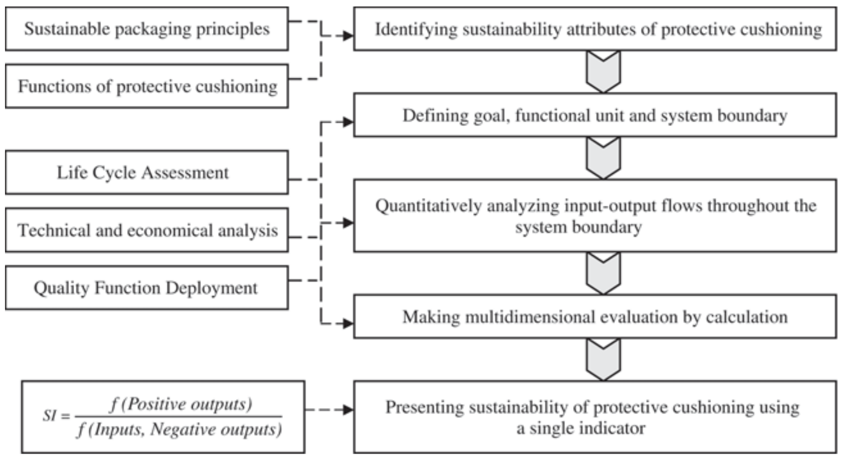


Figure 2. *An overview of the SA approach.*

the new designs. The CD receiver and the new protective cushioning designs were shown in Figure 3(a), (b) and (c). The new protective cushioning products are able to standardize the cushioning design by some structure factors and meet technical requirements of user. The three protective cushioning showed technical variations in Table 2. The old one was also involved for comparing improvement degree on functionality of the protective cushioning.

Analysis and Evaluation

For this study, the functional unit (FU) was defined as a set of cushions for packaging the CD receiver ready for dispatch. The system boundary on basis of the life cycle commenced with the raw material

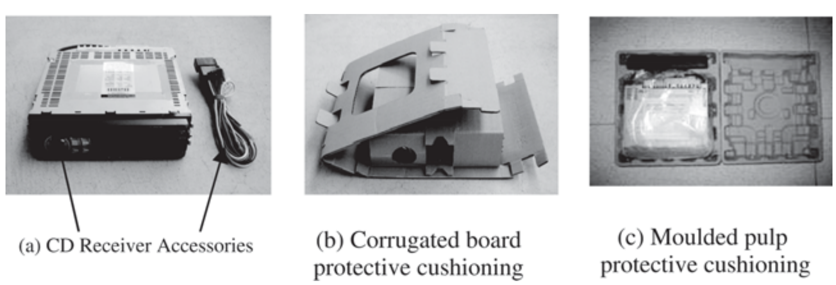


Figure 3. *CD receiver and two new protective cushioning designs.*

Table 2. *Technical Variations of the Three Protective Cushioning Designs.*

Items	EPS Protective Cushioning	Corrugated Board Protective Cushioning	Mould Pulp Protective Cushioning
Materials and forming	Polystyrene, vesicant (8%) and dispersant (5%) By aluminum mould tool used (2)	New pulp and used paper K180/SCP160/K180, B/F (3)	Used cardboard and newspaper By aluminum mould tool (4)
Weight (kg/set)	3.000E-02 (4)	1.52E-01(3)	2.15E-01(3)
Fragility (G_{max})	25 (4)	56 (3)	58 (3)
Packing	Easy (3)	By folding along pressed marks. (2)	Easy (3)
Water absorption	No (4)	Yes (2)	Yes (2)
Speckiness	Very low (4)	Low (3)	Low (3)
Causticity	No (4)	No (4)	No (4)
Mildew resistance	Good (3)	Moderate (2)	Moderate (2)
Weather resisting property	Good (3)	Good (3)	Good (3)
Flammability	Yes (1)	Yes (1)	Yes (1)
Disposal	Landfill or incineration (0)	Recycle or landfill (4)	Landfill (4)

Note: Scores in parenthesis (4 = very good; 3 = good; 2 = moderate; 1 = poor; 0 = very poor.) were given by experts for making calculation.

collection and ended with the protective cushioning disposal, as shown in Figure 1. The study did not include transportation of raw materials, product distribution and disposal of the protective cushioning. As some in-house data was not available, we applied comparable average data assuming a similar situation exists in this study.

By means of investigative and calculative actions, the input-output flows of the evaluation targets within the system boundaries were quantified based on the FU, and multidimensional life cycle inventories (LCI) were created. The LCI of the input-output flows for the evaluation targets in a monetary unit was shown in Table 3. The LCI of main emissions to environment of the evaluation targets based on the FU and subsequent characterization result were shown in Table 4 and Table 5. JEMAI-LCA Pro with associated databases [7], which is developed in accordance with the LIME model was used in the evaluation.

The environmental impacts of the protective cushioning were further assessed by the LIME. The results of integrated environmental impact of the protective cushioning (involving various life cycle stages) were presented in Figure 4 and Figure 5(a). The HCC of the corrugated board

Table 3. LCI of Input-output Flows Based on a Monetary Unit for the Two New Protective Cushioning (a set of cushions for packaging a CD receiver).

Items (CNY)	Corrugated Board Protective Cushioning	Mould Pulp Protective Cushioning
SVcontained contents	9.000E+02	9.000E+02
Ccontained contents[1]	3.150E+02	3.150E+02
ACprotective cushioning[2]	9.450E+00	9.450E+00
Cprotective cushioning[3]	2.400E+00	2.000E+00
DCC[4]	1.800E+00	1.500E+00
VA	1.755E+01	1.755E+01
K	9.382E–01	1.036E+00
RVA	1.647E+01	1.818E+01

[1,2,4]Average of data was adopted ($C_{contained\ contents}$ is 35% of $SV_{contained\ contents}$; $AC_{protective\ cushioning}$ is 3% of $C_{contained\ contents}$; DCC is 75% of $C_{protective\ cushioning}$).
[3]$C_{protective\ cushioning}$ is practical price of the protective cushioning.

protective cushioning and moulded pulp protective cushioning based on the FU were 2.08 and 7.34 JPY respectively. The Eco-indicator'95 and Ecopoint model also demonstrated a similar situation, as shown in Figure 5(b) and (c).

Results and Discussion

The RVA, DCC and HCC representing total inputs and outputs of the protective cushioning were compared through calculation, as shown in Figure 6(a) and (b). The SI of the corrugated board protective cushion-

Table 4. LCI of Main Emissions to Environment for the Two New Protective Cushioning (a set of cushions for packaging a CD receiver).

Main Emissions to Environment (kg)	Corrugated Board Protective Cushioning	Mould Pulp Protective Cushioning
CO_2	2.387E–01	8.327E–01
CH_4	2.895E–07	1.475E–06
N_2O	8.930E–06	8.798E–05
NO_x	8.962E–04	1.780E–03
SO_2	2.914E–04	3.459E–03
SO_x	3.582E–04	1.232E–06
Dust	2.317E–05	2.786E–04
Hydrocarbons	7.960E–07	7.171E–06
COD	1.034E–05	1.462E–05
Industrial waste landfill	1.520E–01	2.150E–01
Sludge (landfill)	2.496E–01	3.440E–01

Table 5. *Characterization of the Main Emissions to the Environment for the Two New Protective Cushioning (a set of cushions for packaging a CD receiver).*

	Indicator Results	
Impact Category	**Corrugated Board Protective Cushioning**	**Mould Pulp Protective Cushioning**
Global Warming	2.414E–01	8.587E–01
Acidification	1.292E–03	4.735E–03
Eutrophication	9.928E–06	1.970E–05
Photochemical Oxidant	3.366E–07	3.033E–06
Solid Waste	5.605E–04	5.590E–04
Resource Consumption	3.369E–07	1.983E–06
Fossil Energy Resource Consumption	1.576E+00	1.002E+01

ing and moulded pulp protective cushioning were 8.46 and 9.03, in the order given, as shown in Figure 6(c). The 2008 exchange rate was adopted (100 JPY equivalent to 7 CNY) [12].

The results of the multidimensional analysis show that the moulded pulp protective cushioning has advantage over the corrugated board protective cushioning in social performance because of more RVA created with less DCC; whereas the integrated environmental impact generated by the moulded pulp protective cushioning is more than three times of that of the corrugated board protective cushioning because of more energy consumption and emissions to air during the forming stage in production. The results of the LCI of main emissions to environment and

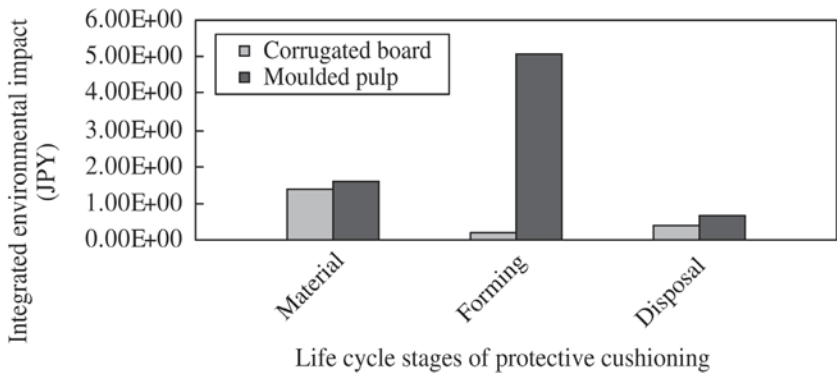


Figure 4. *Results of integrated environmental impact on various life cycle stages of protective cushioning.*

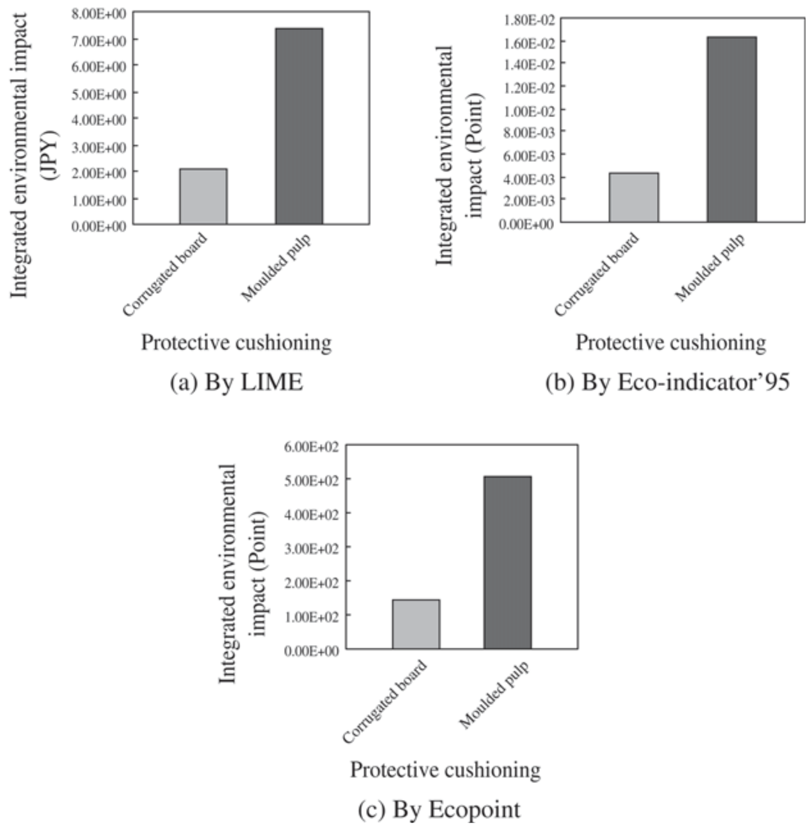


Figure 5. *Final results of integrated environmental impact of two protective cushioning.*

subsequent characterization show that the main environmental burdens of the two protective cushionings are due to atmospheric emissions, COD and industrial waste landfill. They result in impacts upon global warming, acidification, eutrophication, photochemical oxidant, and so on.

The final results of the SA show that the moulded pulp protective cushioning has advantage over the corrugated board protective cushioning in overall benefits, although it consumes more total costs than that of the corrugated board protective cushioning. The findings indicate that moulded pulp protective cushioning design should be the first option for capturing good overall benefits in the development of protective cushioning for the CD receiver. But the moulded pulp protective cushioning would be better for achieving sustainability if the forming stage in production could be improved. However, as the case study results rely partly on assumptions in the data and subjective scores given by experts, specific data should be generated for more accurate results.

CONCLUSIONS

A sustainable assessment approach, incorporating sustainable attributes in social, economic and environmental aspects, was utilized to evaluate protective cushioning designs for electronic products. Total inputs and outputs representing the protective cushioning performance in social, economic and environmental perspectives were identified and expressed with monetary units. Moreover, the overall benefits of the protective cushioning were indicated by the SI. The SI is recognized easily so that the trade-off analysis is transparent to stakeholders throughout the packaging/product system. In particular, a newly developed RVA was used to assess the social performance of the protective cushioning. The findings showed the advantages and disadvantages in various as-

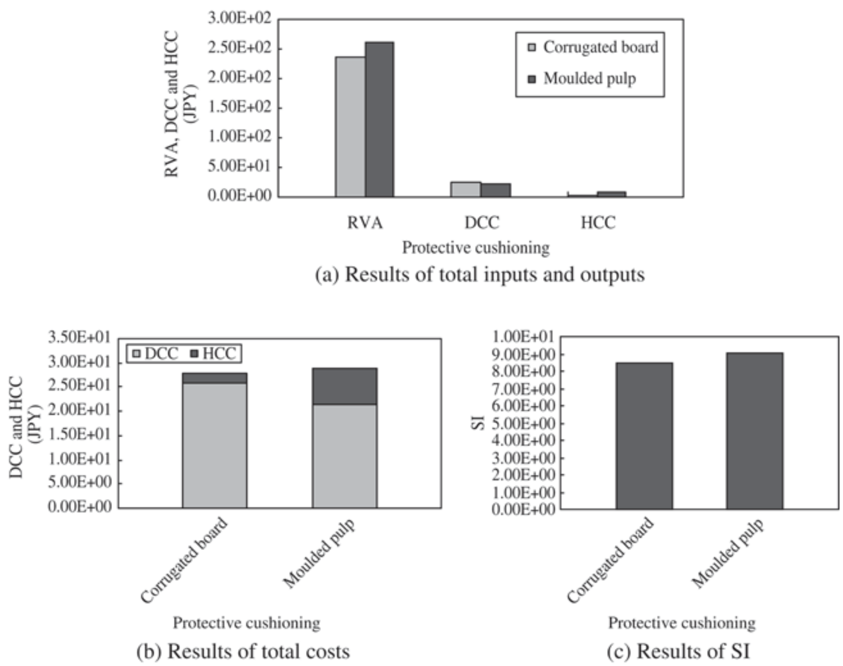


Figure 6. *Final results of the SA of protective cushioning.*

pects of each evaluated targets. They can offer guidance for both packaging designers and packaging consumers. The proposed SA approach can assist in identifying improvements to sustainability of the protective cushioning.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the financial support provided by Japan Society for the Promotion of Science for this study (CSC-10608). The authors would also like to thank the referees for their invaluable comments on this paper and appreciate Dr. Geoff Thomas, RMIT University, Australia for checking English.

REFERENCES

1. Wal-Mart packaging scorecard. *http://www.scorecardmodeling.com/* Accessed on March 15, 2008.
2. Verghese K., Horne R. and Fitzpatrick L. PIQET-A packaging decision support tool. *http://www.sustainablepack.org.* Accessed on November 7, 2008.
3. Wever R. and Tempelman E. The social component of sustainable packaging. *Proceedings of the 24th IAPRI symposium on packaging.* USA, June 2009.
4. Rebitzer G., Ekvall T. and Frischknecht R. et al. Review on Life cycle assessment Part 1: Framework, goal and scope definition, inventory analysis, and applications. *Environment International.* 2004; 30: 701–720.
5. Huo L., Saito K. Multidimensional life cycle assessment on various moulded pulp production systems. *Packag. Technol. Sci.* 2009; 22: 261–273.
6. Park PJ, Tahara K. Itsubo N. et al. Estimation of product sustainability by combining quality, environmental, and economic aspects. *Proceedings of the 6th International Conference on Ecobalance.* 2004; 69–73.
7. Japan Environmental Management Association for Industry. *JEMAI-LCA Pro Manual.* 2007.
8. Itsubo N, Inaba A. A new LCIA method: LIME has been completed. *Int J LCA.* 2005; 8(5): 305.
9. Kobayashi Y., Kobayashi H., Hongu A. et al. A practice method for quantifying Eco-efficiency using Eco-design support tools. *Journal of Industrial Ecology.* 2005; 9(4): 131–144.
10. Sonneveld K., James K. and Fitzpatrick L. et al. Sustainable Packaging: How do we Define and Measure It? *Proceedings of the 22nd IAPRI symposium on packaging.* Brazil, October 2005.
11. Sustainable packaging definition. *http://www.sustainablepackaging.org.* Accessed on December 17, 2006.
12. Exchange rate. *http://www.bankofchina.com/sourcedb/lswhpj.* Accessed on November 20, 2008.

Bin Packing of Potted Plants for Efficient Transportation

RENÉ SCHUMANN[1,*], MATTHIAS POSTINA[2] and JÜRGEN SAUER[3]

[1]*Goethe University Frankfurt am Main, Robert-Mayer-Str. 10, D-60325 Frankfurt, Germany*

[2]*OFFIS Institute for Information Technology, Escherweg 2, D-26121 Oldenburg, Germany*

[3]*Carl von Ossietzky University Oldenburg, Ammerländer Heerstr. 114-118, D-26129 Oldenburg, Germany*

ABSTRACT: The transportation of plants is expensive since plants have a comparable high volume in relation to their value. Shipping costs are directly dependent on the quality of the packing, so as a consequence effective packing secures a competitive advantage for a plant nursery. In this paper we present the potted plant packing problem—a problem from this domain—which is a special instance of a 3D bin packing problem. A heuristic approach is demonstrated to solve the problem on the basis of already existent algorithms. As these initially adopted algorithms were outperformed by human packers, further improvements like stacking were introduced.

1.0 INTRODUCTION

THE transportation of plants is expensive as they require careful treatment due to their fragility. Furthermore a plant has a comparable high volume in relation to its actual value. For shipment, potted plants are normally loaded on transport trolleys (shown in Figure 1) and transportation costs for a given order are calculated based on the number of trolleys needed for shipment. In order to minimize transportation costs, the efficient packing of trolleys is necessary. This becomes even more important with the ongoing shift towards DIY stores and discounters and their demand for smaller orders at shorter distribution intervals.

This paper discusses the potted plant packing problem, which is a special 3D bin packing problem. It is a practical planning problem of one of our customers. We start with an informal description of the problem and switch to a formal presentation afterwards. In section 5 we present the

*Author to whom correspondence should be addressed. Email: reschu@informatik.uni-frankfurt.de

1557-7244/10/01 015-26

Figure 1. *Trolley for plants.*

problem decomposition. First plants are placed in layers, which is a 2D bin packing problem, where a number of small circles have to be placed in one large rectangle. Then the filled layers have to be mounted into the trolleys, which is a 1D bin packing problem. We present strategies to solve these problems. For the layer packing, two different sub-problems can be defined. The packing of equal circles into a rectangle, we call this homogeneous packing, and the placement of unequal circles into a rectangle, which we call heterogeneous packing. A typical strategy applied by human packers to optimize the plant density of a single layer is plant stacking. The concept of stacking plants is adapted to increase the quality of packing plans. The aspect of stacking is shown in section 5. To our knowledge, the detail of presentation is a new approach in the field of packing problems. After the algorithms have been described, we present first results in section 6. Finally we draw our conclusion and outline further work.

2.0 INFORMAL PROBLEM DESCRIPTION

When solving the potted plant packing problem, the major task is computing a valid packing instruction for a given order. An order consists of any number of order items. Every order item relates to a specific article and defines the ordered quantity.

Each article, a potted plant for instance, has various attributes like height, width, depth, weight and type of packing-unit (e.g. a flowerpot). Of course there are further attributes (price for example) which we neglected here since they are not related to the packing problem. Some of these attributes have to be computed, as the dimensions of a plant are subject to change over time.

Each plant is packed into a packing-unit like a flowerpot or a tray. Consequently a packing-unit can hold one or more plants of one article. The shape of a packing-unit can either be rectangular or circular.

It is possible that parts of one order are supplied by external plant nurseries. Such partial orders arrive pre-packed on so called cc-trolleys at the central packing facility and typically remain untouched because repackaging would usually lead to a substantial additional effort.

Our main objective is the computation of a valid packing plan, based on the order data, especially looking at the number of trolleys needed. Every packing-plan consists of a number of trolley-packing-instructions, one for each cc-trolley of an order. Such a trolley-packing-instruction itself consists of a number of layer packing-instructions. They define the exact position of the layer (mounting height) along with detailed packing instructions for all packing-units on the layer. Each single layer consists of a set of packing-units along with the exact position of every packing-unit on the layer. To clarify the problem, a trolley is shown in Figure 1.

As is often the case in optimization problems a goal conflict arises. On the one hand very effective placements have to be computed and on the other hand the plants have to be handled with care and the packed trolleys should be stable. Techniques like stacking of plants turned out to be very relevant for the quality of the computed plans but challenge aspects of stability. Thus the generation of a valid, stable and realizable packing-instruction requires the observance of a number of constraints. Usually a differentiation is made between soft constraints (e.g. keeping order items together on one layer or trolley) and hard constraints like the fixed size of layers or the maximal allowed height of a loaded trolley.

3.0 TOWARDS A FORMAL REPRESENTATION OF THE PACKING PROBLEM

3.1 Problem Characterization

A first step towards a formal description of the potted plant packing problem is a formal characterization of the problem. We follow the commonly used characterization of cutting and packing problems introduced by Dyckhoff [1]. Dyckhoff advocates the use of some typical characteristics to identify similar groups of cutting and packing problems. These characteristics are:

1. dimensionality (1, 2, 3, *n*)
2. kind of assignment (*B*, *V*)
   - (*B*) All objects and a selection of items
   - (*V*) A selection of objects and all items
3. assortment of large objects (*O*, *I*, *D*)
   - (*O*) One object
   - (*I*) Identical figures
   - (*D*) Different figures
4. assortment of small items (*F*, *M*, *R*, *C*)
   - (*F*) Few items (of different figures)
   - (*M*) Many items of many different figures
   - (*R*) Many items of relatively few different figures
   - (*C*) Congruent figures

Detailed discussions about this notation are provided by Dyckhoff [1] and Washer et al. [2]. A problem description of a cutting or packing problem is a 4-tuple, describing the problem in respect to the mentioned characteristics.

Following Dyckhoff's description, the potted plant packing problem belongs to the class 3/*V*/*D*/*R*. This notation encodes that,

- the problem has three relevant dimensions (length, width, height),
- all small objects (plants) have to be placed within a large object (trolley),
- the large objects (trolleys) can have different dimensions (trolley with or without add-on modules)
- and that there are many small objects of relatively few different figures.

This typology is widely used to describe the main characteristics of cutting and packing problems. Currently an improved typology is discussed by Washer et al. [2]. According to Washer, a problem like the plant packing problem can be seen as a special type of the Three Dimensional Multiple Bin Size Bin Packing Problem. A current survey about existing literature of packing and cutting problems can be found in Washer et al. too.

The major disadvantage of these notations is the lack of an overall problem description. Such notations are not able to cover the problem entirely. Additional constraints have to be added to express restrictions like allowed plant alignment (plant upstanding, no bottom-up packing) or stacking constraints (cf. section 5.5). Nevertheless, these notations are widely used because they allow a classification of problems.

3.2 Problem Modeling

One general approach when describing planning problems is to use a tuple of parameters, which are capable of describing the entire problem. Scheduling problems for example are described in such a way by Keng et al. [3] or Sauer [4]. The potted-plant packing problem can be described as a 6-tuple consisting of

- *Resources*—Trolleys with height (*H*) determined by either the truck-roof or by the trolleys' highest mounting point for layers.
- *Objects*—These are the objects the algorithm is supposed to pack. We actually do not focus on the plants themselves, but on an artificial entity called packing-unit instead (a cylindrical virtual container with radius R_c, as shown in figure Figure 2). We use this simplification because a typical instance of the potted-plant packing problem can have some 50.000 different plants. Using this abstraction by building plant categories Cat_n ($n \in \mathrm{IN}$), we can significantly reduce the number of different entities the algorithm has to respect.

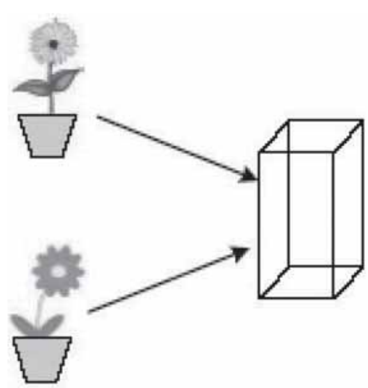


Figure 2. *Concept of categories.*

- *Order*—An order comprises of a set of order items, each declaring a number of specific packing-units which have to be packed on trolleys. Additionally the order details, the customer as well as the delivery and packing time.
- *Hard-constraints*—like:
  - —stability of packed plants
  - —Safety-aspects for stacking plants, ensuring no plant is damaged by stacking
  - —any single packing-unit has to be stored in its entirety on one layer
  - —a layer has to be mounted correctly into one trolley
  - —the overall trolley height is lesser than or equal to the allowed maximum
  - —any single layer can be mounted into one trolley only
  - —a packing-unit cannot be stored bottom up
- *Soft constraints*—such as:
  - —The packing-units of an order item are placed contiguously on layers.
- *Objective function*—The objective function $h(O)$ is to minimize the number of trolleys needed for packing all potted plants of an order onto trolleys.

Trolleys are modeled as resources explicitly, because the typical assumption of an infinite number of trolleys and layers available does not hold. For example, a limited number of add-on modules exist for trolleys which can be used to increase the height of trolleys.

An advantage of such a tuple modeling is that all relevant aspects of the current problem can be captured in the model which now covers the problem domain entirely. This is useful when designing a solution method for the problem, even if this model does not impose a certain solution method.

Note: In our object-oriented prototype, we modeled constraints as implicit statements included in our algorithms. This hinders a generic constraint modeling but allows faster prototyping to stay in step with actual practice.

4.0 RELATED WORK

As the potted plant packing problem is directly adopted from the customer needs it is not possible to compare results with existing approaches since they rely on benchmarks. Those benchmarks typically do not include all constraints to be regarded here. However, it is possible to partially decompose the potted plant packing problem into standard like problems.

Work like [5–7] addresses the placement of equal circles in a rectangle. This complex problem is the special case in our packing of trolley layers. We decide to use the already known quite simple packing strategies for packing equal circles because our plans are more realistic and the results can easily be explained to our partners.

Albrecht et al. [8] present an approach on extending the packing of equal circles. The circles are assumed to be flexible and thus can be deformed when they are squeezed. This is an interesting approach, as plants were squeezed as well during the packing. But as already stated the packing of equal circles is a special case in plant packing and thus the existing approach had to be extended drastically.

Compared to sphere packing from the field of computational geometry where dense packing of identical objects is observed we are dealing with different objects. Not even the shape of our objects is similar when it comes to stacking. Nevertheless, we were inspired by Conway et al. [9] when it comes to pattern identification for plant alignments.

An early problem statement containing the problem of placing different sized circles into a rectangle was presented by George et al. [10]. The presented packing problem is decomposed as well, and the sub-problem of packing different size circles into a rectangle is addressed as well. Domain dependent stability criteria were already defined and thus specialized solutions were generated.

The problem of packing unequal circles into a rectangle is tackled by Schöning et al. [11]. This problem corresponds to our 2D packing problem, but the presented solution has the restriction that the regarded bobbins can only be placed from one site to the other. This restriction was introduced to be able to control a packing robot efficiently. Unfortunately such a restriction is not useful for our problem. The main idea for their packing algorithm was to simulate force of gravity that moves all circles towards the bottom of one layer.

Dowsland et al. [12] are regarding the problem of cutting different circles from one rectangle. This problem has some similarities to the packing of trolley layers. The presented solutions works on local search algorithms and uses the idea to group circles of equal size together in quite regular placements.

Huang et al. [13] also consider the problem of placing unequal circles into a rectangle. The authors present a greedy heuristic to solve the problem. Their algorithms are inspired by human packing habits to a great extend. For that reason we choose this algorithm in our actual implementation as it seems to be an efficient strategy and, moreover, it can be easily explained to our partners (in contrast to a solution based on metaheuristics like stated by Correia et al. [14]). Additionally it can be noted that this algorithm could be further improved by measures as described by Akeb and Li [15]. Actually these improvements are not implemented in our prototype, so far. A slight difference to our problem is that they try to minimize the used size of the rectangle enclosing all circles which is not the case in our application.

5.0 SOLVING THE POTTED PLANT PACKING PROBLEM

In this section we present concepts and algorithms solving the potted plant packing problem. The reuse of existing algorithms and partial solutions was a crucial concern when designing the system. Whenever possible we decomposed the problem into standardized sub-problems. The whole problem can be described by

- identifying patterns (5.4.1 and 5.4.2),
- placing plants on one layer (5.4),
- additional stacking of plants in one layer (5.5) and
- Composing of layers to optimal height of trolleys (5.6).

Thus we present an approach consisting of

- complexity reduction by building plant categories (5.1),
- decomposition of the problem in layer packing and layer distribution (5.2) and
- Presentation of algorithms for layer packing and stacking (Algorithm 1 and Algorithm 2).

Additionally it was intended not to compute all of the intermediate data at runtime. Especially the computation of layers containing different sized plants can be time consuming. For that reason layers are computed offline and stored in a database.

5.1 Reducing Complexity by Building Plant Categories

For the potted plant packing problem the search space is defined as the number of possible valid packing instructions. This number is determined by the number of different plants. It should be obvious that the search space size mainly affects the distribution of plants on layers. As already stated in the problems formal representation in Dyckhoff's notation, there are a large number of small objects with differing dimensions. The entire article database of our customer has more than 50,000 entries and more than 300 different articles, which can be contained in one order. A diminution of the number of these potential packable items—and thereby of the search space—is desirable. This should have a positive impact on overall performance, simplify the problem, and make it more tractable. To implement such a diminution, we decided to build categories of plants with similar dimensions. A category can be regarded as a box or cylinder which is able to contain different plants with similar dimensions. The classification will go beyond the mere exclusion of irrelevant attributes. However, it will also be necessary to look for similarities between different plants. The concept of categories is sketched in Figure 2.

Building categories necessitates a design decision concerning the number of different categories. Reducing the number of categories implies decreasing precision, but also reduces the heterogeneity of figures to pack. This can best be illustrated when looking at the two extremes:

1. There exists only one category, resulting in a maximization of wasted space and a minimization of the number of categories.
2. Each group of plants with identical dimensions has its own category, resulting in a minimization of wasted space and a maximization of the number of categories.

As explained above, those extremes have a substantial impact on the size of the search space for packing layers. To find the optimal number of categories, we advocate the computation of the wasted space for a given number of categories and analyze the gradient of the resulting curve. The graph of such an analysis is shown in Figure 3.

5.2 Problem Decomposition

To be able to compute solutions faster and to reuse existing ap-

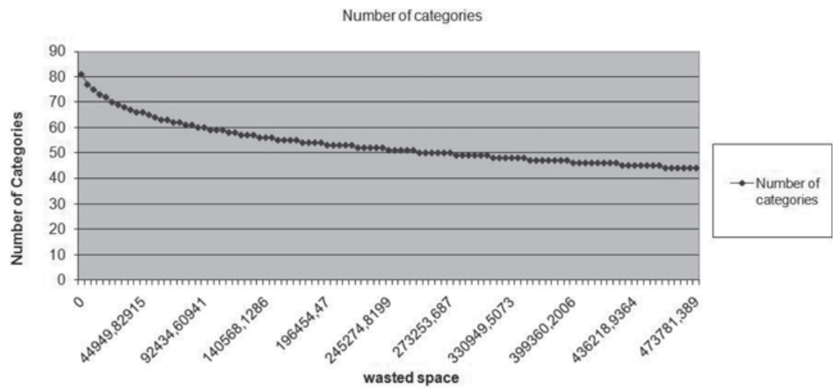


Figure 3. *x-axis: wasted space; y-axis: number of categories.*

proaches as far as possible we decided to decompose this 3-D bin packing problem into a 2-D bin packing and a 1-D bin packing problem. This is possible because one can independently compute the layers, which is a 2-D bin packing problem. These layers have to be mounted into trolleys, corresponding to a 1-D bin packing problem.

Nevertheless our overall goal is to minimize the number of trolleys. Due to the aforementioned problem decomposition the number of needed trolleys is computed within the distribution of layers to trolleys. A critical aspect resulting from the problem decomposition is the design of the objective function for the first planning step, the distribution of plants to layers. It is not necessarily the minimization of layers, but as detailed in the following the minimization of the summed height of all layers. In consequence it is more promising to reduce the number of high layers instead of the total number of layers.

But one can state, that each trolley can carry plants up to a fixed height (H). This (H) is determined by the highest mounting point for layers or the height of the truck, which transports the trolleys. So if a trolley can carry n layers and $h(L_i)$ is the height of layer i, one can state that:

$$H \geq \sum_{1 \leq i \leq n} h(L_i)$$

The overall height of a layer ($h(l)$) is indicated by the height of the tallest plant ($h(p)$) placed thereon. This can be stated as

$$h(l) = h(p_i); p_i \in elements(l); \forall p_j : p_j \in elements(l), h(p_i) \geq h(p_i)$$

For the packing of layers the following objective function $h(O)$ could be used, which leads to a close to minimal number of trolleys.

$$h(O) \geq \sum_{1 \leq i \leq n} h(E_i)$$

5.3 Frame Algorithm for Packing

This section describes the basic planning process. First, all plants of an order are sorted into queues, with a dedicated queue for all categories having the same height. Queues themselves are sorted descending by the heights of the corresponding categories. Then all plants of the first none empty queue are added to the orders working set. If a packing pattern can be applied to a part of the items in the working set, a corresponding layer is introduced and those elements are removed from the working set. This is repeated until all queues are empty and no further packing pattern can be applied. If by that time the working set still contains further elements, additional layers have to be computed by the online layer packing algo-

Algorithm 1 packing frame algorithm

```
Ts = ∅  // trolley set
Ws = ∅  // working set of packing units
Ls = ∅  // layer set
Ap = ∅  // applicable packing pattern
for each o ∈ O // where O is an order and o a order item do
  P = packing-unit(o)
  q = queueForCategory(category(p ∈ P)) //a Queue for each category
  enqueue(q,p)
end for
while ∃q ∈ Q; |q| > 0 OR |Ap| > 0 do
  if |Ap| > 0 then
    p = findBestPattern(Ap)
    Ls = Ls ∪ newLayer(p)
    Ws = Ws − elementsOf(p)
  else
    Ws = Ws ∪ deque(q; q ∈ Q, ∀q′ ∈ Q : |q′| > 0, |q| > 0, h(q) ≥ h(q′))
  end if
  Ap = findapplicable(Ws)
end while
if |Ws| > 0 then
  Ls = Ls∪ onlineLayerComputations(Ws)
end if
Tr = distributeLayers(Ls)
```

Algorithm 1. Packing frame algorithm.

rithm until all plants have been packed. Then all computed layers are packed into trolleys. Trolleys and their dedicated layers are combined which leads to packing instructions. This procedure is sketched in pseudo-code in Algorithm 1.

As already stated the combined height of all layers has a strong influence on the packing instruction's quality of the second planning step, as the layer height, defined in the layer packing, is an important aspect for mounting the layers into the trolleys. As a consequence it is desirable that the summed height of all layers is minimal. To minimize $h(O)$, all plants within an order are sorted descending by their height. The sequence in which plants are added to the working set depends on this sorting. Because of the order in which plants are then added (and thus packed) to the working set, layers will generally contain plants of similar height which minimizes the combined height of all layers.

5.4 Layer Packing

As already mentioned the reuse of computed layers is intended. Therefore, packing patterns are introduced. A packing pattern encodes a valid packing of packing-units of certain groups on a single layer. A packing pattern can contain one or more groups of packing-units. Figure 4 shows an example of such a packing-pattern containing packing instructions on one kind of packing group per layer only.

Our approach is limited, since we only allow round pots and no rectangular trays. So the packing of layers corresponds to a relatively common packing problem; the problem of packing circles into a rectangle. In the notation of Dyckhoff the problem can then be characterized as $2/V/I/R$. Meaning that:

- the problem has two relevant dimensions (length, width),
- all small objects (plants) have to be placed within the large objects (layers),
- large objects are identical in size
- and there are many small objects with relatively few different dimensions.

The height of a plant can be ignored when only considering its placement as a circle in a rectangle. It has to be considered of course, as the dimension defining the layer's overall height, which was mentioned above.

The problem of packing circles into a rectangle can be divided into two sub-problems, namely the packing of circles of equal and unequal sizes. These cases differ in their complexity and are discussed in two separate sections.

5.4.1 Homogeneous Packing Patterns

The packing of equal circles into a rectangle is a standard geometrical problem. Even though optimal packings are known for a growing number of circles (e.g. listed by Nurmela and Östergård [5]) this is a NP-hard task. However, heuristics can be designed which compute solutions with sufficient quality very fast. These heuristics are based on a regular placement of circles. We implemented three regular placement strategies, namely

- grid placement
- placement along the depth
- placement along the width

As one can see in Figure 4 the number of placements can vary depending on the strategy. So far we cannot predict which of these will offer the best performance for a given circle and rectangle size. Since these heuristics are very fast, we always compute all three alternatives and then choose the best.

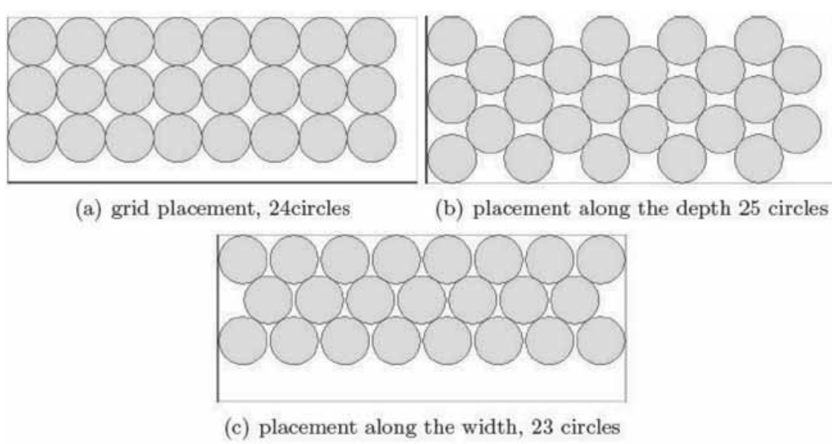


Figure 4. *Regular circle placements.*

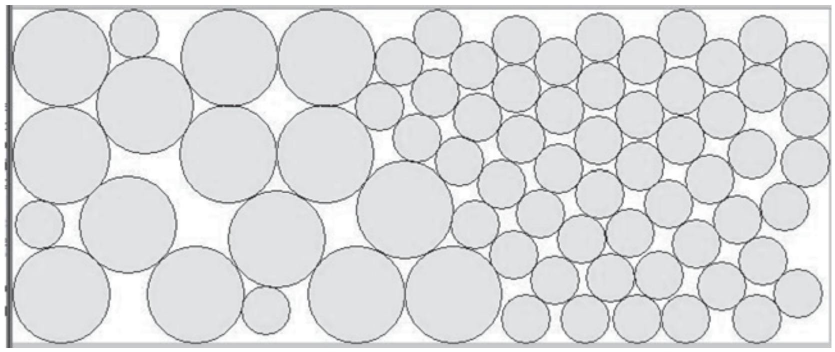


Figure 5. *Placement computed by the maximum hole degree algorithm.*

5.4.2 Heterogeneous Packing Patterns

The placement of circles of differing sizes into a rectangle is more difficult. Only a few publications have dealt with this problem so far, for example [10,11,13,14]. We decided to implement the maximum hole degree algorithm (B1.0) presented by Huang et al. [13] because this algorithm uses a constructive heuristic and thus has two advantages

- it computes deterministic solutions, which is important and
- the results can be explained to non technical staff with minor knowledge in optimization.

Moreover the results presented by Huang et al. indicate that this heuristic can compute competitive solutions. The clou of this algorithm is the subsequent placement of circles into corners. Where a corner is defined by

- two sides of the rectangle,
- rectangular side and a circle or
- two circles.

The first two circles are placed by a simple placement strategy. Then for each circle not already placed in the rectangle, all possible corner placements are computed. The circle being associated with the placement having the minimal distance to another circle or side is then chosen next. This is repeated until no more valid corners are found or all elements have been placed. A detailed description of the algorithm as well as a complexity analysis can be found in Huang et al. [13]. An example of a layer containing two different types of circles is shown in Figure 5.

The original algorithm presented by Huang et al. [13] has the objective to minimize the size of the rectangle needed to place a fixed number of circles. Of course this objective is not useful to solve the potted plant packing problem, as the size of the layers is fixed. The task here is to place as, many circles as possible into a rectangle of fixed size. Thereby the number of circles is in general much higher than the capacity of one layer. This leads to some adjustments of the core algorithm. The numbers of circles that have to be placed within one layer have to be given to the B1.0 algorithm. If the number is too high, i.e. not all circles can be placed within the layer, the algorithm fails. In the other case, space is wasted on the layers. Thus the maximal numbers of circles from the working set have to be estimated. Therefore we compute upper and lower bounds, using the simple strategies for homogeneous layers mentioned above, and use a divide and conquer strategy to compute the maximal numbers of circles that can be placed in one layer.

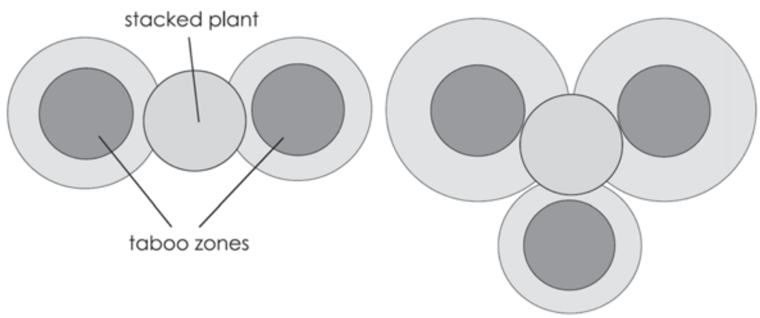
5.5 Stacking

If one compares computed solutions using the already described techniques and packings generated by humans, one can state that humans outperform computed solutions (see section 6). Contrary to programs, human packers have an intuitive understanding of spatial optimization, so stacking plants was observed as a typical human packing habit. To imitate such stacking is consequently the first approach to improve the packing algorithm. This section categorizes the plants stacking problem as the stacking on homogeneous patterns and as the stacking on heterogeneous patterns.

5.5.1 Stacking on Homogeneous Packing Patterns

The terms homogeneous and heterogeneous pattern are related to a layer of a trolley which is already initially packed. Whenever a layer of a trolley is homogeneously packed—which means only plants of the same category with exactly the same pot and plant size were used to fill the basic layer—we say this layer follows a homogeneous pattern. Figure 4 has already shown such homogeneous patterns. In order to support stacking, we need to upgrade the planning algorithms and we have to regard stacking in more detail.

Definition 1: We call a position a *stackable place* whenever a pot could



(a) Non stackable place. (b) Valid stackable place for the stacked plant (assuming same pot hight).

Figure 6. *Visualization of Definitions.*

be placed on the top of a minimum set of three neighbored pots having the same height.

We chose this minimal set of three plants as bed for a stacked plant since plants are fragile goods and falling over leads to a total loss of plant value. So the configuration pictured in Figure 6 (a) would be an illegal configuration according to our definition. For the same reason the stacking procedure needs to take care of the underlying plants. Keeping this in mind, we defined a taboo zone for each plant reflecting its actual shape. These taboo zones have to be respected when looking for valid stackable places. Since we focus on the packing of circular objects it is sufficient to store the center and the radius to indicate a stackable place. Furthermore, we need a statically defined minimal footprint to guarantee a sufficient contact surface and also a maximal boundary to prevent damage from the underlying plants. On the strength of stability, we also restrict the stack height to a maximum of two per trolley layer. Basically this reflects the packing habit of human packers.

Definition 2: We say a stackable place is *valid* for a certain pot category, if the footprint of pots assigned to this category guarantees a sufficient contact surface without violating the taboo zones of the underlying plants.

The set of stackable places is easy to recognize when the basic layer follows a homogeneous pattern. One needs to identify the size of the radius, the size of the taboo zone and the used placement strategy (cf. Figure 4). With this information one can calculate an offset along a direction vector pointing out of each underlying circle center. So finding the center of the stackable places could be seen as a pattern/raster shift of the underlying layer. Such a pattern shift is shown in Figure 7 where the center of each stackable place is moved along the direction vector $\overline{v}$.

5.5.2 Stacking on Heterogeneous Packing Patterns

In analogy to homogeneous pattern, we say a layer follows a heterogeneous pattern, when plants of different categories (with miscellaneous pot diameters) and different pot heights were used to fill the basic layer of a trolley. Figure 5 shows such a heterogeneous arrangement.

Obviously it is much harder to spot stackable places on the top of a layer following a heterogeneous pattern. Since pot heights may vary one needs to identify regions in the layer where at least three neighbored pots have the same pot height. Furthermore these three pots need to be grouped in a way that offers a valid stackable place—which is close enough to allow stacking but wide enough to respect taboo zones of the underlying plants. Moreover, one needs to calculate the centers of the stackable places for each possible triangular constellation individually. Such a center of a stackable place could be calculated as follows:

When regarding a troika of pots of the same height, the centers A, B and C of these three circular objects form a triangle. We want to find a point P having the same distance to each of A,B and C. Such P is the center of the circumscribed circle of the triangle which is the intersection point of the perpendicular bisectors of the sides. This is shown in Figure 8(a).

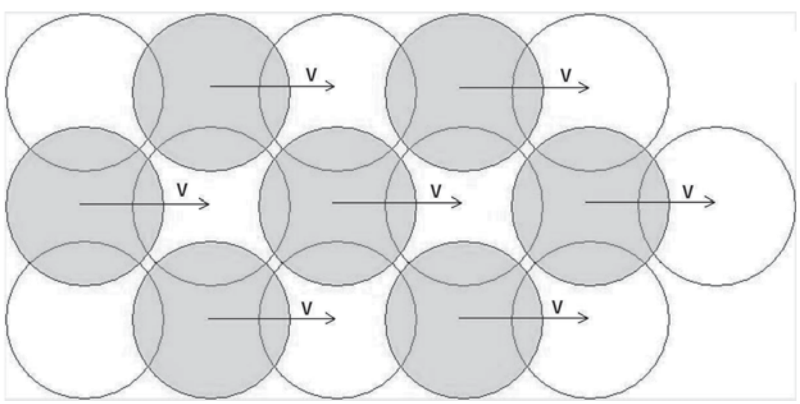
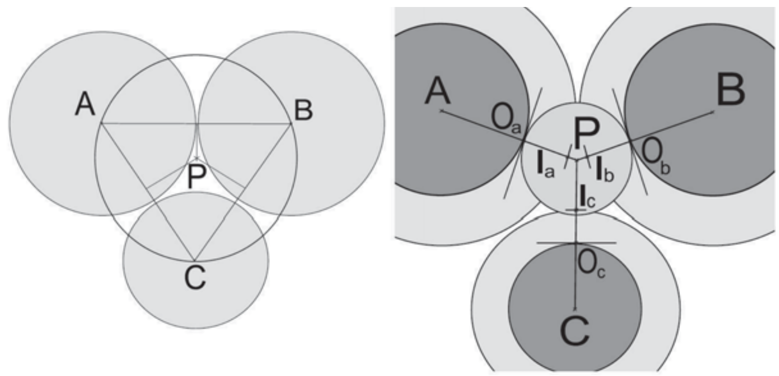


Figure 7. *Postponement along a direction vector v (10 l pots following a homogeneous pattern).*



(a) Finding P as center of a possible stackable place

(b) Identifying the category of the stackable place

Figure 8. *Finding stackable places.*

Now, one needs to show that the identified stackable place having P as center is a valid stackable place for a certain pot category according to Definition 2. In order to do so, we will formalize Definition 2 based on the sketch in Figure 8(b).

Given are the points A, B and C as centers of the three neighbored pots C_a, C_b, C_c with known radii. All of these pots are planted and each taboo zone is known and identified by a taboo radius. We already calculated point P and define $\overline{PA}$, $\overline{PB}$ and $\overline{PC}$ as straight lines—each going through P and one of the centers A,B and C. Further we name the intersection points of these lines with the taboo circles O_i and the intersection points of these lines with the pot bounding circles $I_i (i \in\ a,b,c)$.

To reduce search space we introduced the concept of categories in section 5.1. Still following this concept, each plant is classified as member of a specific cylindrical category. Such a category could be seen as a virtual box hosting plants with similar dimensions. The radius R_c of the cylinder is taken to define a valid stackable place for a certain plant category.

We say the stackable place having P as center is a valid stackable place for a certain plant category Cat_n $(n \in \text{IN})$, if

$$R_c \leq \min[|\overline{PO}_a|, |\overline{PO}_b|, |\overline{PO}_c|]$$

and

$$R_c \geq \max[|\overline{PI}_a|, |\overline{PI}_b|, |\overline{PI}_c|]$$

This calculation method is an early heuristic based on the position of the centers of the regarded pot-troika. Since plant sizes vary over the seasons and even differ between individual plants of the same generation the taboo radii could never be measured exactly. If taboo zones where known exactly one could find better methods for the finding of valid stackable places.

We define the set of stackable places S. Valid stackable places are assigned to plant categories so that each category holds a subset V_n of S, $V_n \subseteq S$ ($n \in IN$ and n is corresponding to the index of the categories Cat_n). The data structure SP is a hash holding the key value pairs of categories and subsets V_n of S:

$$SP = \begin{Bmatrix} Cat_1 => V_1 \\ Cat_2 => V_2 \\ \dots \\ Cat_n => V_n \end{Bmatrix}$$

We further have a second hash CS holding the key value pairs of categories C_n and corresponding plant sets Pl_n ($n \in IN$) which are subsets of all non placed plants:

$$CS = \begin{Bmatrix} Cat_1 => Pl_1 \\ Cat_2 => Pl_2 \\ \dots \\ Cat_n => Pl_n \end{Bmatrix}$$

Algorithm 2 Stacking on heterogeneous patterns

SP `// hash, given as described`

CS `// hash, given as described`

V_i `// subset of` S

Pl_i `// subset of all non placed plants`

while $\exists i \in \mathbb{N} : V_i \neq \emptyset \vee Pl_i \neq \emptyset$ **do**

  $j = \min\{i \in \mathbb{N} : V_i \neq \emptyset \wedge Pl_i \neq \emptyset\}$

  $assign(v_{jx}, pl_{jy})$ `// (` $x, y \in \mathbb{N}, v_{jx} \in V_j, pl_{jy} \in PL_j$ `)`

  $removeElementFromPl_j(pl_{jx})$

  $updateV_x(v_{jy})$

end while

Algorithm 2. Stacking on heterogeneous patterns.

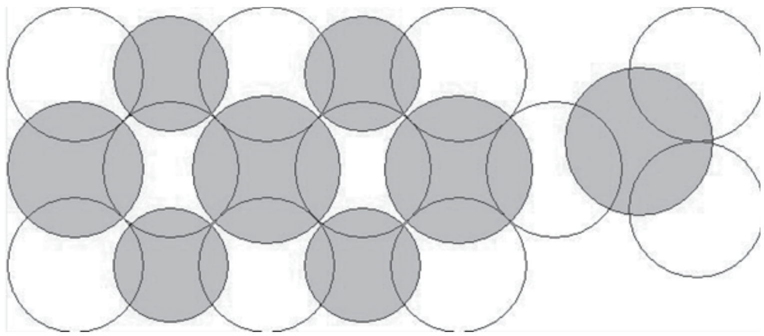


Figure 9. *Stacking on heterogeneous patterns.*

The stacking algorithm formalized in Algorithm 2 works as follows: Given the hashes *SP* and *CS*, the algorithm starts to assign plants in ascending order with respect to their corresponding plant category (which means highest dimension first, ordered by the height of the category) to stackable places (*assign(...)*). During this processing, the hash *SP*—representing valid stackable places per category—is updated permanently ($updateV_x(...)$). This leads to a shrinking set of valid stackable places. Whenever a plant is assigned to a stackable place, such plant is deleted from *CS* ($removeElementFromPl_j(...)$). The algorithm stops the stacking for a trolley layer, when the hash *SP* is empty or when all plants were placed. Figure 9 shows a graphical representation of an exemplary calculation.

5.6 Trolley Packing

The distribution of packed layers to trolleys is, from an abstract point of view, a classical bin packing problem. But in fact the trolley packing algorithm has to answer two key questions.

1. Which layers are mounted into which trolley?
2. Where is each layer placed within the trolley?

The first question concerning the distribution of layers onto trolleys is a classical bin packing problem. This can be solved using standard algorithms like next fit or best fit.

To be able to answer the second question, we have to compute each layer's correct mounting point. This has to be done while observing the height and weight constraints of each layer as well as the trolley's layout and the maximum height allowed during transport. Figure 10 taken from our prototype illustrates the typical trolley layout. Layers are hooked into mounting points, which are generally found at 5 cm intervals from a base of 20 cm up to a height of 190 cm. The available height can be increased through the use of add-on modules up to a total of 225 cm. The placement of layers within a trolley follows a simple strategy:

The tallest layer is hooked into the topmost mounting point that still ensures the adherence of all other constraints, especially the maximum allowed height. All remaining layers are then sorted in ascending order by their weight and inserted top to bottom into the trolley. This strategy aims at two goals. It tries to

- maximize the usage of available space on the truck and
- bring the centre of gravity to the lowest point for stability reasons.

Due to the use of add-on modules the layout of a trolley might change, this requires no change in the method of computation of the layer placement however.

6.0 FIRST RESULTS

We have implemented a prototype (see Figure 10) to evaluate our approach. This prototype was developed for demonstration purposes and to enable our partner to start own field tests.

In this section we compare the computed solutions with and without stacking to solutions generated by humans. Even if we integrated stacking into our solution we have to state, that humans still need fewer trolleys. But, as shown in Table 1 the quality of the solutions increases by using stacking.

Analyzing the trolleys packed by humans it should be mentioned that human packers take advantage of the fact that they are breaking rules the planning strategy has to respect. For example plants were stacked up to three levels per trolley layer or plants were stacked on the base of only two plants instead of three. Furthermore, it was possible for human packers to place more plants on one layer and to pack the plants more densely by squeezing plants. Of course such constraint violation is not thinkable for the algorithm. It is actually in discussion that the packing rules should be applied for humans as well to avoid plant damages and of course the model and algorithms are continuously improved to compute more competitive solutions. Another important aspect for the better re-

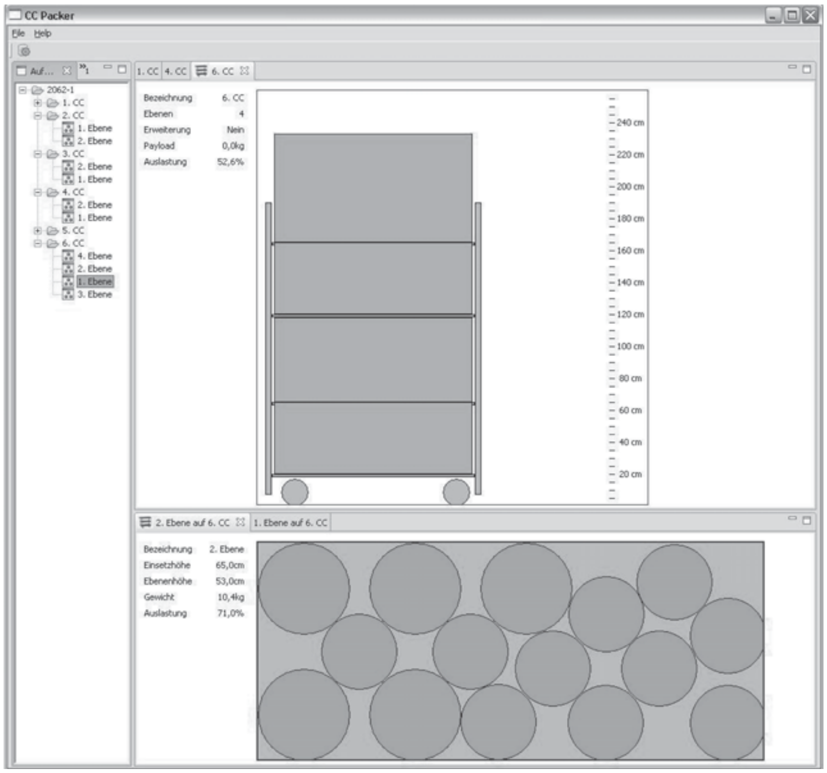


Figure 10. *Screenshot showing a packed trolley..*

sults of human packers is that during the computation pessimistic data for plant sizes is used. So in general real plants are smaller than in our computational model. We are discussing the option to use mean values and not maximum values for plant sizes with our partner, but to do so additional data acquisition is necessary.

Scenario 1 comprises homogeneously and heterogeneously packed layers. Scenario 2 is more difficult because no homogeneous layers exist. Due to the aforementioned model extension concerning the form of pots, the homogeneously packed layers are equal either packed by hand or computed. So the quality difference can be explained by different packed heterogeneous layers and differences between the assumed and real height of the layers. For this reason, the quality of the schedules in scenario 1 is better than in scenario 2.

It is remarkable that in two scenarios (21 and 22) our approach including stacking could produce better results than humans could. In both

Table 1. *Results of the Case Study of Effective Packing.*

		Without Stacking			With Stacking		
Scenario	**#CC Needed**	**#CCs**	**Abs. δ**	**δ in %**	**#CCs**	**Abs. δ**	**δ in %**
1	5	7	2	40%	6	2	20%
2	8	14	6	75%	10	2	25%
3	2	3	1	50%	3	1	50%
4	5	6	1	20%	5	0	0%
5	3	5	2	67%	3	0	0%
6	3	4	1	33%	4	1	33%
7	6	10	4	67%	8	2	33%
8	3	4	1	33%	3	0	0%
9	2	3	1	50%	3	1	50%
10	4	5	1	25%	4	0	0%
11	1	2	1	100%	1	0	0%
12	3	5	2	67%	5	2	67%
13	4	8	4	100%	5	1	25%
14	4	7	3	75%	4	0	0%
15	2	4	2	100%	2	0	0%
16	1	3	2	200%	2	1	100%
17	3	5	2	67%	4	1	33%
18	3	5	2	67%	4	1	33%
19	6	11	5	83%	8	2	33%
20	7	11	4	57%	8	1	14%
21	4	4	0	0%	3	–1	–25%
22	7	11	4	57%	6	–1	–14%
23	6	15	9	150%	8	2	33%
24	8	15	7	88%	10	2	25%
25	5	8	3	60%	7	2	40%
26	4	6	2	50%	5	1	25%
27	9	17	8	89%	12	3	33%
28	5	11	6	120%	9	4	80%
29	4	6	2	50%	5	1	25%
30	2	5	3	150%	5	3	150%
31	5	7	2	40%	6	1	20%
32	8	13	5	63%	10	2	25%
			On Av.	71.6%		On Av.	29.2%

cases the order contained only homogeneous plants of the same category, so only pre-calculated packing patterns were used.

Table 1 shows 30 more test cases ran on our prototype. As one can see stacking can improve the computed results drastically (on average from 71.6% down to 29.2%). Of course the usage of percent values is problematic, as the number of CC-trolleys is a discrete value. But the improvement by stacking is as significant in absolute as in relative numbers.

7.0 SUMMARY AND FUTURE WORK

We present a practical packing problem in this article. As it is provided by our partner, we did not focus directly on solving already standardized benchmark problems, even if we decompose the problem into standard problems. We base our solution method on state of the art packing techniques. We describe how these technologies are applied and extended for the given application. The problem is detailed and a characterization and formalization is presented. For the solution, we decompose the original problem into a 2D and a 1D bin packing problem. Especially the packing of layers, which corresponds to the standard problem of placing circles into a rectangle is discussed. Techniques for the packing of homogeneous layers—containing equal circles—and heterogeneous layers—containing unequal circles—are distinguished. When observing human packers it turned out that plant stacking reflects a usual habit to increase compactness of the packing. Approaches described in other articles do neither deal with packing of goods such as plants nor do they regard stacking of plants or give any useful hints how to solve such a problem. Thus we developed algorithms for stacking on homogeneously and heterogeneously packed layers. All of the described methods are implemented as a prototype and were evaluated by using real world data. Although the results are not as good as plans created and packed by humans, it could be shown that the adaption and integration of stacking into the program increases the quality of plans significantly. We discuss reasons for the different quality of humans and computer generated plans and outline future improvements in terms of higher data quality with non pessimistic assumptions within the input data. The implemented prototype is in a practical evaluation phase, so it can be expected that some tuning has to be done during the tests. It is further assumed that more packing patterns need to be pre-calculated and stored in the database to run the tool efficiently.

For the sake of simplicity we are dealing with plant categories rather than individual plants. However, the categories are either boxes or cylinders whereas a plant pot is usually tapered. i.e., contrary to cylinders the top and bottom diameters of the pots are unequal. Further work lies with detecting if such a simplification leads to proper results or if the data model needs to be updated. First tests using such data models and limits to homogeneous patterns indicate that computed solutions were on a par with solutions generated by human packers in such cases.

Again inspired by human packing habits, the parallel packing of trolley layers is another field of further research. As described, the objective function (minimization of the overall height of all trolleys) is already designed globally but the algorithm works in layers. Using parallel packing one may assume better results, but with respect to complexity and the corresponding runtime of the algorithms we would like to observe further real life results of this approach first. The program based on the planning algorithms is also used in a real time context: In order to be able to calculate the associated shipping costs, the dispatcher needs to know how many trolleys are necessary to fulfill an order, before the actual packing process is undertaken.

Most of the plants relevant in our context are potted in round pots. However, there are exceptions that will require further extensions or new implementations of the packing algorithm to enable their handling as well. A subset of this problem is the consideration of trays, which have a rectangular shape and can hold a fixed number of plants. Current publications mainly focuses on the packing of either rectangles or circles into a larger rectangle (see Moffit and Pollack [16] for packing rectangles and Huang et al. [13] for packing circles). The authors know of no publication addressing an integrated approach for both, circles and rectangles. So far we assume that the dimensions of the packed plants are static and inflexible. This is a simplification and in fact not true for live plants as their dimensions change during the season. Further research in this field could start with the work presented by Albrecht et al. [8] and then try to evaluate whether a more realistic model (including dynamic plant dimensions) would indeed improve quality of the computed instructions.

8.0 REFERENCES

1. Dyckhoff, H.: A typology of cutting and packing problems. *European Journal of Operational Research (EJOR),* 1990, 44, 145–159.
2. Wäscher, G.; Haußner, H.; Schumann, H.: An improved typology of cutting and packing problems. *European Journal of Operational Research (EJOR)*, 2007, 183, 1109–1130.
3. Keng, N. P.; Yun, D. Y.; Rossi, M.: Interaction sensitive planning system for jop-shop scheduling. In *Expert Systems and Intelligent Manufacturing,* M. D. Oliff, ed.; Elsevier, 1988.
4. Sauer, J.: Knowledge-based systems in scheduling. In *Knowledge-Based Systems: Techniques and Applications,* C.T Leondes, ed.; Academic Press: San Diego, 2000; Vol. 4, pp. 1293–1325.
5. Nurmela, K. J.; Östergård, P. R. J.: Packing up to 50 equal circles in a square. *Discrete and Computational Geometry,* 1997, 18, 111–120.
6. Birgin, E. G.; Martínez, J. M.; Ronconi, D. P.: Optimizing the packing of cylinders into a rect-

angular container: A nonlinear approach. *European Journal of Operational Research (EJOR),* 2005, 160, 19–33.

7. Casado, L. G.; Szabó, P. G.; García, I.: Packing up to 100 equal circles in a square. In *Packing up to 100 Equal Circles in a Square;* Universidad de Almería, 1999.
8. Albrecht, A.; Cheung, S. K.; Hui, K. C.; Leung, K. S.; Wong, C. K.: Optimal Placements of Flexible Objects Part II: A Simulated Annealing Approach for the Bounded Case. *IEEE Transaction on Computers,* 1997, 46, 905–929.
9. Conway, J. H.; Sloane, N. J. A.; Bannai, E. *Sphere-packings, lattices, and groups,* Springer-Verlag: New York 1987.
10. George, J. A.; George, J. M.; Lamar, B. W.: Packing different sized circles into a rectangular container. *European Journal of Operational Research (EJOR),* 1995, 84, 693–712.
11. Schöning, U.; Toran, J.; Thierauf, T.; Messner, J.; Blubeck, U.: Three algorithms for packing variable size bobbins. Report: Abt. Theoretische Informatik Universität Ulm: Ulm, 2002.
12. Dowsland, K. A.; Gilbert, M.; Kendall, G.: A local search approach to a circle cutting problem arising in the motor cycle industry. *Journal of the Operational Research Society (JORS),* 2007, 58, 429–438.
13. Huang, W. Q.; Li, Y.; Akeb, H.; Li, C. M.: Greedy algorithms for packing unequal circles into a rectangular container. *Journal of the Operational Research Society (JORS),* 2005, 56, 539–548.
14. Correia, M. H.; Oliveira, J. F.; Ferreira, J. S.: Cylinder packing by simulated annealing. *Pesquisa Operacional,* 2000, 20, 269–286.
15. Akeb, H.; Li, Y.: A hybrid heuristic for packing unequal circles into a circular container. In *Proceedings of the 2006 International Conference on Service Systems and Service Management,* 2006; Vol. 2, pp. 922–927.
16. Moffitt, M. D.; Pollack, M. E.: Optimal rectangle packing: A meta-csp approach. In *Proceedings of the 16th International Conference on Automated Planning \& Scheduling (ICAPS-2006)*, 2006.

Evaluation of Polymer-clay Nanocomposite Barrier Coatings using Mixtures of Two Different Nanoparticles

SUNGWAN JEON[1], JINWOO KWAK[2] and BRUCE WELT[1,*]

[1]*Packaging Science Program, Agricultural and Biological Engineering, University of Florida, Gainesville, FL 32608*

[2]*Dept. of Material Science and Engineering, University of Florida, Gainesville, FL 32608*

ABSTRACT: Among a variety of barrier solutions in the packaging industry, polymer clay nanocomposites (PCN) coatings are attractive due to the potential to enhance barrier properties without modifying the base polymer. PCN barrier coatings require exfoliated microstructures of impermeable anisotropic fillers that can extend the tortuous path of permeating gas molecules. The fundamental expression for gas permeation was demonstrated with a variety of models based on Neilson's detour theory to design better barrier properties of PCN. Based on the physical detour path theory, a system using mixture of two particles with different aspect ratios was suggested to obtain higher barrier properties than with either particle alone by extending tortuous paths. The purpose of using two nanoparticle sizes was to attempt to achieve greater filler packing density and thus, greater tortuous path lengths resulting in greater barrier properties. In this study, a model of the proposed PC^2N system and an equation to estimate relative permeability (R_p) of PC^2N was developed. In order to investigate effects of PC^2N coating on barrier properties, PC^2N solutions were prepared and applied to substrates of known oxygen transmission rate. Significant barrier enhancement was not observed at constant overall nanoparticle volume fraction loadings regardless of particle mixtures used. As a result it was concluded that the final R_p of PC^2N was governed by total volume fraction of filler rather than particle type and size for the particles used. It was found that as volume fraction increased, particle orientation became less important. Surprisingly, it was found that coated films using mixtures of nanoparticles (PC^2N) offered superior optical clarity for a given level of barrier over coatings using just one particle type alone (PCN).

*Author to whom correspondence should be addressed.

1557-7244/10/01 041-22

1.0 INTRODUCTION

MULTI-LAYERED structures devised by the United States military for food packaging materials typical contain an aluminum foil layer to extend shelf-life of foods. However, foils are prone to pinholes which may cause some problems to achieve barrier functions. And the packaging materials containing a foil layer cannot be microwavable. Therefore, high-barrier material alternative to foil layers has to be considered.

Polymer clay nanocomposites (PCN) are attractive in packaging applications due primarily to enhanced barrier properties [1]. Improvements to barrier properties with low concentrations of nano-scale clay particles have been reported by many research groups [2,3]. Several models have been suggested to predict gas barrier properties in polymers and PCN materials. According to Nielsen's model [4], barrier properties of PCN can be changed by the microstructure of particle-polymer matrix as well as the aspect ratio of particles. Nielsen proposed a detour path model that the barrier property of a material can be adjusted by varying detour path of gas molecules. Other barrier models based on Nielsen's model have been proposed to suggest a rational barrier design of microstructure of composite materials by estimating the relative permeabilities (R_p) of them.

Three particle-polymer microstructures are generally recognized that are related to degree of dispersion of particles in polymer. These are referred to as non-intercalated, intercalated and exfoliated and may be loosely interpreted as poorly dispersed with irregularly spaced aggregates, dispersed with regularly spaced aggregates, and completely dispersed particles, respectively. Exfoliated structures provide superior gas barrier properties when compared to the other two structures.

Incorporation of inorganic particles into polymers is difficult due to high temperatures, high viscosities and polarity incompatibilities between particles and polymer. Therefore, commercial PCN materials tend to involve proprietary processing technologies and command high prices. Kwak et al. [5] explored the possibility of developing an aqueous based PCN that could be applied as a coating or paint and then dried in place to achieve a high barrier coating on inexpensive, commodity base materials.

Development of candidate PCN coating solutions requires consideration of particle-polymer compatibility, particle dimensions (aspect ratio) and coating adhesion onto base polymers. Properties of PCN materials depend on particle properties including layer charge, water adsorption capacity, cation exchange capacity (CEC), aspect ratio, viscosity of the final composite system, and the arrangement of cation exchange ions in the intergallery region between negatively charged particle layers. Spacing between two clay platelets can be enlarged via cation exchange. Once the intergallery region is opened, exfoliation of clay platelets in the polymer matrix usually can be achieved with high shear processing [6].

For any coating to be effective, higher roughness for physical interlocking between coated layer and surface of substrates or functional groups to accommodate chemical effects are required[7]. Even if a PCN coating were used within a multilayer film structure, good adhesion would be required to achieve high barrier. Adhesion is enhanced through increased surface area and increased concentrations of polar functional groups on surfaces. Various techniques have been proposed to modify the surface of polymeric materials such as corona discharge [8], plasma [9,10] and chemical modification. Atmospheric plasma treatment is becoming popular due relative treatment uniformity and the ability to modify gas mixtures to suit applications. Plasma is created by applying energy to a gas to produce excited species and ions. Polymer film surfaces are activated by plasma treatment. Optimization of conditions for effective plasma treatment was done previously by Kwak et al. [11].

Therefore, the purpose of this work is to (1) apply a model to predict relative permeability, Rp, of nano-composite incorporating two different particles (Laponite JS and Cloisite NA^+—referred to as PC^2N), (2) prepare PC^2N solutions and apply to substrates of known oxygen transmission rate in order to experimentally measure R_p, and (3) produce multilayer barrier films using PC^2N applications that provide promise as a replacement for foil in high-barrier, multi-layer films.

DESIGN OF PC^2N MODEL

Preparation of an effective PCN requires consideration of the microstructure of clay platelets to achieve lower permeability. In general, well-exfoliated microstructures in polymer matricies promises lower permeability by expanding detour length of gas molecules. Typically, published research has shown one type of nano-particle is used to create PCNs with well exfoliated microstructures [12]. However, it is believed that a mixture of two particles with different sizes and aspect ratios may provide greater tortuous paths and therefore, greater barrier properties than PCNs produced from just one particle variety. Hence, PCN produced with two particle types is referred to as "PC²N." Figure 1 shows the concept of enhancing particle packing density using two types of nano-particles [5]. A structure as shown in Figure 1 should have the same effect as increasing total volume fraction of particles, which would enhance barrier properties of the PCN.

A model was developed to predict R_p of PCN consisting of two types of clay particles having different aspect ratios. The R_p of the PC²N can be calculated using the following Equation (1).

$$R_p = \frac{2H_1(1-\phi_T)}{\left(L_1 + L_2\left(\frac{\phi_2}{\nu_{eff\,2}}\right)\right) + 2H_1} \tag{1}$$

Where, L_1, L_2 and H_1 can be defined as lateral length including edge-to-edge distance, $(l_1 + b_1)$ and $(l_2 + b_2)$ of clay 1 and 2 and vertical length $(h_1 + w_1)$ occupied by clay 1 per unit volume, respectively [5]. Where, effective volume which is occupied space by one clay particle can be calculated using Equation (2) and (3).

$$\nu_{eff\,1} = (l_1 + b_1)^2 (h_1 + w_1) \tag{2}$$

$$\nu_{eff\,2} = (l_2 + b_2)^2 (h_2 + w_2) \tag{3}$$

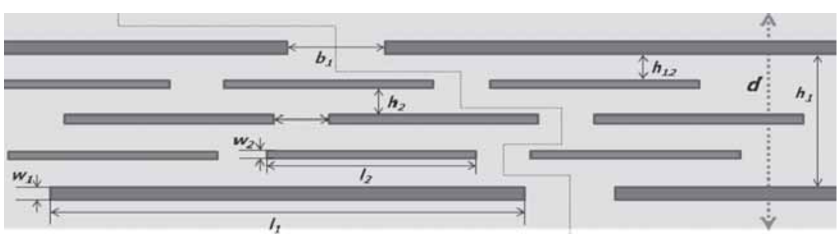


Figure 1. *Schematic diagram showing a filled polymer with two different clay types (blue rectangle represents clay 1 with the dimension of width of* w_1 *and length of* l_1 *and grey scaled rectangle is clay 2 with the dimension of width of* w_2 *and length of* l_2*. The separations of each particle are denoted as* b_1 *and* b_2 *for lateral edge-to-edge distance and* h_1 *and* h_2 *for face-to-face distance between the same clay type platelets. The face-to-face distance between two different type platelets is denoted as* h_{12}*. Note that the solid arrow line is the tortuous path for a gas molecule in PCN (P) and thick dot arrow line can be described as the shortest path (d) through unfilled polymer (*P_o*)).*

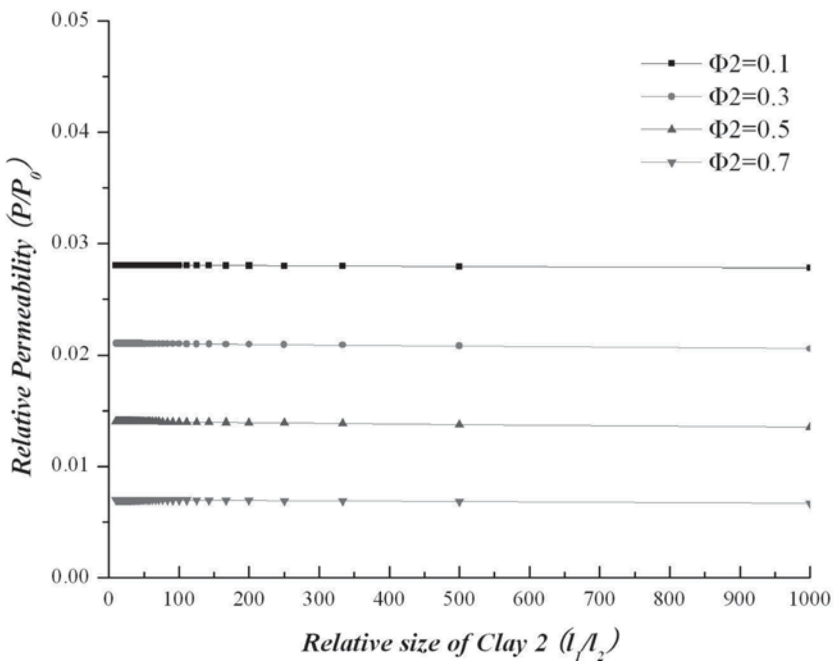


Figure 2. *Effects of relative size of clay 2 on relative permeability of PC²N: obtained by Equation 1 (l_1 = 100 nm, l_2 = 0.1 ~ 10 nm, $w_1 = w_2$ = 1 nm, b_1 = 10 nm, $b_2 = l_2/10$, h_1 = 1 nm, h_2 = 0.001 nm, ϕ_1 = 0.1).*

Based on this model for PC²N systems, barrier property prediction may be made.

Consideration of PC²N Barrier Property

Volume Fraction

When two different types of clays are dispersed in a polymer matrix, the size effect of the two particles has to be known. High barrier properties may be achieved by designing the maximum detour length in the microstructure of composite materials.

Effects of Relative Size of the Smaller Nanoparticle (clay2)

Figure 2 shows effects of relative size of clay 2 on R_p of PC²N. The relative size of clay 2 appears to offer no effect on decreasing permeability. Figure 2 predicts constant R_p values at each volume fraction of clay 2. Higher aspect ratio of clay 2 promises a longer tortuous path, but this requires a lower quantity of clay 2. This equation was made based on the assumption of effective volume. Once an occupied space of clay 2 is fixed by the effective volume of clay 1, the detour path of gas molecules always shows similar length regardless of aspect ratio, resulting in a compensating effect. At higher aspect ratio of clay 2, the quantity of these particles has to be diminished to be occupied the same effective volume created by clay 1. Therefore, total distance, d' in the system is similar for all aspect ratio of clay 2. However, as clay 2 volume fraction increases, R_p decreases due to increased number of platelets of clay 2 in the same effective volume created by clay 1 particles. From this relationship, it can be deduced that volume fraction of clay 2 is more a significant factor than size of platelets at fixed volume fraction of clay 1; this should result in a composite with better barrier properties.

Effects of d-Spacing of Larger Clay Particle (clay 1)

Another critical factor in the process of making PC^2N is the choice of alkyl ammonium ions. Ion exchange of cations (Na^{2+} or Ca^{2+}) in the intergallery region of natural clays by alkyl ammonium ions leads to expansion of this region as well as modification from hydrophilic to organophilic, thus enabling diffusion of polymer chains. The number of carbons in alkyl ammonium and the layer charge of the clay particle will determine the arrangement of alkyl ammonium chains in the intergallery region. Diffusion of polymer chains will be determined by shear forces and molecular weight of polymer [6].

Therefore, d-spacing between clay platelets vary depending on parameters that can be controlled during processing. In the PC^2N system, d-spacing of clay 1 affects R_p as shown in Figure 3. The larger the d-spacing of clay 1 at constant volume fractions of two clays, the higher the R_p value will be. This phenomenon may be caused by a lower number of clay 1 platelets per unit volume as increased d-spacing of clay 1 leads to more sparse distribution of clay 1, corresponding to decreased tortuosity. There would be no increase in added detour path, d_a, contributed by clay 2 particles in a fixed volume fraction of clay 2. Effects of increased volume fraction of clay 2 on R_p has the same tendency as the one shown in Figure 2. Detour length increased by insertion of clay 2 into the intergallery region has constant value when the volume fraction of clay 2 is fixed. D-spacing can be increased by cation exchange and shear processing, but it is also possible to increase d-spacing by insertion of clay 2 between clay platelets 1.

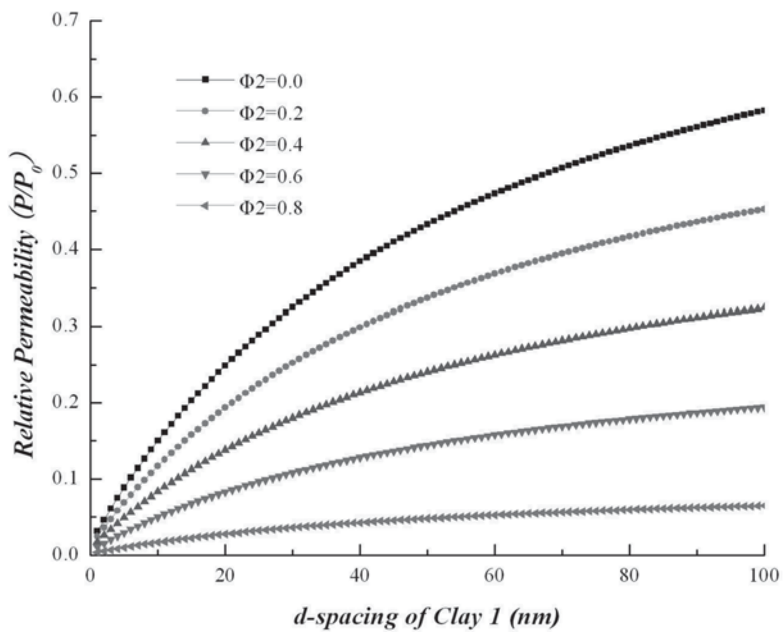


Figure 3. *Effects of d-spacing of clay 1 on relative permeability of PC²N: obtained by Equation 1 (l_1 = 100 nm, l_2 = 5 nm, $w_1 = w_2$ = 1 nm, $b_1 = l_1/10$, $b_2 = l_2/10$, h_1 = 1 ~ 100 nm, h_2=0.001 nm, ϕ_1 = 0.1).*

Effects of Arrangements of Smaller (clay 2) Particles

Expected R_p values are based on the assumption that there are no local changes in d-spacing of clay 2 (h_2) as d-spacing of clay 1 (h_1) generated during the insertion process can result in a deviation from the general barrier properties. However, the case that some increment of Δh can occur as shown in Figure 4, should be considered. This increase in d-spacing would occur by insertion of exfoliated clay 2 in the polymer matrix into the intergallery region with the aid of cation exchange reactions and shear processes. Thus h_1' is required to be substituted for h_1 to estimate R_p of this material and h_1' can be defined as shown in Equation (4).

$$h_1' = l(w_2 + h_2) - h_2 + 2h_{12} \quad (4)$$

Figure 5 illustrates how the arrangement of clay 2 particles affects d-spacing of clay 1 corresponding to its R_p. When a constant number of platelets of clay 2 at a fixed volume fraction are dispersed in the intergallery of clay 1, more layers are formed, therefore reducing the number of clay platelets in each layer. This means that although d-spac-

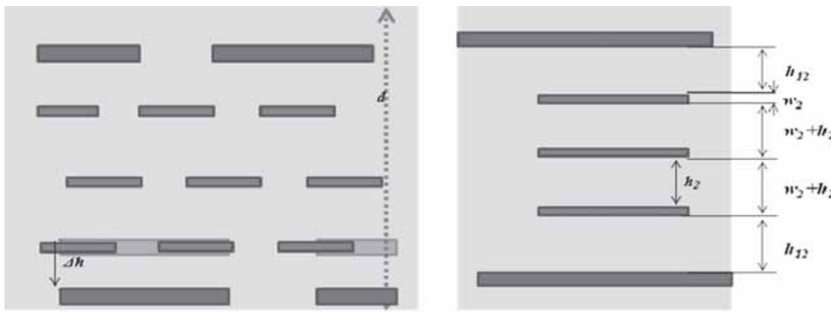


Figure 4. *Schematic diagram showing effects of changes in d-spacing.*

ing can be increased by creating more layers, the possibility that a diffusant encounters a platelet when passing by a layer would be lower. As a result, effects of increased tortuous path cannot exceed a critical R_p by compensation effects of d_a of clays as d-spacing of clay 1 increases. The initial dramatic decrease in R_p when h_1' approaches 0 suggest that d_a plays a key role by forming a densely packed arrangement in the reduc-

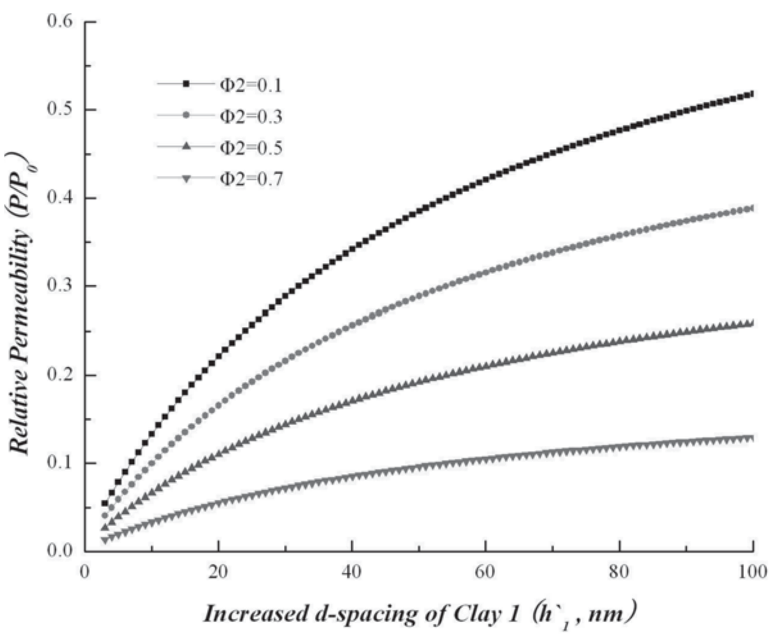


Figure 5. *Effects of increased d-spacing of clay 1 on relative permeability of PC²N obtained by Equation (1) (l_1 = 100 nm, l_2 = 5 nm, $w_1 = w_2$ = 1 nm, $b_1 = l_1/10$, $b_2 = l_2/10$, Φ_1 = 0.1).*

tion of permeability. Therefore, densification of platelets with higher volume fraction may be the ideal approach to achieve a superior barrier material. However, when the spacing between clay platelets including a lateral distance is not sufficient for a diffusant to bypass, the diffusant will detour the particle stack without any penetration between clay platelets, resulting in higher R_p. In other words, the state of aggregation of particles can have a significant effect on permeability, resulting in increased R_p at higher volume fraction. Therefore, an understanding of the aggregation is required for all types of nanocomposites materials since this not only affects barrier properties, but also other physical properties. In this work, the state of aggregation of clay 1 particles is investigated by varying the width of clay 1 from 0 to 1000 nm in Equation (1).

Effects of State of Aggregation on Relative Permeability (R_p*) of PC*^{2}*N system*

As shown in Figure 6, a higher degree of aggregation results in higher R_p because aggregates of particles that are not separated from the stacked agglomerate lower N_1, shorten the detour length and even reduce the

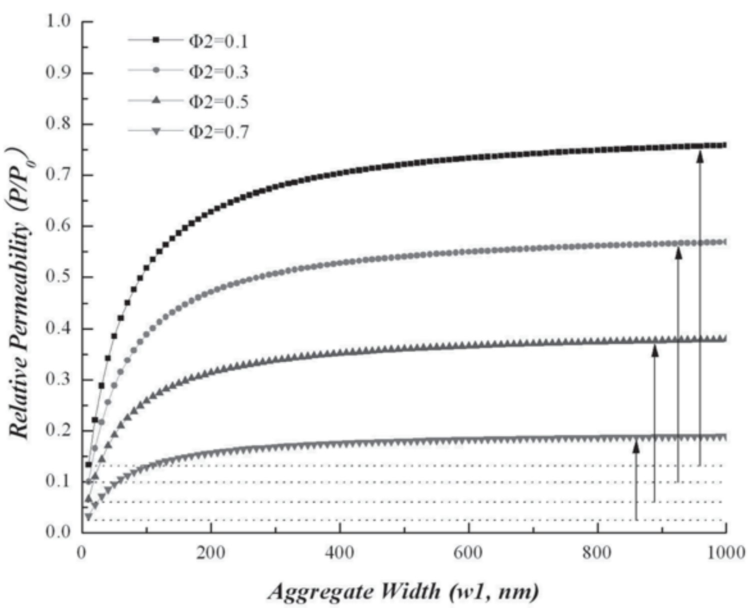


Figure 6. *Effects of the state of aggregation of clay 1 on relative permeability of PC*^{2}*N: obtained by Equation (1) (*$l_1 = 100$ *nm,* $l_2 = 5$ *nm,* $w_2 = 1$ *nm,* $b_1 = l_1/10$*,* $b_2 = l_2/10$*,* $h_1 = 1$ *nm,* $h_2 = 0.001$ *nm,* $\Phi_1 = 0.1$*).*

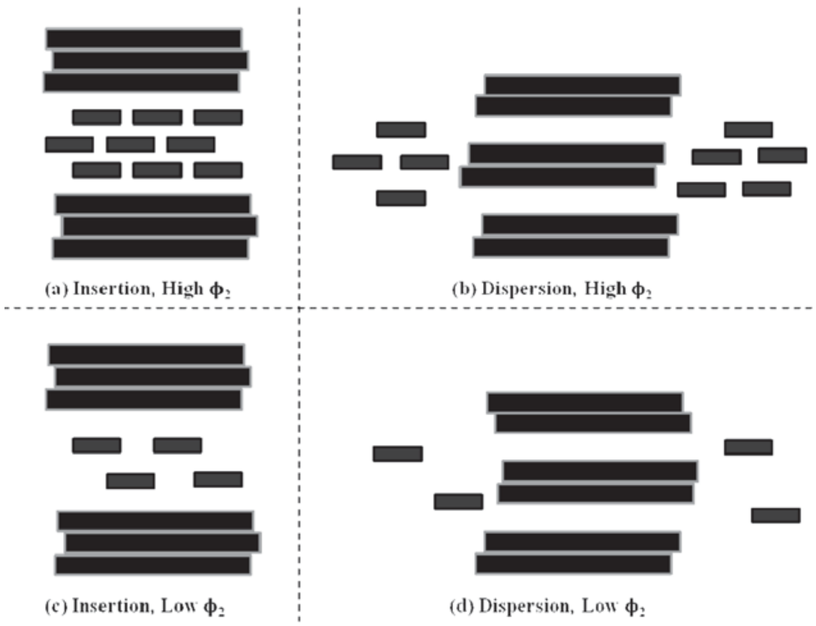


Figure 7. *Arrangement of clay 2 particles in PC^{2}N system: (a) Insertion of high volume fraction of clay 2 particles into intergallery regions of clay 1, (b) Dispersion of high volume fraction of clay 2 in a polymer matrix, (c) Insertion of a low volume fraction and (d) Dispersion of a low volume fraction.*

possibility of successful insertion of clay 2. This figure also shows that as volume fraction of clay 2 increases, the gap between R_p of initial point at w_1 of 1 nm and one obtained at w_1 of 1000 nm is decreased. Increased volume fraction of clay 2 will result in increased d_a due to insertion or dispersion in polymer matrices comparing to lower volume fraction as shown Figure 7. If the assumption that d-spacing of clay 1 is determined only by a cation exchange reaction and shear forces, R_p will depend on d-spacing of clay 1 at fixed ratio of clays. This dependence is shown in Figure 8. As the volume fraction of clay 1 increases, R_p of all d-spacing conditions will converge to 0 at the volume fraction of 0.9. The dense arrangement of clay 1 results in lower R_p and therefore, there would be no change in the effect of d_a caused by clay 2 once its volume fraction is set.

Orientation

Previous studies on barrier properties based on tortuosity or diffusion detour theory have focused on the aspect ratio of clay platelets [13,14] and volume fraction of filler [3,15,16]. Existing barrier models are based on the one common assumption that all clay platelets are dispersed in an orientation that is normal to the diffusant. However, the 3-dimensional displacement of clay platelets in the actual exfoliated PC²N system will cause some deviation from theoretical permeability. Bharadwaj reported the effect of orientation of clay platelets on R_p of PCN [15]. Based on this work, effects of orientation of clay platelets on R_p of PC²N can be estimated as shown in Figure 9. This figure shows that low relative permeability (R_p) is achieved when orientation parameter (s) approaches 1. Low R_p can be obtained by increasing aspect ratio of clay platelets as described in Figure 9.

Bharadwaj et al. [15] developed the following equation that considers orientation in predicting R_p.

$$\frac{P}{P_0} = \frac{1-\phi}{1+\frac{L}{2W}\phi\left(\frac{2}{3}\right)\left(S+\frac{1}{2}\right)} \tag{5}$$

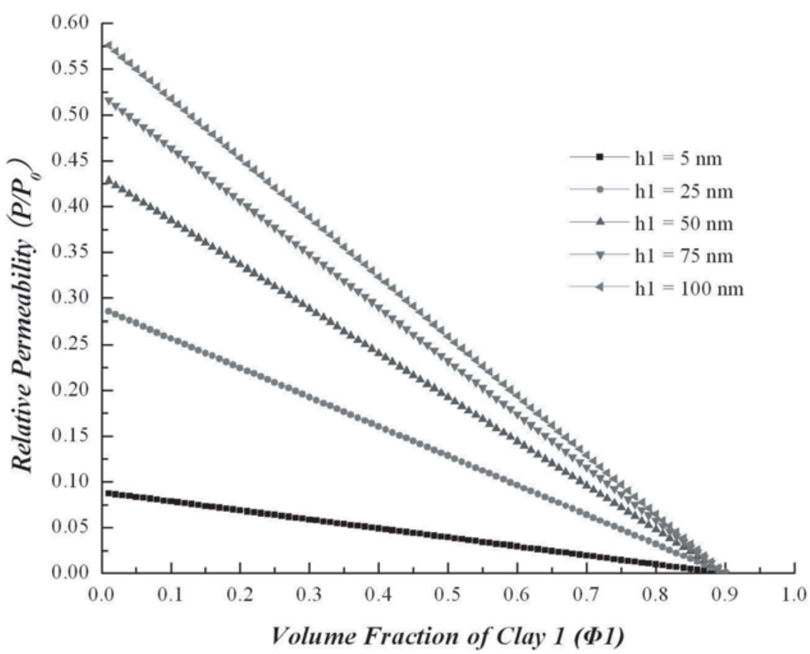


Figure 8. *Effects of the volume fraction of clay 1 on relative permeability of PC²N obtained by Equation (1) (l_1 = 100 nm, l_2 = 5 nm, $w_1 = w_2$ = 1 nm, $b_1 = l_1/10$, $b_2 = l_2/10$, h_2 = 0.001 nm, Φ_2 = 0.1, Φ_2 = 0.01 ~ 1).*

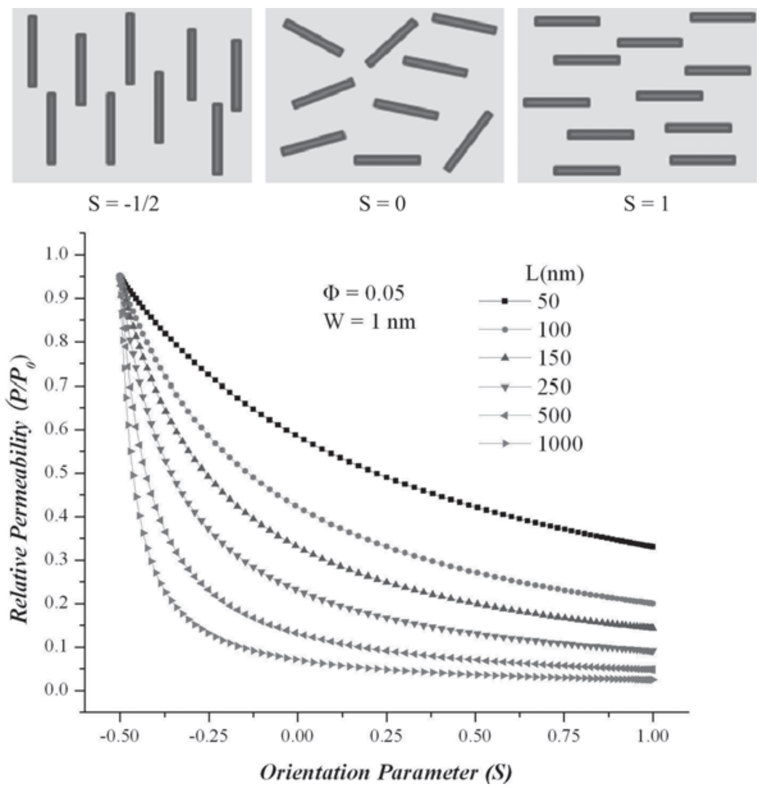


Figure 9. *Effects of particle orientation on relative permeability in exfoliated nanocomposites.*

Equation 6 was derived from Equation (5), for use with PC^2N.

$$\frac{P}{P_0} = \frac{1-(\phi_L - \phi_C)}{1+\left(\frac{L_1}{2W_1}\phi_1 + \frac{L_2}{2W_2}\phi_2\right)\left(\frac{2}{3}\right)\left(S+\frac{1}{2}\right)} \tag{6}$$

Figures 10, 11, and 12 are plotted by Equations (5) and (6). All R_p values were calculated using actual size data of Cloisite Na^+, and Laponite JS. Both clays have similar tendency that R_p is increased as orientation angle (θ), which represents the angle between the direction of preferred orientation and the sheet normal unit vectors increases. However, Cloisite Na^+ has slightly lower R_p due to its larger aspect ratio.

Effects of orientation of polymer chains on barrier properties of a polymer was examined by Somlai et al. [17]. The relationship between

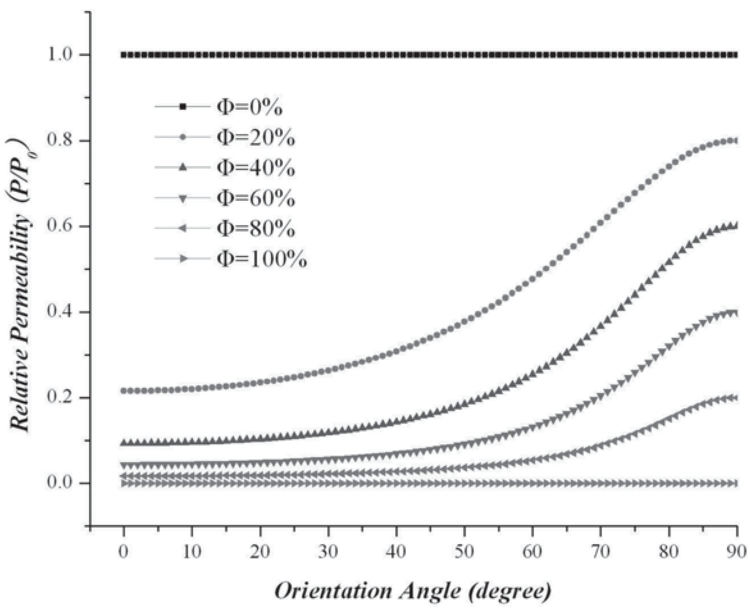


Figure 10. *Effects of orientation angle (degree) and volume fraction (Φ) on relative permeability of Laponite JS.*

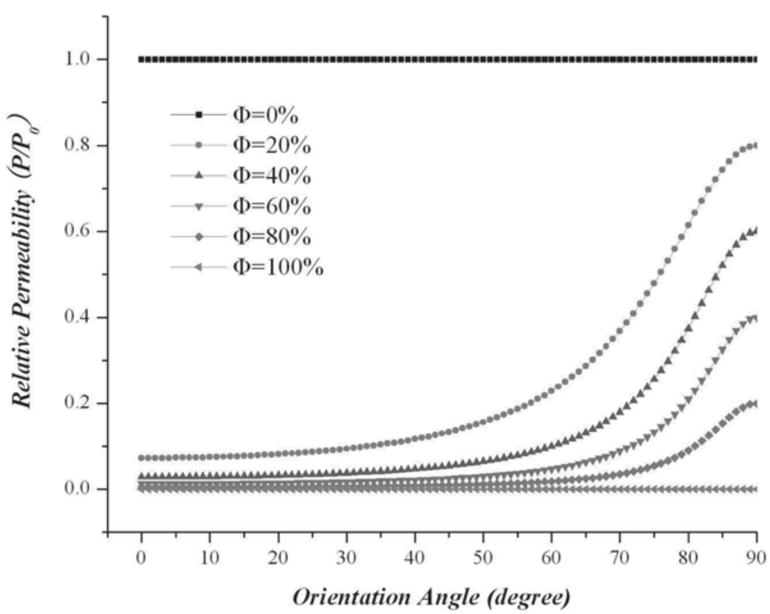


Figure 11. *Effects of orientation angle (degree) and volume fraction (Φ) on relative permeability of Cloisite* Na^+*.*

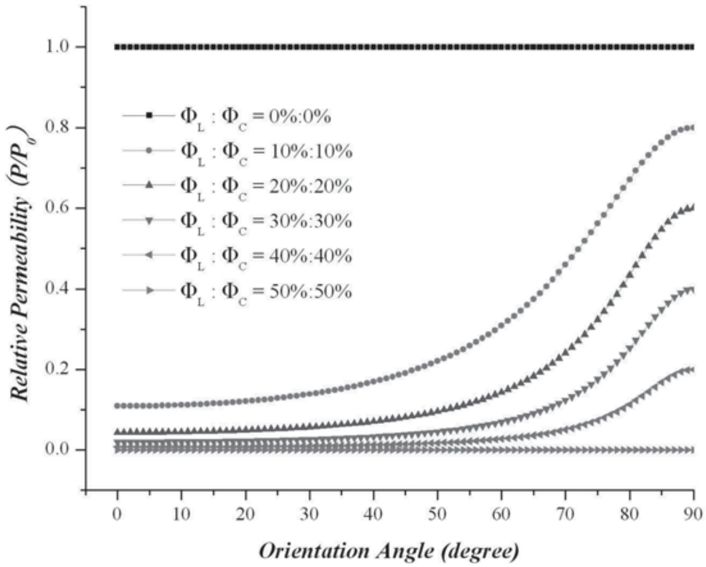


Figure 12. *Effects of orientation angle (degree) and volume fraction (Φ) on relative permeability of mixture composed of Laponite JS and Cloisite Na^+.*

orientation of polymer chains and permeability is not well reported in their work.

It is well known that processes that oriented polymer films tend to enhance strength and barrier properties. It is believed that greater molecular alignment causes an increase in crystallinity and therefore, barrier properties. When orientation is performed on polymer films with clay nanoparticles it is likely that the particle alignment is enhanced as well. In this study, an assumption that all clay platelets are aligned perpendicular to the diffusion direction of gas molecules was made to simplify the barrier equation of PC^2N. Therefore, the effects of other possicble varriables on barrier properties of PC^2N was concerned theoretically as suggested above and the barrier properties in the actual PC^2N coating system was investigated

MATERIALS AND METHODS

Materials

PCN and PC^2N solutions were made with a synthetic layered silicate, Laponite JS and a natural montmorillonites, Cloisite Na^+ (Southern Clay

Table 1. *Specific Information of Material used for PCN and PC^2N Composite Films.*

	Polymer matrix		Inorganic filler	
Box Style	LMW[1] PVOH		Laponite JS	Cloisite Na^+
MW[2]	3,000	Density (g/cm^3)	0.95	2.86
Viscosity (CP)[3]	3~4 cp	Aspect ratio	25	75~150
		d-spacing[4] (nm)	0.001	1.17

[1]Low molecular weight;
[2]Number-average molecular weight;
[3]4% aqueous solution at 20°C;
[4]$d_{(001)}$

Products, Louisville, KY). PVOH was supplied by Scientific Polymer Product, Inc. The specific informations of materials used for the preparation of composite films are listed in Table 1.

Organic ammonium chloride (OAC), [2-(Acryloyloxy)ethyl]-trimethyl ammonium chloride was used for intergallery modification. A multi-layered film consisting of oriented polypropylene (OPP) and linear low-density polyethylene (LLDPE) (Master Packaging Inc., Tampa, FL) was used as a substrate for coating.

Preparation of PCN and PC^2N Solutions

Two PCN solutions containing Laponite JS or Cloisite Na^+ respectively were prepared using the same method. At first, OAC buffer solution which amount is compare to 1 wt% of clay was added into a corresponding amount of deionized water at 40°C and pH 10 were premixed for 2–3 minutes with a magnetic stirrer. Each clay powder was added each beaker gradually to prevent aggregation. Prepared clay solutions were placed at room temperature for about 1 day to allow for cation-exchange in order to increase inter-particle distances. 8 wt% polymer stock solutions were prepared by dissolving PVOH in deionized water at 80°C and stirring with magnetic stirrer for 6–8 hours. Prepared PVOH stock solution was added to clay solutions and mixed for 1 hour with a magnetic stirrer and then mixed with a high shear mixer (KadyMill-L, Kady® International, Scarborough, ME) for 10 minutes. Prepared PCN solutions were placed in a hood at ambient temperature for 3–10 hours depending on viscosity in order to allow entrained air bubbles to dissipate.

To prepare a PC^2N solution, 8 wt% polymer stock solution was prepared by dissolving PVOH in deionized water at 80°C and stirring with magnetic stirrer. Each clay solution was prepared in the same manner described in the preparation of PCN solution and placed at room temperature for the cation exchange reactions. Laponite JS solution was added to polymer stock solution with agitation. Vigorous agitation with a mechanical stirrer for 10 minutes was conducted to prepare an exfoliated Laponite JS in a PVOH matrix.

Prepared PCN solutions consisting of PVOH and Laponite JS are added to Cloisite Na^+ clay solutions. Further, strong shear force with a mechanical stirrer was required for the final insertion process. This process was done by inserting PVOH mixed with relatively smaller Laponite JS particles into the intergallery region of Cloisite Na^+. To investigate effects of the ratio between Cloisite Na^+ and Laponite JS in the PC^2N system on OTR (cc/m^2/day) values, 10:0, 7:3, 5:5, 3:7, and 0:10 of ratios were used.

Preparation of Coated Samples

Atmospheric pressure plasma (APP) treatments were performed using a Dyne-A-Myte VCP (Enercon Industries Co., Menomonee Falls, WI). Air and nitrogen gas (about 50/50) were used to create plasma. The ranges of flow rate of nitrogen gas, working distance, times of treatment, and the speed of conveyer belt are 10–25 (l/min), 5–25 (mm), 2–4 times, and 5cm/s-10cm/s respectively. Modified surfaces of substrate films were coated using coating rods with PCN solution. Coated samples were dried for 24 hours at 40°C in a vacuum oven to abstract micro bubbles.

OTR Measurements

Oxygen transmission rates of substrate, PVOH, PCN, and PC2N samples were measured at 0 RH% and 23°C in accordance with the procedure described in ASTM D3985-05[46] and ASTM F1927-98[47] using a Model OX-TRAN 2/20MH (Mocon Corporation, Minneapolis, MN). Permeation cell area was 50 cm^2.

Results and Discussions

Figure 13 shows effects of coating layers on OTR. OTR values of PVOH (P), PVOH with Laponite JS (PL), PVOH with Cloisite Na^+ and

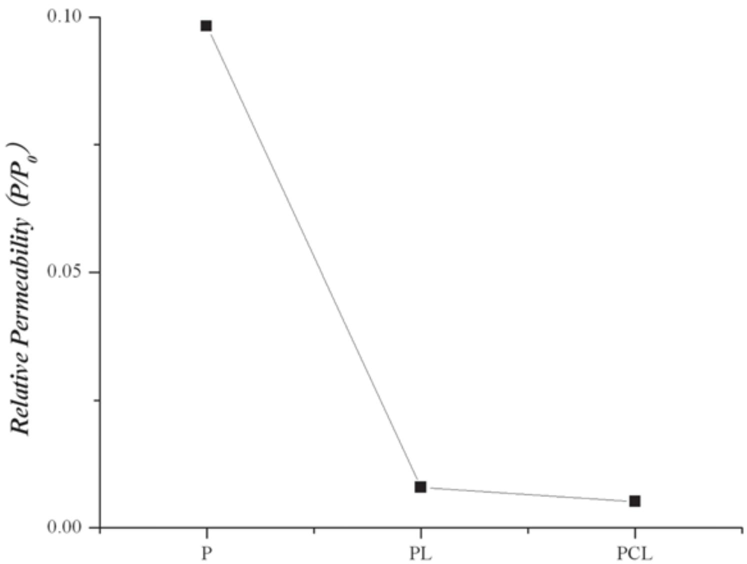


Figure 13. *Relative Permeability of Samples; P (8 wt% of PVOH), PL (8 wt% of PVOH and Laponite JS), PCL (8 wt% of PVOH, 4 wt% of Cloisite Na^+ and Laponite JS).*

Laponite JS (PCL) coated samples were measured. OTR of PVOH-coated sample has about 10% of OPP/LLDPE substrate film.

Generally, increasing crystallinity of homo-polymers tends to decrease gas diffusion rates and therefore OTR as well. Practical limits to degree of crystallinity limit this benefit. Addition of nano particles simulates additional crystallinity, thus reducing OTR beyond what is possible by the polymer alone (Figure 13). In this work, PCL-coated sample showed the lowest OTR, with PL-coated samples showing slightly higher OTR than PCL-coated samples.

These results showed that a mixture of different sized particles (PCL-coated samples) offered only a slight improvement over one type of particle (PL-coated samples). This is likely due to the fixed filler volume of 8% filler by weight used to prepare all samples. Perhaps a greater benefit could be seen if particle packing density is shown to increase with the mixture of particles.

Effects of different aspect ratios in PC^2N systems on OTR are identified by varying the ratio of Cloisite Na^+ to Laponite JS (Figure 14). There was no significant difference in OTR values based on weight percent (wt %). However, when the volume of Laponite is compared to that of Cloisite in terms of their densities, Laponite can occupy more space in the system than Cloisite for the same weight of clay used. This is contrary to results predicted from the model.

Figure 15 shows that lower OTR resulted from increasing clay weight percent. This tendency was confirmed by our previous work that examined Laponite JS as shown in Figure 16 [11]. Figure 16 shows OTR as a function of clay concentration loaded in the final dried PL film. The lowest OTR value was shown at 50 wt% of clay in the final dried film and, in the range of 20 to 40 wt% of clay concentration, there was no significant difference in OTR values. This plot also shows a dramatic increase in OTR values starting at 50 wt% of clay concentration which was likely due to particle aggregation and film stability as clay concentration increased.

In this study, enhanced barrier properties were shown to follow increased total effective volume of filler particles that were exfoliated in a PC^2N system. Also increasing total volume fraction of filler within a practical range is a relatively simple way to improve barrier property by about one order of magnitude for relatively high transmitting films such as OPP/LLDPE. Although PC^2N system did not show significant differ-

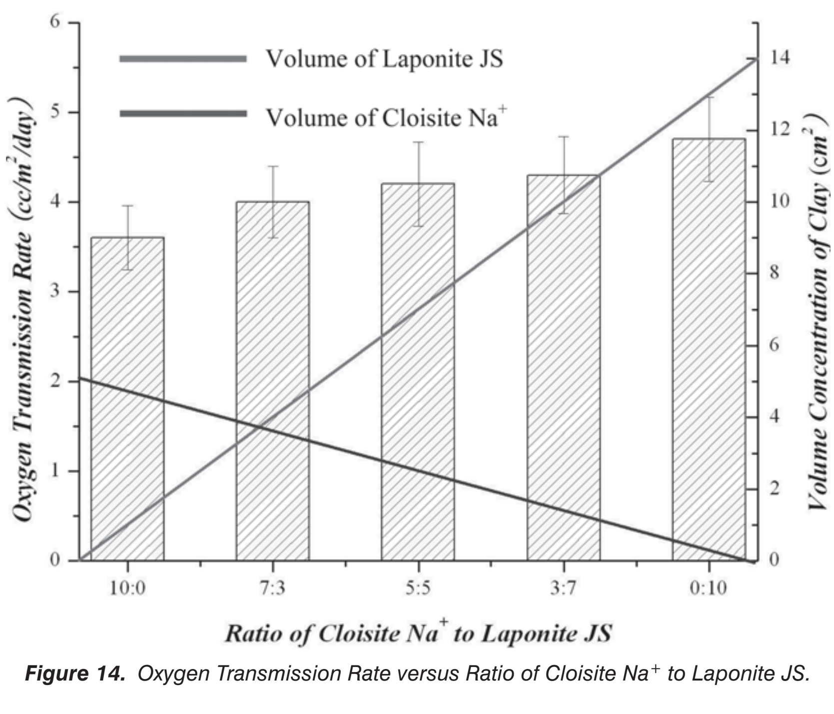


Figure 14. Oxygen Transmission Rate versus Ratio of Cloisite Na⁺ to Laponite JS.

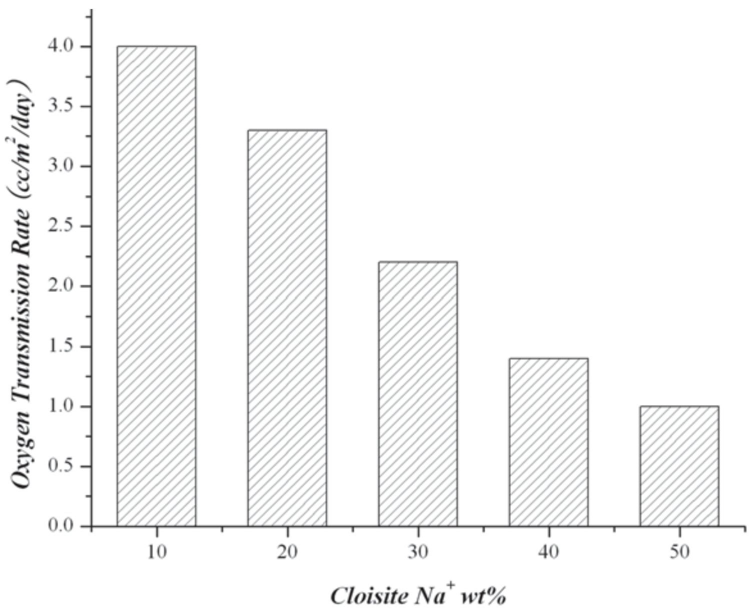


Figure 15. *Oxygen Transmission Rate vs. PVOH with Cloisite Na^+ film according to Cloisite Na^+ wt%.*

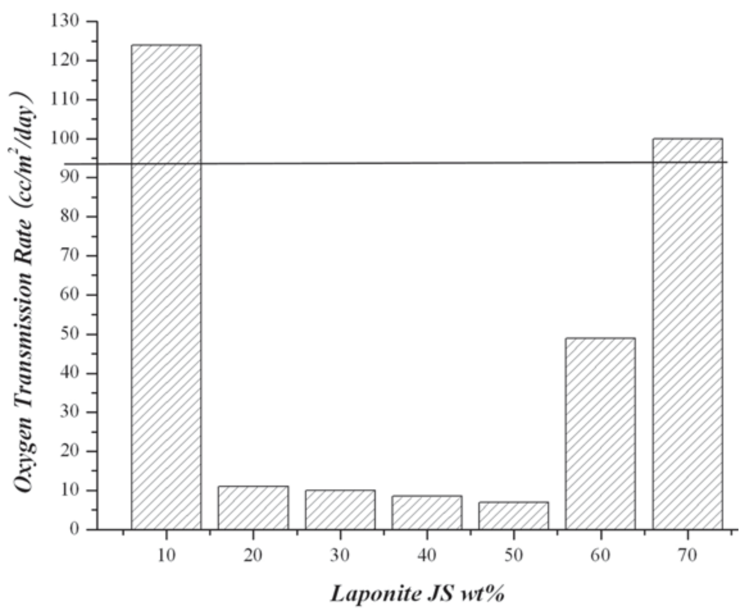


Figure 16. *Oxygen Transmission Rate vs. PVOH with Laponite JS film according to Laponite JS wt%.*

ences in barrier properties, PC²N composite films (mixture of particles) appeared to offer better optical clarity than Poly Vinyl Alcohol / Cloisite Na^+ (PC) films with comparable OTR.

CONCLUSIONS

Initially, it was believed that a mixture of two particles with different aspect ratios may provide greater packing densities, longer tortuous paths and therefore, enhanced barrier properties than PCNs produced from just one particle variety. To investigate the effect of using a mixture of two particle types on barrier properties quantitatively, a model was developed. Several expectations were suggested based on the model. First, the relative size of the smaller particle (clay 2) has no effect. Second, the larger the d-spacing of the larger particle (clay 1) at constant volume fractions of both clays, the higher the R_p value is obtained. Third, when a constant number of platelets of the smaller particle (clay 2) at a fixed volume fraction are dispersed in increased intergallery of clay 1, more layers are formed, therefore reducing the number of clay platelets in each layer. Therefore, although d-spacing can be increased by creating more layers, the possibility that a diffusant encounters a platelet when passing by a layer is decreased. As a result, effects of increased tortuous path cannot exceed a critical R_p. Forth, a higher degree of aggregation results in higher R_p because aggregates of particles that are not separated from the stacked agglomerate tend to lower the number of effective particles (N_1), shorten the detour length and even reduce the possibility of successful insertion of clay 2. Finally, R_p of PC^2N was governed by total volume fraction rather than by the specific relative ratios between clay 1 and 2. The variance in R_p with degree of particle orientation was explored. Most models are based on the assumption that all clay platelets are dispersed in an orientation that is normal to the diffusant. The discrepancy from theoretical barrier models can have significant effect on R_p, especially for the PCN materials containing the filler with higher aspect ratio. To estimate the effect of the orientation of platelets on R_p, the orientation parameter (s) and related R_p equation reported by Bharadwaj et al. [15], were used for PC^2N system and it was found that as particle volume fraction increased, importance of platelet orientation decreased.

Initially, effects of different coating layers (P, PL, and PCL) on OTR showed that addition of nano particles reduces OTR beyond what is possible by the homopolymer. Secondly, even though no significant difference in OTR values among all ratios for mixing two clays was obtained, the similar tendency that the clay having higher aspect ratio result in slightly lower OTR values was discovered. In conclusion, enhanced barrier properties were shown to follow increased total effective volume of filler particles that are exfoliated in polymer matrix and this tendency confirmed theoretical expectations for PC^2N. Although PC^2N system did not show significant enhancement in barrier properties, PC^2N composite films with a moderate barrier property appeared to offer greater optical clarity than comparable PCN with just one, larger nanoparticle.

ACKNOWLEDGEMENTS

This work was funded by the US Army Natick Soldier RD&E Center—Combat Feeding Directorate, Advanced Materials Engineering Team.

REFERENCES

1. Sudip R, Siew Y. Q, Allan E, Xiao D. C. The Potential Use of Polymer-Clay Nanocomposites in Food Packaging, *International Journal of Food Engineering* 2006;2:1–11.
2. Maged A. O, Vikas M, Massimo M, Ulrich W. S. Polyurethane Adhesive Nanocomposites as Gas Permeation Barrier. J. Macromolecules 2003;36:9851–9858.
3. Jaime C. Grunlan, Ani Grigorian, Charles B. Hamilton, Ali R. Mehrabi. Effect of clay concentration on the oxygen permeability and optical properties of a modified poly(vinyl alcohol). *J Appl Polym Sci* 2004;93:1102–1109.
4. Nielsen LE. Models for the Permeability of Filled Polymer Systems. *J Macromol Sci* (Chem) 1967;A1:929–942.
5. Jinwoo K, Sungwan J, Bruce W, Charles B, Siobhan M. Barrier Property Estimation of Polymeric Clay Nanocomposite System Containing Two Particle types. Submitted 2009.
6. Powell Clois E. Process for treating smectite clays to facilitate exfoliation. Patent, 2001;US 6,271,298,B1.
7. Ferencz S. Denes, Sorin Manolache. Macromolecular plasma-chemistry: an emerging field of polymer science, *Prog. Polym. Sci.* 2004;29:815–885.
8. Sepeur S, Kunze N, Werner B, Schmidt H, UV curable hard coatings on plastics. *Thin Solid Films* 1999;351:216–219.
9. Kwang Soo K, Chang Mo R, Chan Sup P, Gil Soo S, Chan Eon P. Investigation of crystallinity effects on the surface of oxygen plasma treated low density polyethylene using X-ray photoelectron spectroscopy. *Polymer* 2003;44:6287–6295.
10. Claire T, Christelle T, Pascal T, Jean D, Philippe L. Atmospheric pressure plasmas: A review. *Spectrochimica Acta Part B* 2006;61:2–30.
11. Jinwoo K, Bruce W, Charles B. Enhancement of Oxygen Barrier with Polymer Clay Nanocomposite Coatings on Polypropylene Treated with Atmospheric Pressure Plasma. *J Appl Packag Res* 2009;3:39–56.
12. Bo Xu, Q.Z, Yihu S, Yonggang S. Calculating barrier properties of polymer/clay nanocomposites: Effects of clay layers. *Polymer* 2006;47:2904–2910.

13. Tie Lan and Thomas J. Pinnavaia. Clay-Reinforced Epoxy Nanocomposites. *Chem. Mater.* 1994;6:2216–2219.
14. Kazuhisa Y, Arimitsu U, Akane O. Synthesis and Properties of Polyimide-Clay Hybrid Films. *J Polym Sci A: Polym Chem* 1997;35:2289–2294.
15. Bharadwaj K. Modeling the Barrier Properties of Polymer-Layered Silicate Nanocomposites. *Macromolecules* 2001;34:9189–9192.
16. Maged A. Osman,Vikas Mittal, Massimo Morbidelli, Ulrich W. Suter. Polyurethane Adhesive Nanocomposites as Gas Permeation Barrier. *Macromolecules* 2003;36:9851–9858.
17. Somlai L.S, Liu R.Y.F, Landoll L.M, Hiltner A, Baer E. Effect of Orientation on the Free Volume and Oxygen Transport of a Polypropylene Copolymer. *J. Polym. Sci. Part B: Polym. Phys.* 2005;43:1230–1243.

GUIDE TO AUTHORS

1. Manuscripts shall be sent electronically to the editor, Dr. Jay Singh at singhjapr@gmail.com using Microsoft Word in an IBM/PC format. If electronic submission is not possible, three paper copies of double-spaced manuscripts may be sent to Jay Singh, Industrial Technology, Cal Poly State University, San Luis Obispo, CA 93407. (Telephone 805-756-2129). Manuscripts should normally be limited to the space equivalent of 6,000 words. The editor may waive this requirement on special occasions. As a guideline, each page of a double-spaced manuscript contains about 300 words. Include on a separate title page the names, affiliations, and addresses of all the authors, and identify one author as the corresponding author. Because communication between the editor and the authors will be electronic, the email address of the corresponding author is required. Papers under review, accepted for publication, or published elsewhere in journals are normally not accepted for publication in the *Journal of Applied Packaging Research*. Papers published as proceedings of conferences are welcomed.
2. Article titles should be brief, followed by the author's name(s), affiliation, address, country, and postal code (zip) of author(s). Indicate to whom correspondence and proofs should be sent, including telephone and fax numbers and e-mail address.
3. Include a 250-word abstract and 5–6 keywords.
4. If electronic art files are not supplied, submit three copies of camera-ready drawings and glossy photographs. Drawings should be uniformly sized, if possible, and planned for 50% reduction. Art that is sent electronically should be saved in either a .tif or .JPEG format for superior reproduction. All illustrations of any kind must be numbered and mentioned in the text. Captions for illustrations should all be typed on a separate sheet(s) and should be understandable without reference to the text.
5. DEStech uses a numbered reference system consisting of two elements: a numbered list of all references and (in the text itself) numbers in brackets that correspond to the list. At the end of your article, please supply a numbered list of all references (books, journals, web sites etc.). References on the list should be in the form given below. In the text write the number in brackets corresponding to the reference on the list. Place the number in brackets inside the final period of the sentence cited by the reference. Here is an example [2].

   Journal: 1. Halpin, J. C., "article title", *J. Cellular Plastics,* Vol. 3, No. 2, 1997, pp. 432–435.
   Book: 2. Kececioglu, D. B. and F.-B. Sun. 2002. *Burn-In Testing: Its Quantification and Optimization,* Lancaster, PA: DEStech Publications, Inc.

6. Tables. Number consecutively and insert closest to where first mentioned in text or type on a numbered, separate page. Please use Arabic numerals and supply a heading. Column headings should be self explanatory and carry units. (See example at right.)
7. Units & Abbreviations. SI units should be used. English units or other equivalents should appear in parentheses if necessary.

Table 5. Comparison of state-of-the-art matrix resins with VPSP/BMI copolymers.

Resin System	Core Temp. (DSC peak)	Char Yield, %
Epoxy (MY720)	235	30
C379: H795 = 1.4	285	53

8. Symbols. A list of symbols used and their meanings should be included.
9. Page proofs. Authors will receive page proofs by E-mail. Proof pages will be in a .PDF file, which can be read by Acrobat Reader. Corrections on proof pages should be limited to the correction of errors. Authors should print out pages that require corrections and mark the corrections on the printed pages. Pages with corrections should be returned by FAX (717-509-6100) or mail to the publisher (DEStech Publications, Inc., 439 North Duke Street, Lancaster, PA 17602, USA). If authors cannot handle proofs in a .PDF file format, please notify the editor, Jay Singh at singhjapr@gmail.com.
10. Index terms. With proof pages authors will receive a form for listing key words that will appear in the index. Please fill out this form with index terms and return it to the publisher.
11. Copyright Information. All original journal articles are copyrighted in the name of DEStech Publications, Inc. All original articles accepted for publication must be accompanied by a signed copyright transfer agreement available from the journal editor. Previously copyrighted material used in an article can be published with the written permission of the copyright holder (see #14 below).
12. Headings. Your article should be structured with unnumbered headings. Normally two headings are used as follows:

    Main Subhead: DESIGN OF A MICROWAVE INSTALLATION
    Secondary Subhead: Principle of the Design Method

    If further subordination is required, please limit to no more than one (*Third Subhead*).
13. Equations. Number equations with Arabic numbers enclosed in parentheses at the right-hand margin. Type superscripts and subscripts clearly above or below the baseline, or mark them with a caret. Be sure that all symbols, letters, and numbers are distinguishable (e.g., "oh" or zero, one or lowercase "el," "vee" or Greek nu).
14. Permissions. The author of a paper is responsible for obtaining releases for the use of copyrighted figures, tables, or excerpts longer than 200 words used in his/her paper. Copyright releases are permissions to reprint previously copyrighted material. Releases must be obtained from the copyright holder, which is usually a publisher. Forms for copyright release will be sent by the editor to authors on request.

General: *The Journal of Applied Packaging Research* and DEStech Publications, Inc. are not responsible for the views expressed by individual contributors in articles published in the journal.