

**Effects of Different Sealing Conditions on the Seal Strength of Polypropylene Films Coated with a bio-based Thin Layer**

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Preview

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5 Effects of Different Sealing Conditions on the Seal Strength of  
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7 Polypropylene Films Coated with a bio-based Thin Layer  
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## Abstract

This paper presents the results of an investigation through the Design of Experiment (DoE) technique regarding the influence of temperature, dwell time and bar pressure on the heat-seal strength of oriented polypropylene films coated with a gelatin-based thin layer. This chemometric approach allowed achieving a thorough understanding of the effect of each independent factor on the two different responses (maximum force and strain energy) considered in this work as a measure of the strength necessary to break the bond across the sealed interface. Surprisingly, the factor affecting both responses the most was the bar pressure rather than the sealing temperature. Moreover, whereas the bar pressure negatively affected the seal strength of coated polypropylene films, the sealing temperature had a positive effect. Dwell time did not have any significant influence as a main factor, while influencing negatively the seal strength as an interaction term (i.e. *time\*pressure*), together with the further interaction *temperature\*pressure*. The mathematical models obtained for the two responses provided different results in terms of fitting capability ( $R^2$ ) and prediction ability ( $Q^2$ ). In particular, for the maximum force response,  $R^2$  and  $Q^2$  were equal to 0.571 and 0.405 respectively, whereas the model supporting the strain energy response gave  $R^2 = 0.932$  and  $Q^2 = 0.937$ , highlighting that for quantifying the seal strength, the energy necessary to break a seal is a better measure than the maximum force. The highest seal strength values obtained during this work were of 0.6615 N and 19.6 Nmm for maximum force and strain energy, respectively.

KEY WORDS: bio-coating; Design of Experiment (DoE); gelatin; heat-sealing; seal strength.

## INTRODUCTION

Coatings can be defined as very thin layers (from some nanometers to a few micrometers) that can be applied on different substrates, with the goal of improving one or more targeted properties. The term coating encompasses several applications that envisage the use of polymers of both synthetic (e.g. fluorocarbons, polyurethanes, acrylics, vinyl ether polymers, polyesters, alkyd and phenolic resins, silicones) and natural (e.g. starch, casein, gelatin, waxes, cellulose derivatives) origin. Some examples include adhesives, protective barriers, slip/anti-slip agents, conductors, decorations, inks and paints. Nowadays coatings are used for numerous purposes, pertaining to different fields like packaging, automotive, pharmaceutical, marine, photography, textile, army and architectural applications. In addition, the deposition techniques have increased in the last few years, due to the development of newer materials, which contributed to make the world of coating a rather broad one.<sup>1</sup>

Despite the well established use of synthetic coatings for a large number of applications, great interest has recently been expressed through the possibility of developing new coatings directly from renewable resources, especially for food packaging purposes.<sup>2-6</sup> This is due to different reasons. Firstly, the increase in the price of crude oil together with the uncertainty related to its durability, has imposed the necessity to manufacture new structures in order to partially or totally replace oil-derived polymers in the future. Arising from this consideration packaging optimization is nowadays a pressing task for the whole scientific community in the field, as demonstrated by the large number of works retrievable in the scientific literature. Among the different ways that may be pursued to face this issue, the replacement of multi-layered structures with lighter solutions appears a compelling and attainable possibility, and coating deposition on plastic webs is probably one of the most convenient and efficient techniques to accomplish this goal. The interest in coatings

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4 development for packaging applications has recently increased, in the attempt to achieve  
5 better solutions in terms of production costs, final performance and environmental impact.  
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7 Secondly, besides this 'green' benefit, bio-based coatings can provide other obvious  
8 advantages mostly related to their versatile nature and chemical structure. For example, bio-  
9 macromolecules, more so than synthetic polymers, enable tailored structures able to 'sense'  
10 external stimuli (e.g. pH, temperature, humidity) which can act as triggers in all those  
11 applications (e.g., controlled release packaging) where a physical modification of the original  
12 matrix is a necessary pre-requisite to start the release of active compounds (e.g. drugs,  
13 antimicrobials, antioxidants) previously incorporated into the coating.<sup>7,8</sup> Finally, wide  
14 availability and relatively low cost may be two additional key factors making tomorrow's  
15 research on bio-macromolecules such as lipids, proteins and polysaccharides an appealing  
16 field.  
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33 The obtainment of bio-coatings with similar properties to those of synthetic origin is  
34 an important task in order to make them marketable. To accomplish this goal, bio-coatings  
35 should primarily exhibit excellent affinity when coupled with the plastic substrates. In other  
36 words, they should not modify the original positive characteristics of the substrate (e.g.  
37 mechanical strength, transparency). On the contrary, additional benefits after their deposition  
38 are expected, which would provide an ultimate high-performance system. In our previous  
39 paper<sup>9</sup> we investigated the effect of a lipid-protein bio-coating on barrier (oxygen and water  
40 vapour), friction (static and kinetic coefficients) and optical (haze, transparency and UV  
41 transmission) properties of three different plastic films (polypropylene, polyethylene  
42 terephthalate and low-density polyethylene) after coating deposition. We concluded that,  
43 despite some negative effects which need to be addressed, such composite layers may  
44 represent a potential way to meet the ever increasing demand for more sustainable solutions.  
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4 Moreover, we pointed out the necessity for further investigation, especially concerning some  
5 potential aspects like the possibility of using such bio-layers as a heat-sealant. This hypothesis  
6 arises from our belief that the special features of gelatin (the main component of the bio-  
7 coating) as a sealing agent can be properly exploited to obtain natural sealants for food  
8 packaging application. Indeed, the adhesives properties of collagen (from which gelatin is  
9 derived) were already known in the New Stone Age, when this substance was used as a glue  
10 for many applications.<sup>10</sup> The adhesive properties of gelatin, credited to be responsible for the  
11 adhesion properties of biopolymers, can be ascribed to its chemical nature, which involves the  
12 presence of polar functional groups such as hydroxyl (OH), carboxylic acid (COOH) and  
13 aminic (NH<sub>2</sub>) on its molecular backbone.<sup>11</sup> These special features of gelatin appear  
14 particularly attractive in the development of a new gamma of bio-based sealants, which would  
15 make way for the replacement of typically thick heat-sealant plastic polymers (e.g.,  
16 polyethylene), thus drastically reducing the amount of waste dumped into the environment.  
17 Further benefits which would arise from the use of such coatings rely on their natural origin,  
18 which would make contact with packaged food not hazardous when the whole formulation  
19 envisages substances listed by law as either food ingredients or food additives. Consequently,  
20 a first potential application of such 'green' layers could be the replacement of the so-called  
21 cold sealant, thus avoiding the expensive and time-consuming deposition of the sealing layer  
22 only onto the edge of the package (direct contact with the food is forbidden by law).

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49 The aim of the present paper is to evaluate the heat-seal strength of polypropylene  
50 strips coated with a biobased layer acting as a bio-sealant, as a function of the sealing  
51 temperature, pressure and dwell time, by means of the DoE technique.  
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## 59 **MATERIALS AND METHODS**

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## Materials

Biocoated plastic films were prepared using the same raw materials and reagents as reported in our previous work<sup>9</sup> and according to the procedure protected by International patent WO 2008/075396 A1.<sup>12</sup> Briefly, pigskin gelatin powder type A 133 Bloom (Weishardt International, Grauliet Cedex, France), acetic acid esters produced from monoglycerides (Grindsted<sup>®</sup> Acetem 70-00 P, Danisco A/S, Langebrogade, Denmark), and glycerol (Giomavaro, Brugherio, Italy) were used to prepare the starting water solution (Milli-Q, 18.3 MΩ) in the following concentration: 10 wt%, 2.5 wt% and 5.0 wt%. Firstly, the gelatin-glycerol mix was heated to 75°C for 1 h and the pH adjusted to the isoelectric point of the gelatin ( $\approx 8.5$ ) using sodium hydroxide (NaOH) 1 M. At this point, the lipid component was added and mixed for 2 min with a homogenizer (Ika-Werke, Stanfen, Germany), at the speed of 24000 rpm. After degassing the solution using a vacuum pump, a constant amount (1.0 cm<sup>3</sup>) of coating solution was used to coat the corona-treated side (24 x 18 cm) of the plastic film (oriented polypropylene-OPP, 20.0  $\pm$  0.5  $\mu$ m thick - Radici Film, S. Giorgio di Nogaro, Italy) previously placed on an automatic film applicator (Ref. 1137, Sheen Instruments, Kingston, UK) equipped with a steel horizontal rod to obtain a wet coating thickness of approximately 10  $\mu$ m (data provided by the factory). Coating deposition was performed according to ASTM D823-07 – Practice C,<sup>13</sup> at a constant speed of 150 mm min<sup>-1</sup>. Coated films were firstly dried using a constant and perpendicular flux of mild air (25  $\pm$  0.3°C for 2 min.) at a distance of 40 cm from the applicator, and then stored under controlled conditions (23  $\pm$  2°C, 40  $\pm$  2.0% RH) for 24 h. Finally, they were kept in a sealed anhydrous dessicator for 24 h. The thickness of the uncoated plastic film was measured with a micrometer (Dialmatic DDI030M, Bowers Metrology, Bradford, UK) to the nearest 0.001 mm at 10 different random locations. For the determination of the thickness of the biodegradable layers

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4 coated on the plastic films (Table 1), a 10 x 10 cm sample was cut and weighed ( $M_1$ ). The  
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6 coating was mechanically removed using hot water (80°C) and the resulting base film  
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8 weighed ( $M_2$ ). The apparent thickness of the coating was obtained using the following  
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10 equation.<sup>14</sup>  
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$$l = \frac{M_1 - M_2}{\rho} \times 100 \quad (1)$$

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17 where:

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19  $M_1$  = unit total mass (plastic film and coating) ( $\text{g dm}^{-2}$ )

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21  $M_2$  = unit mass of the plastic film ( $\text{g dm}^{-2}$ )

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23  $\rho$  = density of the aqueous solution ( $\text{g cm}^{-3}$ )

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26  $l$  = thickness ( $\mu\text{m}$ )

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29 being  $M_1 - M_2$  and  $\rho$  known.  
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### 34 Seal strength determination

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37 Strips of 2.54 cm wide and 14 cm long were obtained using a precision sample cutter  
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39 (mod. MMT, Thwing-Albert, West Berlin, NJ). Two strips at a time were placed on top of  
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41 one another, and an area of 2.54 x 1.5 cm was heat-sealed using a thermal heat-sealer  
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43 Polikrimper TX/08 (Alipack, Pontecurone, Italy) provided by smooth plates. Temperature,  
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45 pressure and dwell time of the sealing plates were adjustable, and a microprocessor  
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47 maintained the set temperature of each bar throughout the experiment. Sealed samples were  
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49 kept for 24 h at  $23 \pm 0.5$  °C and  $50 \pm 2.0$  % RH in a climatic chamber in order to achieve  
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51 chemical stabilization before testing. Seal strength of heat-sealed samples was measured  
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53 through the so-called T-peel test according to the standard method as described by ASTM F  
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55 88-07a<sup>15</sup> by means of a dynamometer (mod. Z005, Zwick Roell, Ulm, Germany) fitted with a  
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57 100 N load cell and connected to two clamps placed at a distance of 10 cm one from the other.  
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4 Each run was performed at a crosshead speed of  $300 \text{ mm min}^{-1}$ . The peak load required to  
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6 break the joints (maximum force) and the area under the 'load (N)-deformation (mm)' curve  
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8 from zero to the breakage point (i.e. the strain energy) were directly drawn by the software  
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10 'TestXpert V10.11 Master' (Zwick Roell, Ulm, Germany) as a measure of the seal strength.  
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## 14 15 16 **Experimental design** 17

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19 In order to achieve a good seal, it is necessary to achieve a clear awareness of the  
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21 effects of temperature, dwell time and pressure on the seal strength of the final structure (e.g.  
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23 a packaging system).<sup>16</sup> To accomplish this goal, the DoE technique was used as the most  
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25 powerful tool providing the maximum information by the smallest number of trials, with the  
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27 advantage of evaluating multiple parameters and their interactions minimizing costs and time  
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29 of research.<sup>17</sup> In this work, the experimental investigation was carried out by a screening test,  
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31 using a fractional factorial design. Three quantitative controllable factors were considered for  
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33 the screening test: bar temperature ( $X_1$ ), dwell time ( $X_2$ ), and pressure ( $X_3$ ). Each of these  
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35 independent variables was assessed at two equidistant levels (-1 and +1) from the centre point  
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37 (0), as reported in Table 1. In particular, temperature ranged from  $70^\circ\text{C}$  to  $110^\circ\text{C}$ , dwell time  
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39 from 0.5 s to 1.5 s, and pressure from 2.5 to 4.5 bars. Whereas the selected dwell time and  
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41 pressure values account for the most common packaging operations, those related to  
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43 temperature arise from the attempt to develop a better performing solution, i.e. able to seal at  
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45 a lower temperature than conventional thermoplastic sealants. Two dependent variables were  
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47 selected as a measure of the seal strength, i.e. the effort required to separate the heat seal after  
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49 cooling: maximum force ( $Y_1$ ) and strain energy ( $Y_2$ ). A  $2^3$  full factorial design was finally  
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51 chosen, with 33 total runs (10 corner points replicated three times and 3 centre-points). The  
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53 worksheet obtained from this design is reported in Table 2. As far as the selection of the  
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regression model is concerned, it was assumed that  $i$  mathematical functions,  $f_z(z = 1, 2, \dots, i)$  exist for each response,  $Y_z$ , function of  $m$  independent factors,  $X_k$  ( $k = 1, 2, \dots, m$ ), such that:

$$Y_z = f_z(X_1, X_2, \dots, X_m) \quad (1)$$

where:  $i = 2$  and  $m = 3$ . Finally, the  $f_z$  function was assumed to be approximated by a polynomial equation:

$$Y_z = b_{z0} + \sum_{k=1}^{m=3} b_{zk} X_k + \sum_{k \neq j}^{m=3} b_{zjk} X_k X_j + \varepsilon \quad (2)$$

where:

- $Y_z$  = dependent variable;
- $b_{z0}$  = response value when all factors are set at medium level (centre point);
- $b_{zk}$  = linear regression coefficient;
- $b_{zjk}$  = interaction regression coefficient;
- $\varepsilon$  = residual response variation not explained by the model.

The selected regression model and the correlated data analysis provided a proper understanding of the interactions between factors and the correlation between each factor and different responses, by estimating the numerical values of the model terms, i.e. the regression coefficients. MODDE software package (MODDE 2006, version 8.0; UMETRICS AB, Umea, Sweden) was used for the evaluation of raw data and regression analysis in the screening design, according to the least squares analysis technique.

## RESULTS AND DISCUSSION

### Statistical Analysis

Table 3 shows all main linear and interaction regression coefficients calculated for both responses. Among them, the regression coefficient  $X_1X_2$  (the interaction effect

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4 *temperature\*time*) was removed from our models, which were then refitted to the data. The  
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6 not statistical significance of the interaction term *temperature\*time*, although fairly odd, can  
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8 be explained considering that whereas the seal strength increased as the temperature increased  
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10 throughout the experiment, the effect of dwell time on the seal strength was positive only for  
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12 the shortest setting (0.5 s), namely when the two coated strips were brought together for a  
13  
14 sufficient time to diffuse across the interface. As the dwell time increased, a detrimental effect  
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16 on the seal strength occurred, differently from the thermoplastic polymers, for which an  
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18 increase in dwell time is normally associated to an increase in seal strength. Table 4  
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20 summarizes the four statistical parameters which are generally recognized as the most  
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22 important to judge the validity of a model.<sup>18</sup> The best results were obtained for the ‘strain  
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24 energy’ model in terms of fitting capability ( $R^2$ ), prediction ability ( $Q^2$ ) and reproducibility. It  
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26 means that a high percentage of the variation of the response is explained by the model (i.e.  
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28 the model satisfactorily fits the data); a high percentage of the variation of the response is  
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30 predicted by the model (i.e. the model is able to predict the data); the pure error, i.e. the  
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32 variation of the response under the same conditions compared to the total variation of the  
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34 response, is small, as it can be seen in Table 5. Also the parameter ‘model validity’ (which is  
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36 a measure of the lack of fit) has to be considered good, since values over 0.25 are normally  
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38 judged acceptable. The differences between the two models (‘maximum force’ vs. ‘strain  
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40 energy’) may be ascribed to the poor reproducibility, i.e. poor control over the experimental  
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42 error, encountered when the maximum force drawn from each ‘load-deformation’ curve was  
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44 considered as a measure of seal strength. This result confirms that the best measure of seal  
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46 strength across an interface is the amount of energy necessary to break it, i.e. to separate the  
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48 two surfaces, rather than the maximum force required to break the bond.<sup>19,20</sup>  
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Lastly, the two polynomial equations that allow the modelling of the relationship between factors and each response are:

$$\text{Maximum force } (Y_1) = 0.453 + 0.022X_1 + 0.006X_2 - 0.077 X_3 - 0.027X_1X_3 - 0.043X_2X_3$$

$$\text{Strain energy } (Y_2) = 10.591 + 1.750X_1 + 0.233X_2 - 2.251X_3 - 1.626X_1X_3 - 1.633X_2X_3$$

Analysis of variance (ANOVA) is summarized in Table 5. For both models the mean square regression (i.e. the variance explained by the model) in the first  $F$ -test was significantly larger (approximately one order) than the mean square residual (i.e. the amount of variance unexplained by the model). This means that both polynomial equations adequately represented the data for maximum force and strain energy, even though by a different extent. Furthermore, the lack of fit test showed that model error and replicate error were small and of similar size, suggesting that there was no lack of fit. It demonstrates that the two models were sufficiently accurate to predict each corresponding response, although in different ways.

### Effect of Factors on Responses

The regression models developed within the screening design highlighted the influence of each factor (temperature, dwell time and bar pressure) on the two responses ('maximum force' and 'strain energy'). As represented in Figure 1, the most influencing factor was the 'bar pressure', which acted in inverse proportion on both responses. Further, 'strain energy' was positively affected by 'bars temperature', whose minimal effect was, on the contrary, not-statistically significant for the 'maximum force' response as shown in Table 4. For both dependent variables mention must be made of the significant interaction effect

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4 *time\*pressure*, whereas the interaction effect *temperature\*pressure* was significant only for  
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6 the 'strain energy' response (Table 4).  
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9 For an intuitive interpretation of the relationship between factors and responses, the  
10 response surface contour plots are undoubtedly the most effective tools since they are able to  
11 indicate the direction towards which the different variables should be set when specific  
12 response values are aimed. Some epitomizing response surface plots are illustrated in Fig. 2  
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14 (a, b and c). The relationship between temperature and pressure in defining the strain energy  
15 response is shown in Figure 2a. The two factors influenced the selected response oppositely,  
16 i.e. an increase in temperature led to an increase in the strain energy, whereas a reduction of  
17 the same response occurred due to an increase in pressure. The same plot highlights the  
18 interaction effect between the above-mentioned two factors. Whereas for the lower pressure  
19 values an increase in temperature produced an increase in seal strength, for the higher  
20 pressure setting (> 4.0 bar) an increase in temperature did not lead to any improvement, so  
21 that the seal strength of the tested samples (expressed as strain energy) was approximately the  
22 same (~ 8.22 Nmm) at any sealing temperature within the experimental region investigated in  
23 this work, regardless of the dwell time. It is worth noting that, although within this  
24 experimental design the term 'dwell time' is not significant as a main effect, it becomes  
25 relevant as an interaction term. It is shown in Figure 2b, where the strong interaction effect  
26 between dwell time and pressure for both responses is reported. In particular, both response  
27 contour plots display that the effect of bars pressure on maximum force and strain energy is  
28 strictly dependent on the set value of dwell time. It is more clearly evident in Figure 3: when  
29 the dwell time is set at its lower coded value (-1), the effect on strain energy arising from an  
30 increase of bar pressure from -1 to +1 is negligible (from ~10.9 Nmm to ~9.7 Nmm).  
31 Conversely, in correspondence to the highest dwell time (1.5 s), moving from 2.5 bars to 4.5  
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4 bars led to a more pronounced strain energy decrease (from ~14.7 Nmm to ~6.9 Nmm). To  
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6 correctly quantify the *dwelt time\*pressure* interaction, the effect of temperature also needs to  
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8 be taken into account. It is appreciable from Figure 2c that an increase in temperature caused  
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10 a dramatic decrease in strain energy due to the interaction effect between dwell time and  
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12 pressure. This is especially obvious when the dwell time was set at its highest value (1.5 s). In  
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14 this case, changing the bar pressure from the lowest (-1) to the highest (+1) coded values  
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16 produced a strain energy difference as large as the selected sealing temperature was high.  
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18 Accordingly, for a sealing temperature of 70°C the above difference was equal to 3.5 Nmm  
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20 (given by the difference 10.82 Nmm – 7.32 Nmm), whereas it was equal to 6.88 Nmm and  
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22 10.8 Nmm when the temperature was set at 90°C and 110°C, respectively.  
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28 These results elucidate the different effect exerted by the three main parameters of the  
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30 sealing process on the bonding performance of the bio-coating. Temperature and bar pressure  
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32 were of primary importance as a main effect, though in a diametrically opposed way, while  
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34 dwell time played an important role only as an interaction effect. In other words, seal strength  
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36 improved as temperature increased, whereas it was negatively affected by increasing the bar  
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38 pressure. However, the positive effect of temperature completely disappeared when  
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40 approaching pressure values of approximately 4.5 bars, as much as the negative effect of  
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42 pressure was amplified by increasing the sealing temperature (interaction effects). Further,  
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44 both interactions were emphasized by prolonging the dwell time, which however had no  
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46 significant effect as an individual factor, as already mentioned. These results do not  
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48 completely agree with those obtained by Theller<sup>16</sup>, Stehling and Meka<sup>21,22</sup>, who pointed out  
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50 that heat sealability of plastic films in bar sealing applications is primarily controlled by  
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52 sealing temperature and dwell time, rather than pressure. Therefore, our finding represents an  
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54 original feature of such bio-sealants.  
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These results can be explained through the joints formation mechanism underlying the sealing process. The two coated film strips are brought together by means of two heated bars, which simultaneously exert a pre-set pressure during contact, whose duration should be properly selected to ensure enough heat reaches the sealant polymer, while avoiding profitless time wasting. The heat transfer from the plates through the plastic films allows melting the crystalline gelatin-based sealant, whereas the applied pressure promotes the interfacial interaction across the two coated surfaces. The longer the contact time of such an interaction, the more interpenetrating it will be. If the above mentioned three parameters were properly set, a heat seal of sufficient strength would be formed, due to the recrystallization of the melted gelatin upon cooling. However, as shown by this investigation, the sealing coating properties may be strongly affected by changing the temperature setting, time and pressure. In particular, the negative effect on seal strength due to an increase in bar pressure (from 2.5 bars to 4.5 bars) could be explained in terms of ‘squeezing’ effect exerted by the pressure. We hypothesize, that increasing the bar pressure, the intimate contact between the two coated sides of the plastic film is exaggerated in so much as the bio-coating comes out from the sealing contact area. This effect is of course emphasized by the bars temperature setting, since the melting process of the biosealant is faster at higher temperatures for a given pressure value. The final result is the weakening of the seal due to a drastic reduction in the thickness of the touching coated surfaces. In addition, especially for polymers obtained from biomacromolecules, the detrimental effect arising from high temperatures should not be excluded, particularly as the dwell time is increased. Besides the chemical properties of the substrate itself, it must also be noted that the coating thickness is a pivotal parameter in defining the seal properties. In this work, the apparent coating thickness was on average of  $1.2 \pm 0.08 \mu\text{m}$ , hence a very low value in comparison with the sealing layers commonly used.

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5 Probably, increasing the coating thickness by a small extent would produce some benefits,  
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7 especially due to the relatively higher thermal conductance of the whole sample (substrate  
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9 plus coating), which could somehow delay the adverse effects caused by the interaction  
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11 between high temperature, pressure and dwell time, as previously mentioned.  
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14 Finally, it is worth citing the mode of failure of the heat-sealed joints as investigated  
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16 by visual inspection of the sealed interface after rupture. It is theoretically assumed that the  
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18 mode of failure can be both cohesive and adhesive depending on where the seal fails.<sup>19</sup> When  
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20 the seal strength is greater than that of the substrate (e.g., the plastic packaging), tearing mode  
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22 failure is cited. Conversely, when the rupture is between the sealant-coated surfaces, the  
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24 failure occurs in the peeling mode.<sup>23</sup> In this work, polypropylene sealed strips separated from  
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26 each other according to the peeling mode, as confirmed by the presence of the bio-coating on  
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28 both sealed faces of the plastic strips. On the other hand it would be fairly difficult to have a  
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30 tearing mode failure with seal strength values as high as 19.6 Nmm (in terms of strain energy)  
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32 and 0.66 N (in terms of maximum force), which are typical of easy-opening peel seals.  
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## 40 CONCLUSIONS

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42 This research investigated the seal strength attribute as a function of the most  
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44 important sealing parameters of a coating developed by using macromolecules of natural  
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46 origin. The results obtained by means of the Design of Experiment technique firstly  
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48 confirmed the strain energy as the best measure for quantifying the seal strength, graphically  
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50 given by the area under the load-deformation curve recorded during the so-called T-peel test.  
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52 Furthermore, it has been possible to achieve detailed information on the influence of sealing  
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54 temperature, dwell time and bars pressure on the seal strength of the gelatin-based coating,  
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56 which exhibited unique sealing characteristics when compared to conventional heat sealant  
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4 thermoplastic films. In particular, ‘bar pressure’ and ‘bars temperature’ turned out to be the  
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6 most influencing factors on the strain energy of the specific formulation studied in this work.  
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8 The interactions *time\*pressure* and *temperature\*pressure* played also an important role.  
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10 Moreover, the DoE approach and the related modelization of experimental data allowed  
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12 achieving important findings that can be profitably exploited in practice. More specifically,  
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14 the response surface plots analysis enabled the proper combination of the three sealing  
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16 parameters according to the desired seal strength of the biocoating.  
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21 However, it should be pointed out that the obtained results concern only the specific  
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23 gelatin-based formulation used, i.e. they can not be extended to any potential formulation in  
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25 which the same ‘ingredients’ are used, since the sealing attribute is strongly influenced by the  
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27 combination of the starting molecules. Similarly, different sealing attitudes are expected when  
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29 the thickness of the produced coatings differ from that used in this work. Thus, in order to  
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31 spawn more general findings, further research on the effect of each formulation-variable as  
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33 well as of the coating thickness on the sealing behavior is necessary. In addition, it has to be  
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35 stressed that our investigation has been carried out within a well defined experimental region.  
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37 It means that for any factors combination outside it, new conclusions should be drawn by new  
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39 experiments.  
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45 On the basis of the results presented in this study it can be concluded that a new class  
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47 of coatings with unique features may be generated from readily-available macromolecules of  
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49 natural origin. In the near future this type of new biocoatings could serve several purposes in  
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51 an intelligent and sustainable fashion, although the road ahead is still rough going and various  
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53 issues need to be addressed.  
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## Tables

**Table 1. Factors and their levels for the 2<sup>3</sup> full factorial design**

Variable	Unit	Symbol		Levels		
		Coded	Uncoded	-1	0	+1
Plates temperature	°C	X <sub>1</sub>	T	70	90	110
Dwell time	s	X <sub>2</sub>	t	0.5	1.0	1.5
Plates pressure	bar	X <sub>3</sub>	p	2.5	3.5	4.5

**Table 2. Worksheet of the 2<sup>3</sup> full factorial design**

Exp. N°	Run order	Variable levels			Responses	
		X <sub>1</sub>	X <sub>2</sub>	X <sub>3</sub>	Y <sub>1</sub> <sup>a</sup>	Y <sub>2</sub> <sup>b</sup>
1	13	70	0.5	2.5	0.4075	7.7466
2	8	110	0.5	2.5	0.5534	15.1154
3	14	70	1.5	2.5	0.5423	11.3884
4	29	110	1.5	2.5	0.6513	19.6056
5	12	70	0.5	4.5	0.4448	9.3578
6	23	110	0.5	4.5	0.3639	9.9506
7	26	70	1.5	4.5	0.3325	6.9899
8	21	110	1.5	4.5	0.2637	6.1143
9	27	90	1	3.5	0.3309	9.3118
10	28	90	1	3.5	0.3934	10.2752
11	5	90	1	3.5	0.3938	9.7227
12	1	70	0.5	2.5	0.4653	7.5287
13	10	110	0.5	2.5	0.563	15.8183
14	11	70	1.5	2.5	0.6615	12.8105
15	22	110	1.5	2.5	0.5973	16.2851
16	15	70	0.5	4.5	0.4248	10.1362
17	24	110	0.5	4.5	0.3454	9.7927
18	9	70	1.5	4.5	0.3066	6.4393
19	25	110	1.5	4.5	0.4637	8.4886
20	16	90	1	3.5	0.4005	9.5027
21	3	90	1	3.5	0.5954	10.4872
22	2	90	1	3.5	0.6147	9.7227
23	17	70	0.5	2.5	0.3195	6.6536
24	30	110	0.5	2.5	0.575	13.9764
25	32	70	1.5	2.5	0.489	11.647
26	4	110	1.5	2.5	0.5388	17.5018
27	20	70	0.5	4.5	0.4603	10.0543
28	7	110	0.5	4.5	0.4362	10.1353
29	33	70	1.5	4.5	0.3136	7.3019
30	19	110	1.5	4.5	0.3529	7.2927
31	31	90	1	3.5	0.3717	9.4005
32	18	90	1	3.5	0.4661	11.736
33	6	90	1	3.5	0.5341	11.237

<sup>a</sup> Maximum force (N). <sup>b</sup> Strain energy (Nmm).



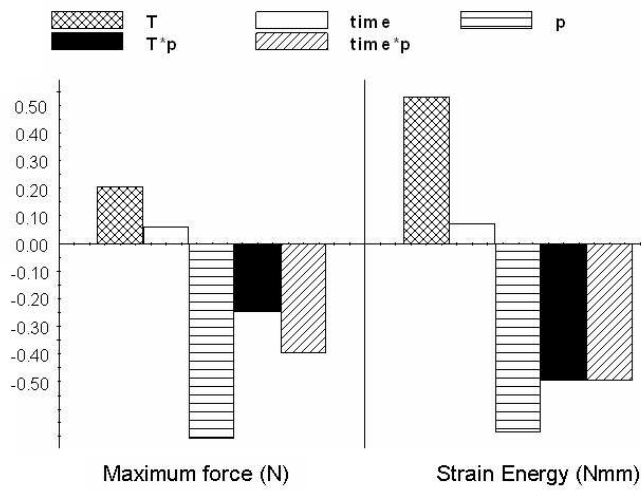
## Figures Legend

Figure 1. Coefficient overview plot for 'maximum force' and 'strain energy' responses.

Figure 2. (a) Response contour plot for *Temperature\*Pressure*; response: strain energy. Time: 1 s.  
(b) Response contour plot for *Time\*Pressure*; responses: maximum force (left) and strain energy (right). Temperature: 70°C. (c) Response contour plot for *Time\*Pressure*; response: strain energy. Temperature: 90°C (left) and 110°C (right).

Figure 3. Interaction plot for *Time\*Pressure*; response: strain energy.

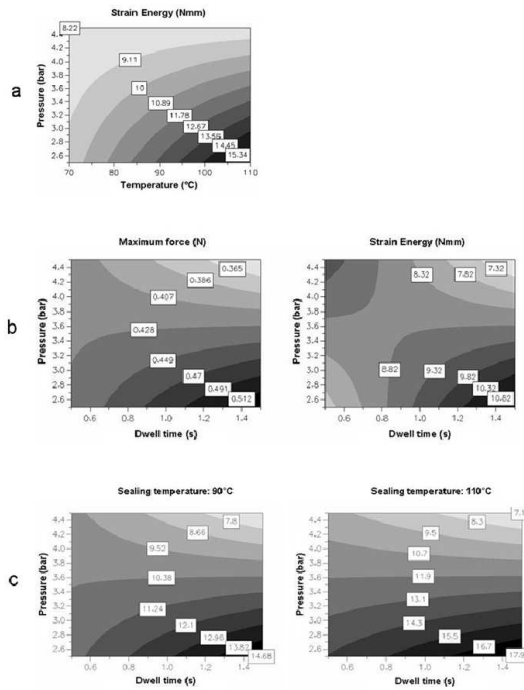
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Coefficient overview plot for 'maximum force' and 'strain energy' responses.  
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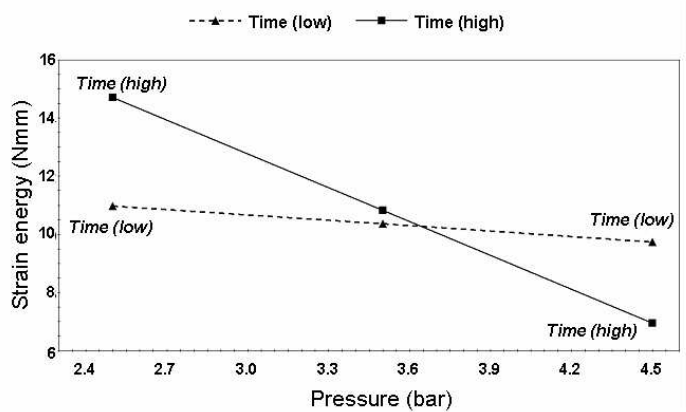
Review





(a) Response contour plot for Temperature\*Pressure; response: strain energy. Time: 1 s. (b) Response contour plot for Time\*Pressure; responses: maximum force (left) and strain energy (right). Temperature: 70°C. (c) Response contour plot for Time\*Pressure; response: strain energy. Temperature: 90°C (left) and 110°C (right).  
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Interaction plot for Time\*Pressure; response: strain energy.  
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Review