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Comparative Analysis of Aluminum and Aluminum Free Recycled Multilayered Beverage Carton Packaging

ABSTRACT

Packaging industry's raw material producers face continuous challenges of sustainable development. Besides raw material and energy consumption mitigation, essential functions of product protection must be maintained. Re-use and recycling of waste materials must be provided, which is required by the European Union directives as well. Multi-beverage carton recycling is even more difficult due to the use of various materials, due to their diverse properties and qualities. The introduced paper is part of a complex study, which aims to prove that utilizing dry-grinding technology and no additives, semi-finished products can be produced for the packaging industry.

KEY WORDS

multi-layered packaging, beverage cartons, dry-grinding, recycling, eco-design

Anita Magdolna Szabó[1],
László Koltai[2],
Lóránt Fodor[1]

[1]*Department of Machine and Product Design, Budapest University of Technology and Economics*
[2]*Department of Media Technology and Light Industry Engineering, Óbuda University*

Corresponding author:
Anita Magdolna Szabó
anitam.szabo@gmail.com

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Introduction

The first carton beverage packaging was patented in 1944 by Ruben Rausing, founder of Swedish company Tetra Pak. This tetrahedron-shaped, plastic-covered packaging from the 1950 was used for dairy products. Aluminized aseptic packaging in the mid '60s allowed longer shelf life (Leander, 1996; Sedig, 2002; Crattenand, 2011). Today, in addition to the storage of dairy products, other liquid food types are packaged into similar, multi-layered cartons.

According to the literature, composition of 1 liter beverage cartons, aseptic boxes containing aluminum: box weight is 28 g (75% paper, 20% PE, 5% Al). In case of aluminum-free boxes: box weight is 29 g (91% paper, 9% PE) (European parliament and council directive, 1994).

Different layers have different functions. Polyethylene (PE) separates food from the other layers, and protects paper layers from moisture, keeping its strength. The cardboard also protects the product from sunlight and aluminum (Al) creates an oxygen barrier (Baka, 2013). This multi-layer laminated packaging material is produced by lamination and coating processes. During lamination a thin layer of glue is used to combine the layers, while during coating, a liquid phase solvent or solvent-free coating is applied onto the matrix that forms a thin film surface on the surface after drying (Simon, 2012).

In the European Union approximately 100 million liter liquid or semi-liquid foods and beverages are packaged in beverage cartons. This sums up as 977.000 tons of combined packaging and recycling. This, beyond developments, gives constant challenge for manufacturers. In the European Union the rate of recovery in beverage

carton recycling has doubled in the last ten years. Return rates in Hungary, 19% as of now, are improving. Still they are below the European average of 39%. Recycling rate is greater than the average in Germany (65%), Belgium (62%) and Austria (42%) (Ace, 2011a; 2011b).

Basic separation demands high energy and technology background, so in many cases high costs are involved. There are two ways for industrial processing of the selectively collected boxes. The most widespread method is the separation of components (Figure 1), the other, rarely used method is the grinding method without separation (Figure 2) (Italos Karton Környezetvédelmi Egyesülés, 2010; Tetrapak, 2013a).

Board-products of the dry grinding technology are utilized by the construction and furniture industries. These products are sold in many countries around the world under various brand names Tetrapak (2013b). For example, in Slovakia Tetra K1, K2, K3, in Germany Tectan (EVD, 2010), in Argentina T-Plak (Compostar, 2013; TPlack, na), in Brazil Reciplak (Recipak, na; Ecopack, 2012), in China Chiptec, in Kenya Lamiboard, in Pakistan Greenboard and in Turkey Yekpan.

Today, ensuring sustainable development is a fundamental requirement for European packaging industry. The 94/62/EC- No. 1994 of December 20, 2005/20/EC (9th March 2005) amended EU directive to packaging manufacturers is committed to the highest possible level of recycling of packaging waste generated (Ace, 2013b).

Aims of the research to prove that utilizing dry-grinding technology and without additives semi-finished products for the packaging industry can be produced in the multilayer beverage carton recycling.

Preparation for laboratory testing

Initially, 1L filling volume Tetra Pak of (aseptic) packaging material containing aluminum or non-aluminum was studied. Investigated samples were obtained commercially. After the product was removed and the package was cut up, surfactant and water-surface cleaning was applied. Before the measurement, samples were air conditioned for 36 hours in the laboratory, under conditions of 50 ± 2% relative humidity and 23 ± 1°C temperature. The equilibrium moisture content of the samples was at an average of 2.3%. The results of the laboratory tests we describe in the section 5.

Component determination

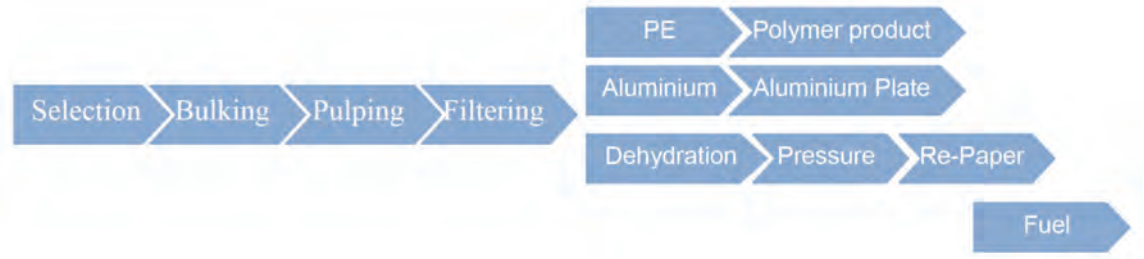
Tetra Pak boxes feedstock composition determination of a newly developed method was applied to determine the components of Tetra Pak boxes (Simon, 2012). The accuracy of the method corresponds to our research purposes, although a very small amount of PE can be recognized besides the separated cellulose (Figure 3).

Polyethylene (PE) is affecting our experiments, determining its amount by 99.55 v/v% toluene boiling with subsequent 50 v/v% ethanol and hot water wash.

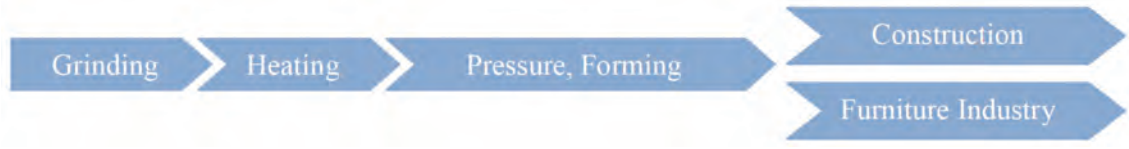
Grammage determination

Average grammage of the packaging material was examined according to the ISO 536:2012 standard (1) before further investigations. There were five parallel measurements of 100x100 mm surface test specimens, with analytical precision.

$$m_A = \frac{m}{A}\left[\frac{g}{m^2}\right] \qquad (1)$$



» **Figure 1:** *Recycling with separation of components*



» **Figure 2:** *Recycling without separation*

Where m_A - Grammage [g/m^2], m- mass of the specimen [g], A- surface area of the test specimen [m^2].

Thickness determination

Subsequently, test specimen thickness was determined using semi-automatic Lorentzen-Wettre type thickness measuring device, with an average of 10 measurements for each sample, according to the ISO 534:2011.

Apparent Sheet Density determination

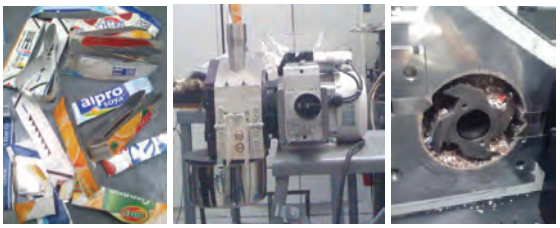
The apparent sheet density of packaging value was determined by calculation (2) these indicates compactness of the structure.

$$T = \frac{m_A}{s \cdot 1000}\left[\frac{g}{cm^3}\right] \qquad (2)$$

Where T- Apparent Sheet Density [g/cm^3] m_A- Grammage [g/m^2], s – thickness of the specimen[mm]

Grinding

Preparation for grinding, as described in "Preparation for laboratory testing" section, was that prepared cartons were cut into 20 mm wide stripes, then were cut up and grinded by a 4 knife FRITSCH Cutting Mill. The process is shown in Figure 3. Then resulting powder particle size distribution was examined.



» **Figure 3:** *FRITSCH Cutting Mills*

Fractioning

The laboratory fractionation shaker used for fractioning has defined hole size sieve series between Ø3.15 mm and Ø0.063 mm, with an eccentric drive and a timer. Each sieve has the remaining chips that can be read by 0.01 g accuracy. In five simultaneous measurements, a 60 seconds shaking timer, 100 g – 100 g samples per raw material were fractionated.

The structure of the grinding was examined (Figure 7) by Tuxen type BCT-type stereo microscope with video camera and external light. TS View was the image processing software used.

Test specimen manufacturing

After fractioning, test specimens were produced from the grinded materials for further analysis. The 1.5 mm thick test specimen was prepared using COLLIN P 200 E-type heat press. According to the literature upper and lower face of the press was adjusted to T = 180°C temperature. 120 mm x 120 mm x 1.5 mm test specimen plates were prepared. The 1.5 mm metal frame was filled up by the pulp and placed in the heated press. After closing of the press, and setting the temperature reheat to T0 = 180°C, pressure was increased from p0 = 0bar to p1 = 100 bar, for 5 minutes (t = 300 s). Then cooling the press heads down to T1 = 40°C, and lowering pressure to p2 = 0 bar, prepared specimen were removed.

» **Figure 4:** *P200 COLLIN this heat press*

Test specimen

At the preparatory stages of basic and mechanical tests, specimens were kept at 50±2% relative humidity on T = 23°C±1 temperature. After this experiments took place.

Optical structure experiments

Specimens were examined by Tuxen type BCT-type stereo microscope with video camera and external light. TS View was the image processing software used.

Grammage determination

The grammage value determined by calculation of the specimen as described in previous section.

Thickness measurement

Thickness of the test specimen was measured by Lorentzen Wettre digital thickness gauge at ten points according to ISO 534:2011.

Mechanical tests

Specimens were measured against resistance by mechanical stress experiments. Aluminum-free, and aluminium constituent beverage carton specimens, their material composition, the irregular distribution of their com-

ponents and on the basis of thickness deviation it was assumed that during further investigations against the individual test points, specimens will behave differently in response to the given mechanical load. We assumed that the measured data will provide comparable values.

The tensile strength, elongation, flexural rigidity and tear strength of specimens were determined during mechanical testing. Assuming that tensile strength of the aluminum-free specimen is going to be lower than the aluminum constituent ones.

Tensile test

Tensile test was carried out according to ISO 1924-1:0992 in the Frank-type pendulum system Schopper tensile machine, 50 mm clamping length. Test specimen size was 15 mm x 70 mm. Tensile strength of specimens was tested with ten-ten parallel measurements. Using the readings, tensile strength values determined by calculation of the specimens tensile strength, which is 1m wide sample relative tensile strength (3).

$$S_T = \frac{F_T}{b}\left[\frac{kN}{m}\right] \quad (3)$$

Where S_T- Tensile Strength (N/m), b- Clamping Length (m)

From the absolute elongated values, read from the device, knowing the clamping length average linear strains of the specimens were calculated (4).

$$\varepsilon = \frac{\Delta L}{L_0} \quad (4)$$

Where ΔL- the Difference Between the Original Length (L_0) and the Elongated Length (L1), L_0- Initial Length (mm).

Specimens with various material composition of are comparable, due to the Tensile index values. This is nothing more than tensile strength weighed by grammage. However, this requires the grammage values: mass of the heat-pressed specimen expressed in grams per square meter, defined in section “Mechanical tests” section. As a result tensile index was determined (5).

$$I_T = \frac{S_T}{m_A}\left[\frac{Nm}{g}\right] \quad (5)$$

Where I_T - Tensile index (Nm/g) S_T- Tensile strength (N/m), m_A – Grammage (g/m^2)

Tearing strength test

Tearing strength of the specimens is used to determine the Elmendorf machine according to ISO 1974:2012. Tear resistance, the mean force required to continue the tearing of an initial cut, was parallel measured on the ten test specimens. Before starting the test, the sector-shaped pendulum is secured in a suitable position for capturing the specimen, than 63x100 mm specimens were camped by the clamping jaws and the affixed knife cut 20 mm into the specimen. To be able to continue the further tearing process, pendulum anchorage was released and allowed to swing. Tear strength was readable on the scale of the device, thus tear resistance was determined by calculation (6).

$$S_{Tear} = \frac{F_{Tear} \cdot 16}{n} \cdot 9{,}81[mN] \quad (6)$$

Where: S_{Tear} = Tearing force (mN), F_{Tear} = is read from the scale value (p), n = Number of test specimens examined.

In the formula, the 9.81 multiplier is needed from read tear force to mN conversion. The tear strength depends on the grammage of the paper, so the tear strength of the different papers used for the comparison is known as tear index (7).

$$I_{Tear} = \frac{S_{Tear}}{m_A}\left[\frac{mNm^2}{g}\right] \quad (7)$$

Where I_{Tear} = Tear index (mNm^2/g), as a base for S_{Tear} = Tear strength (Nm) m_A = Grammage (g/m^2).

Bending stiffness test

The specimen stiffness was measured with the Lorentzen type of instrument according to ISO 5628:2012. The device measures the force required for the 15 degree bending of the specimen, in case of a 25 mm lever arm. The received data is in pond units that needed conversion and summary to mN. During the experiments, 38 x 70 mm sized specimens were used.

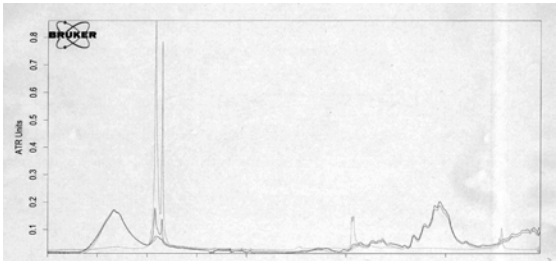
The two kind of specimens was tested with ten-ten parallel measurements. Using the readings, bending force values were determined by calculation of the bending stiffness (Koltai, 2013).

Results

Results of component determination

Applied to the component determination process described in “Component determination“ section, the results are presented in bone dry test sample. As results concluded, the PE content of the 1-liter aluminum-free Tetra Pak carton is at an average of 11.43%, while carton with aluminum is at 16.61%, with 5.61% aluminum content.

The polietilene was detectabled with Fourier transform infrared spectroscopy (FTIR). The result shows a Figure 5.



» **Figure 5:** *Control after toluene boiling with FT-IR equipment: there are detectable polyethylene traces*

Results of grammage determination

The Grammage was determined by calculation as described in "Grammage determination" section. These values are Tetra Pak aluminum-free m_A = 132g/m^2, Tetra Pak with aluminium m_A = 145 g/m^2.

Results of the thickness measurement

The thickness of test sample was determined as described in "Thickness determination" section. These values are Tetra Pak aluminum-fre s=466 µm, Tetra Pak with aluminum s= 444 µm.

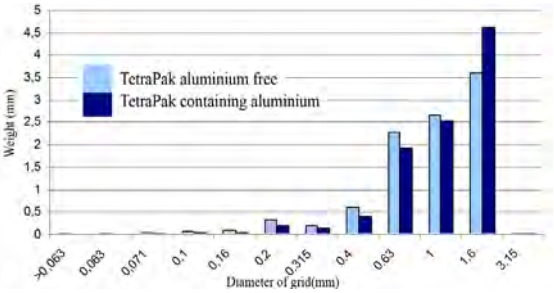
Results of apparent sheet density

The apparent sheet density was determined from thickness, surface area and weight values as described in "Apparent Sheet Density determination" section. These values are Tetra Pak aluminum 0.32 g/cm^3 and the aluminum-free sample was 0.28 g/cm^3.

Fractioning

After grinding- what we described in "Grinding" section - we performed fractionation as described in "Fractioning " section. It can be concluded that the largest fraction was between 1.6 mm and 3.15 mm range. There was fractionated chips of the aluminum-free sample of 4.61 g, which is 46% of the total fraction, while from the aluminum ones 3.61 g was that the total fraction of 36% into this range.

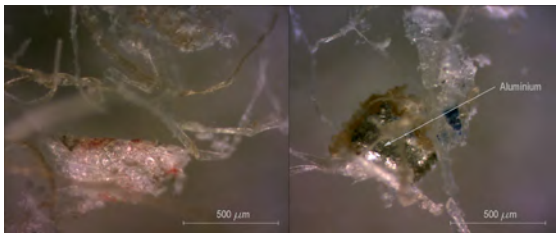
Based on the average of the measured values diagram, it is observed that although both show about the same size distribution of raw materials, the aluminum-containing granules of the Tetra Pak Ø 1.6 mm sieve had 1.00 grams, while the Ø 0.063 mm sieve had 0.37 grams more. For both aluminium constituent and aluminum-free Tetra Pak particle diameter was mostly in the range of 0.63 mm and 1.6 mm sieve diameter. The Ø1.6 mm sieve for aluminum-free particles had 36.1%, while aluminium constituent had 46.1% of the particles remained. In the Ø1.0 mm sieve, there were approximately the same amount, aluminum-free 26.7% and aluminum constituent 25.5%. The Ø0.63 mm sieve had aluminum-free 22.8%, and 19.4% of the aluminum constituent particles. On the 0.071 mm sieve there were typically scrabs of cellulose fiber, while on the Ø1.6mm one beverage carton pieces were found. The results shows Figure 6.



» **Figure 6:** *Grinding chip size distribution*

The structure of the grinding

Investigating structure of the grinding as described in "Fractioning" section, it can be clearly observed that the associated packaging material's most significant components are cellulose fibers that are greatly separated from each other, due to dry state grinding effect. Bundles were observed adhered to PE foil pieces, and the associated aluminum (Figure 7).

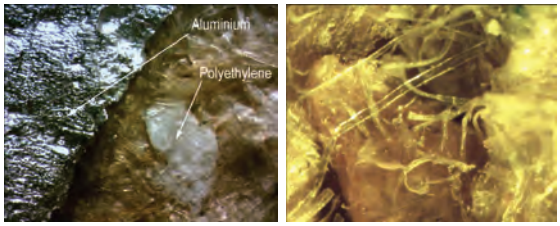


» **Figure 7:** *Below 0.315 mm particle microscopes, CS stereomicroscope TS view*

Results of test specimens

Optical structure examination results

The recordings made as described in "Optical structure experiments" section. On these pictures aluminum, molten polyethylene (PE) and cellulose fibers can be well recognized. (Figure 8)



» **Figure 8:** *Surface image, BTC stereo microscope TS view*

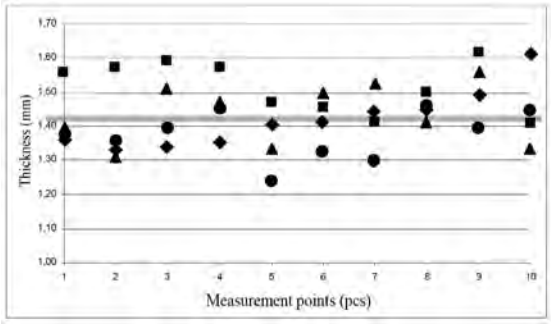
Despite the mechanical strength of the recordings, it can be concluded that the structure has a specific incoherence. Due to the three material components, specific cavity system can be observed on the surface of the sample. This structure shows the structure of fiber-reinforced composites, wherein the PE is matrix material, the reinforcing material in the cellulosic fiber and aluminum. The molten polyethylene (PE) is able to keep the structure of the sample. Problematic structural element is the aluminum (Al), as compression occurred below the melting point of aluminum so that, in areas where a greater surface area (2-3 mm) aluminum veil, the elements can connect to a lesser extent compared to areas where only cellulose and polyethylene (PE) is present.

Results of grammage determination

Grammage determination as defined in "Mechanical tests "section. These values for Tetra Pak aluminum-free m_A = 94g/m^2, Tetra Pak aluminium m_A = 128 g/m^2.

Results of tickness measurement

The thickness value of the specimen was read with the method described in "Thickness measurement" section. The average thickness is calculated from the measured values was s = 1.46 mm. The measured values compared to s = 1.46 mm average thickness, showed a 12% difference.



» **Figure 9:** *The average thickness of 1.5 mm thick frame made as the specimen*

This difference may be due to irregular material distribution that various components react differently for heating at T = 180°C. There was no detectable difference between the aluminum-free, and the aluminum-containing specimen thickness deviation (Figure 9)

Results of mechanical tests

Results of tensile test

We measured in the present experiment, with presented at "Tensile test " section instrument, and calculated using the formula number 3. These values are Tetra Pak aluminum-free S_T = 987N/m, Tetra Pak with aluminium S_T = 2396 N/m. The average elongation of hot-pressed specimens was determined with the reading values and the formula 4. These values are Tetra Pak aluminum-free specimen ε = 0.05, for the Tetra Pak aluminium specimen ε = 0.055.
The tensile strength index was calculated as formula 5. The values of grammage was using, what was presented in "Results of grammage determination" section. In the present study, tensile index of the analyzed specimens for Tetra Pak aluminum-free were I_T = 10.49 Nm/g and for Tetra Pak aluminium I_T = 18.7 Nm/g.

It can be concluded that there is almost 44% difference between the two specimens. The higher tensile index of the aluminum containing specimen is mainly due to the higher PE content, so 30% increase in the polyethylene ratio a 44% increase can be achieved in the value of Tensile index. The result is important because when lower PE content is associated with the packaging material, it can be processed using this technology, by addition of relatively little waste PE. There is significant difference between test specimen tensile indexes depending on their raw materials, even if they have the same grinding conditions, the same production temperature and the same thickness ratio. According to our prior assumption tensile index of the Tetra Pak test specimens containing aluminium than the aluminium-free ones.

Results of tearing strength

The test method described in "Tearing strength test" section and formula 5 was determined using the average tearing strength. These values of an aluminum-free Tetra Pak is S_{Tear} = 386.899 mN, while in case of aluminium Tetra Pak it is S_{Tear} = 596.454 mN. Then the tear index was calculated to using the formula 6. The tear index for aluminium-free Tetra Pak is I_{Tear}= 4.116 mNm^2/g, while the Tetra Pak containing aluminium is I_{Tear} = 4.660 mNm^2/g. The tear index of specimens containing aluminium was 11.67% higher than the aluminium-free test specimen.

Results Bending stiffness experiment

The test method described in "Bending stiffness test" section determined using the average force required to bending. We have found that the average bending strength is the following: for aluminum-free Tetra Pak F_S = 32.57 mN, while aluminium Tetra Pak F_S = 66.97 mN. This value is 51.37% higher in the case of the specimens containing aluminum, the stiffer structure is due the PE content.

Conclusion

Sustainable development, as EU requirement introduce it, takes the whole life cycle into account and it is an extremely important area. Our research is a new alternative for beverage carton packaging recycling of dry grinding technology.

Experiments reported in this article start with this technology, along with new method of using no additives, only mechanical and thermal energy is made using specimens were examined. Our experiments show that our previous hypothesis is true and the polyethylene (PP) affects the specimen structure.

Present study packaging specimens are made of aluminum and non-aluminum beverage cartons. We confirmed in our experiments, that preliminary working hypothesis that the polyethylene (PE) ratio in the sample speciment are affected by the structure of the bodies and the mechanical properties.

Specimens containing aluminium comprise a polyethylene (PE) volume 33% greater than aluminium-free ones. Their tensile strength was greater by 44% and their bending stiffness was 51.37% higher than the aluminum-free ones. However, tear strength difference between the two specimens was only 11.67% for the benefit of the aluminium and more polyethylene containing Tetra Pak specimen. The two types of specimens had nearly the same elongation.

References

1. Ace (Alliance for beverage cartons and enviroment) (2011a) Beverage carton recycling rates 2011 [Online] http://www.beveragecarton.eu/mediaroom/3/50/Beverage-Carton-Recycling-Rates-2011#.UoPnoHB¬SiHc [Accessed 9th of May 2013]
2. Ace (Alliance for beverage cartons and enviroment) (2011b) Continuous increase in Beverage Carton recy-cling across Europe [Online] http://www.beveragecarton.eu/uploads/Modules/Publications/2011acepr020712.pdf [Accessed 9th May 2013]
3. Baka, É.: Mitől klímabarát az italos karton? [Online] http://www.iksznet.hu/index.php [Accessed 9th May 2013]
4. Crattenand, F., Tetra Pak International S.A. Ingram in The Gale Directory of Company Histories, Retrieved 30 October 2011
5. Compostar (2013) Productos compostar [Online] Avaible from: http://www.compostar.ar/productos_conpostar.php [Accessed 9th May 2013]
6. Ecopack (2012) Produtos [Online] Avaible from: http://www.ecopak.com.br/site/pro¬dutos.php [Accessed 9th of May 2013].
7. European parliament and council directive 94/62/ec – of 20 december 1994 – on packaging and packaging waste http://europa.eu/legislation_summaries/environment/waste_management/l21207_hu.ht-m#amendingact [Accessed 9th October 2013]
8. EVD (2010) Material and manufacturing [Online] Avaible from: http://www.evd-diez.de/material/material-and-manufactur¬ing.html [Accessed 9th of May 2013]
9. Italos Karton Környezetvédelmi Egyesülés. (2010). Papírgyári hasznosítás. [Online] Available from: http://www.iksznet.hu/cikkek/papirgyari-hasznositas. (Accessed 9th of May 2013) [Accessed 9th May 2013]
10. Koltai L. (2013) Csomagoló és papíripari anyagismeret, Budapest, Óbuda University, Rejto Sandor Faculty of Light Industry and Environmental Protecting Engineering, pp.47-50, 56-59, 72-75, 82-86.
11. Leander, L., Tetra Pak (1996) ISBN 91-630-4789-6. A vision becomes reality. A company history with a difference, lund: tetra pak international), pp. 18
12. Recipak (na) As Melhores ideias para o futuro de meio ambiente [Online] Avaible from: http://www.recipak.com.br/ [Accessed 9th of May 2013].http://www.ecopak.com.br [Accessed 9th May 2013]
13. Sedig, K. (2002) Swedish Innovations, Stockholm: The Swedish Institute (ISBN 91-520-0910-6), pp. 45
14. Simon, I. (2012) Tetrapak dobozok cellulóztar-talmának meghatározása a termékdíjjal összefüggésben, a nemzeti adó- és vámhivatal szakértői intézetében, Determination Of Cellulose Content in Tetra Pak Containers at The Forensic Institute of The National Tax and Customs Administration, In The Context of The Environmental Tax, BSc thesis, University Óbudai
15. Tetrapak (2013a) Cartons are recyclable [Online] Avaible from:http://www.tetrapakrecycling.co.uk/recyclable_cartons.asp [Accessed 9th of May 2013]
16. Tetrapak (2013b) http://zoblue.com/assets/prof/tetrapak/faq.html [Accessed 9th May 2013]
17. Tplack (na) Fabricación de material de construcción y muebles a partir de cartones para bebidas usados [Online] Avaible from: http://www.agendajoven.org.ar/5_mercadovirtual/procesotplak.htm [Accessed 9th of May 2013]