

## Research Article

# Nanosensors for a Monitoring System in Intelligent and Active Packaging

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A theoretical wireless nanosensor network (WNSN) system that gives information about the food packaging condition is proposed. The protection effectiveness is estimated by measuring many factors, such as the existence of microorganisms, bacteria, gases, and contaminants. This study is focused on the detection of an antimicrobial agent (AA) attached on a polymer forming an active integrated package. All monitoring technologies for food conservation are analyzed. Nanobiosensor nanomachine (NM), which converts biological or chemical signals into electrical signals, is used. A mathematical model, which describes the constituent's emigration from the package to food, is programmed in MatLab software. The results show three nanobiosensors forming a WNSN. The nanobiosensors are able to carry out the average concentration for different spots in the package. This monitoring system shows reading percentages in three degrees and different colors: excellent (green), good (cyan), and lacking (red). To confirm the utility of the model, different simulations are performed. Using the WNSNs, results of AA existing in food package (FP) through time were successfully obtained.

## 1. Introduction

The new food packaging technologies are developed in response to consumer demands and trends in industrial production of conservation, product freshness, flavor stability, longer service life, monitoring, and traceability. The mentioned topics allow quality control and compliance with health requirements. In this sense, the packaging is a constantly evolving field, achieving a leading role in the conservation and improvement of the characteristics of food [1].

The safety of food products is one of the main objectives of food law. Packaging protects food from external factors, such as heat, light, humidity control, oxygen level, relative pressure, enzymes, odors, microorganisms, insects, contaminants, dust, gaseous emissions, and lipid oxidation [2].

To prolong the food life it is necessary to delay the enzymatic activity and biochemical reactions using different

strategies, such as temperature control and humidity control, by adding chemical components such as salt, sugar, carbon dioxide, antioxidants, and antifungal, antimicrobial, natural acids, and removing oxygen or compounds of these in packaging [3].

## 2. Packaging Technologies

**2.1. Active Packaging (AP).** Active packaging is one of the most innovative concepts and the main engine of nanomaterial applications in the food packaging industry. This technology is based on the concept of incorporating specific components (additives with antimicrobial and antioxidants properties and fungicides) in the packaging systems, in order to prolong the life, maintaining or improving the condition and quality of the packaged food [4]. These AP enable release or absorption of substances or additives from the packaged

food or the surrounding environment, for example, retarding lipid oxidation in the stored food and similarly limit any undesirable flavors caused by direct addition of these additives in foods [5].

**2.2. Intelligent Packaging (IP).** Considered as the next generation of packaging systems, the IP are technologies to monitor and provide information of the status of the food storage, the packaging, or the internal and external environment by ensuring the quality indicators [6]. These packaging systems are capable of performing intelligent functions such as detection, acquisition, recording, monitoring, communication, and application of scientific logic, to facilitate decision making, extending the life and warning potential problems [7]. This type of packaging analyzes the system, processes information, and presents it, without generally exerting any action on the food.

**2.2.1. Time and Temperature Indicators (TTIs).** Control devices are easy to use and can be integrated as part of the package, and most of them allow consumers to check the quality of food through a color response that matches or correlates with the quality of a food at a given temperature [8]. Depending on the operating mechanism, they may be based on physical systems such as diffusion, chemical systems such as polymerization reactions, and biological systems such as an enzymatic reaction or require some initial activation, for example, by ultraviolet light [9].

**2.2.2. Integrity Indicators.** The most common are the indicators of leakage, to detect perforations and faults in the sealed container. The most commonly used indicators are gas, oxygen ( $O_2$ ), and carbon dioxide ( $CO_2$ ) [10]. The simpler indicators of integrity are time indicators, which provide information about the period in which a product has been opened.

**2.2.3. Freshness Indicators.** The idea of freshness indicators is to give direct information about product quality, emitting a signal when the food condition becomes unacceptable during storage, transportation, sale, and consumption [11]. The signal operates on the basis of change of the indicator, which occurs as a result of different compositions within the atmosphere of the package, due to the chemical and microbiological changes of the packaged product [12].

**2.2.4. Radio Frequency Identification.** Radio frequency identification (RFID) is a wireless data collection technology that uses electromagnetic waves (EW) to transfer between a transmitter and/or receiver [13]. These RFID tags comprise an antenna and a storage unit microchip memory, embedded in a material called substrate (Figure 1). For intelligent food packaging, the use of RFID tags is possible; obtaining real-time data on the packaging allows control of the integrity, authenticity, antitheft protection, anticounterfeiting, quality, and traceability [14].

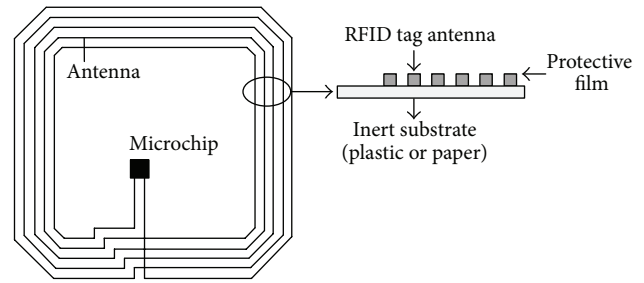


FIGURE 1: Schematic representation of the RFID tags.

### 3. Nanotechnology in Smart Packaging

The indicator/nanosensor interacts with internal factors (food components) and/or environmental factors. As a result of this interaction the indicator/nanosensor will generate a response (visual signal, electric signal) that correlates with the state of the food product. The information generated not only is useful for communication with consumers, providing them with information on safety and quality of products but also can be used by the producers in their decision support systems to determine when and what measures should be taken over the entire production process and distribution channels for products [15].

Recent technological advances and ongoing scientific research are being directed towards the field of printed electronics, nanotechnology carbon, silicon photonics, and biotechnology [16]. These scientific advances offer the possibility of developing a new generation of nanosensors with applications in food packaging.

**3.1. Difficulties of the IP Technology.** There have been great advances in the last decade; however several obstacles remain to overcome large-scale commercial application. The main obstacles are reducing the size and rigidity (improved flexibility), reduced development and production costs, increased sensitivity and robustness (resistance to mechanical stress, exposure to light, and temperature variations), and strict compliance with the law and low consumer acceptance ("nanophobia" to new nanotechnology), although they can help to improve the safety and quality of food products [15].

**3.2. Nanosensors in IP.** Nanosensors have great potential to accelerate the rate of detection, identification, and quantification of pathogens, decaying substances, and allergy-causing proteins [17]. Therefore, these nanodevices have the potential to significantly impact food security. Generally, nanosensors are placed on food packaging to monitor internal and external conditions of the products [18] and in the precise identification of various contaminants in food [19].

**3.3. WNSNs in Intelligent Packaging Industry.** The nanocommunication refers to the exchange of information between NM, which can be done through nanomechanical, acoustic,

electromagnetic, and chemical or molecular mass communication [20]. Wireless communication, the process of collecting energy, and limited computer resources are the main features that guide the design of protocols and architectures WNSNs [21].

Communication between nanosensors expands the capabilities and applications of nanodevices; for the moment, it is unclear how these NM will communicate. There is the vision of two main alternatives for communication at the nanoscale: molecular communication [22, 23] and nanoelectromagnetic communication [24, 25]. The detection range of existing nanosensors requires them to be in the phenomenon being measured, and the area covered by a single nanosensor limits its scope of action [26]. Therefore a network of nanosensors will be able to cover larger areas and communicate with each other through a wireless channel [27].

In this paper we propose a theoretical system of integrated packaging AP and IP, which represent the migration of AA which is used to protect the food. The objective of this work is to simulate the concentration of AA on each instant of time impregnated in the polymer matrix of an FP. In our study, thymol is used as AA. Thymol oil is obtained from a group of aromatic plants called Thyme plants. It can be used in foods and packages as an antimicrobial agent. Different studies have demonstrated the effectivity of this agent against the pathogens transmitted in the food, such as *Listeria monocytogenes*, *Salmonella typhimurium*, *Escherichia coli*, *Shigella dysenteriae*, *Bacillus cereus*, and *Staphylococcus aureus* [28]. The impregnation process was carried out with CO<sub>2</sub> values above the critical point (7.38 MPa and 304.1°K). By doing this, the attachment of polymeric matrices used in the AP formation is performed. The tests were run with three different pressure values and the same temperature using supercritical CO<sub>2</sub>SC conditions. Under this scenario, AA becomes soluble and leaves the polymeric film with different initial AA concentration. A set of three nanobiosensors have been connected into a WNSN in order to obtain the concentration value. Each NM will compute an estimation of the AA migration in different container points. The global concentration percentage will be displayed by using a specific color according to the concentration of AA in the matrix of the container. A food packaging is considered as a ternary system (environment-package-food), where beneficial and detrimental interactions occur. One of these interactions is a phenomenon called migration that has been mathematically modelled, programmed, and simulated in the MatLab, and the model was adjusted by using experimental data from studies conducted in laboratory (Table 1) [29–32].

With this method, it is possible to monitor the status of a FP during its life cycle, providing consumer information about the state of the container antimicrobial protection and also providing the producer with information about when the protection is insufficient and does not allow a reutilization. Figure 2 describes the theoretical system of monitoring proposed using WNSNs. It is expected that in the future the AP and IP integration will be a promising market.

TABLE 1: Initial concentration, partition coefficient, and diffusion coefficient of AA in the linear low-density polyethylene (LLDPE), for different conditions of impregnation and same temperature 313.15°K. Food simulant (FS) 95% ethanol.

Pressure [MPa]	Thickness films [ $\mu\text{m}$ ]	
7	165.8	
9	170.6	
12	181.2	
$C_p$ [mg/kg]	$K_{P/FS}$	$D_p$ [ $\text{m}^2/\text{s}$ ]
4300	58	$1 * 10^{-12}$
10700	25	$3 * 10^{-12}$
13600	15	$5 * 10^{-12}$

## 4. Mathematical Model

**4.1. Mass Transfer Equations.** Figure 3 shows an outline of the system to be analyzed in this work. Three stages are pondered; the first in the polymer matrix is considering the migrant concentration profile present in the matrix (layer I), then taking into account the distribution in the simulant phase in contact with the polymeric matrix, and finally the concentration profile of the boundary layer at the vicinity of the interface (layer II). In this study a symmetric concentration profile is considered.

From the results of the partition coefficient between the polymer and the food simulant ( $K_{P/FS}$ ), obtained from experimental analysis of migration, along with the values of initial concentration of the migrant, the polymer properties, and food simulant, the mathematical model correlating the diffusion coefficient of the migrant in the polymer ( $D_p$ ) was calculated.

Several authors have proposed mathematical models to give analytical solutions to Fick's laws [33, 34]. The mathematical model used consists of applying a simplified numerical solution of Fick's law to calculate the diffusion coefficient values of AA through the polymer films.

The equation that estimates the mass transfer of the migrant by molecular diffusion, according to Fick's law, through the polymer matrix is

$$J_1 = \frac{D_p}{L/2} \cdot \left( C_m^P(x=0, t) - C_m^P\left(x = \frac{L}{2}, t\right) \right), \quad (1)$$

where  $J_1$  is the transfer flow of the migrant [ $\text{kg}/\text{m}^2\text{s}$ ],  $D_p$  is the active diffusion coefficient in the plastic film [ $\text{m}^2/\text{s}$ ],  $L$  is the film thickness [m],  $C_m^P(x=0, t)$  is the concentration of migrant within the polymer [ $\text{kg}/\text{m}^3$ ], and  $C_m^P(x=L/2, t)$  is the concentration of migrant at the interface on the side of the polymer [ $\text{kg}/\text{m}^3$ ].

In the interphase there is the partition coefficient of the migrant in equilibrium represented by

$$K_{P/FS} = \frac{C_m^P(x=L/2, t)}{C_m^{FS}(x=L/2, t)}. \quad (2)$$

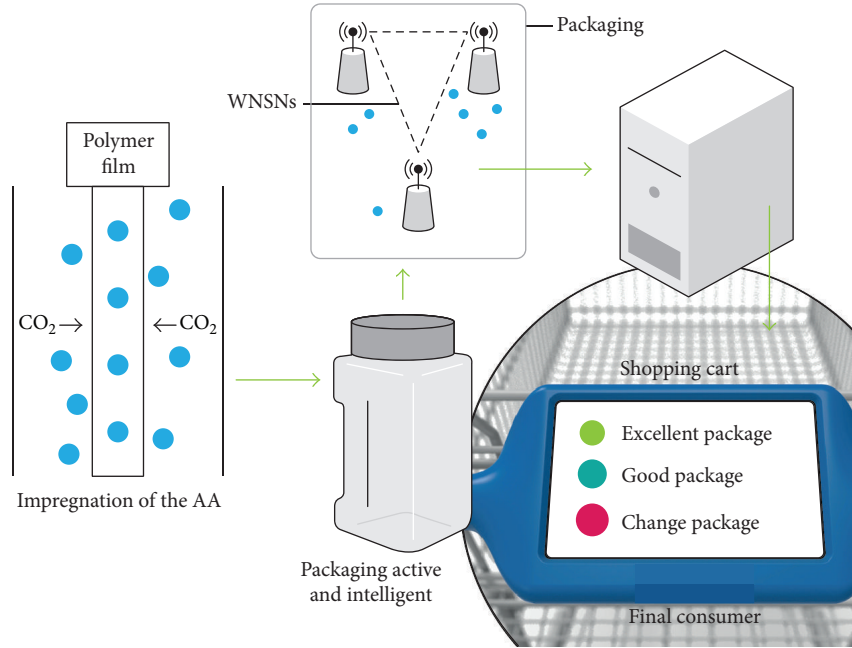


FIGURE 2: Schematic representation for a theoretical monitoring system using WSNs with AP.

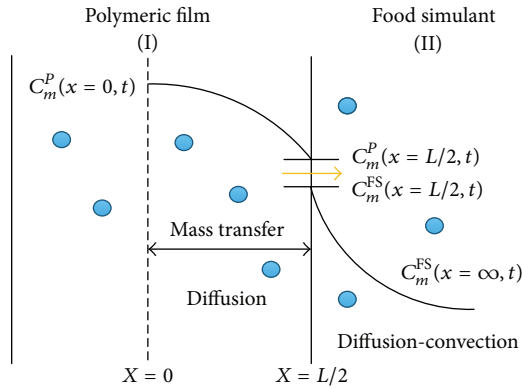


FIGURE 3: Scheme of the migration process: concentration profiles and equilibrium conditions in the surrounding of the material/simulant interphase.

Lastly, the transfer through the boundary layer in the simulant phase at the vicinity of the interphase is described by

$$J_{II} = K \cdot \left( C_m^{FS} \left( x = \frac{L}{2}, t \right) - C_m^{FS} (x = \infty, t) \right), \quad (3)$$

where  $K$  is the mass transfer coefficient that quantifies the natural convection in the simulant phase [m/s] and  $C_m^{FS}(x = \infty, t)$  is the migrant within the food simulant [kg/m<sup>3</sup>].

The initial concentration of the migrant in the polymer is considered with the following initial condition:

$$C_m^P(x = 0, t = 0) = C_m^P. \quad (4)$$

In this way, for  $t = 0$ ,  $C_m^P(x = 0, t) = C_m^P(x = L/2, t) = C_m^{P_{mo}}$ .

It is assumed that initially there is no migrant in simulant, which defines a second initial condition:

$$C_m^{FS} \left( x \geq \frac{L}{2}, t = 0 \right) = C_m^{FS_{mo}} = 0. \quad (5)$$

This equation implies that if  $t = 0$ , then  $C_m^{FS}(x = L/2, t) = C_m^{FS}(x = \infty, t) = 0$ .

As a boundary condition, the represented condition of symmetry is indicated by

$$\left( \frac{\partial C_m^P(x, t)}{\partial x} \right)_{x=0} = 0. \quad (6)$$

The model is solved performing an iterative calculation of the estimate of flow of the mass transfer through the interface when  $J_I = J_{II}$ , and the iteration is performed until the differences between the values of the diffusion coefficient of the migrant in the polymer between experimental and simulated values of  $C_m^{FS}(x = \infty, t)$  are minimal, applying the method of the root mean square error (RSME%) proposed by [35], by the following equation:

$$RSME = \frac{1}{m_{P,O}} \cdot \sqrt{\left( \frac{1}{N} \right) \cdot \sum_{i=1}^N \left( (m_{FS,t})_{\text{experimental},i} - (m_{FS,t})_{\text{predicted},i} \right)^2}, \quad (7)$$

where  $m_{P,O}$  is the initial amount of migrant in the polymer [ $\mu\text{g}$ ],  $N$  is the number of experimental points for each curve of migration,  $i$  is the number of observations, and  $m_{FS,t}$  is the amount of migrant in the simulant in time [ $\mu\text{g}$ ].

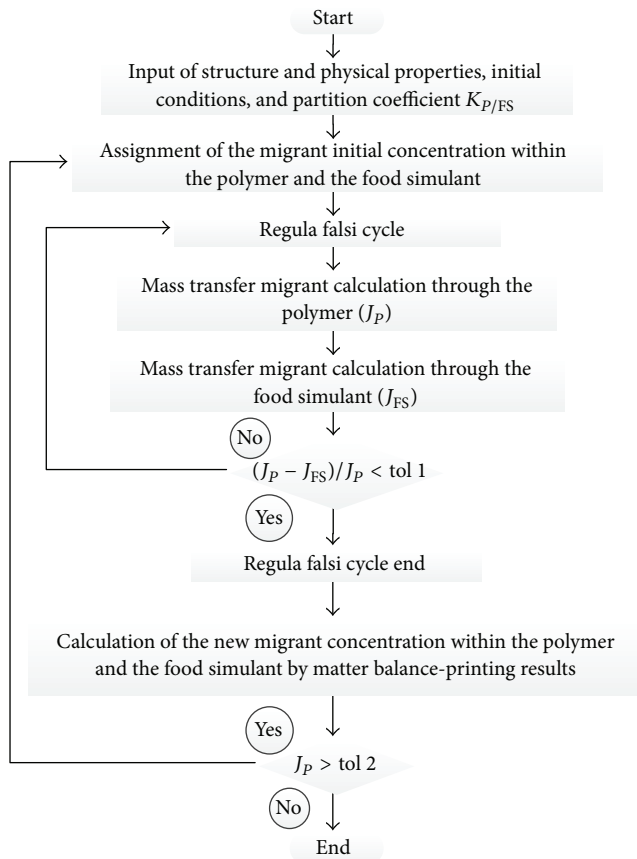


FIGURE 4: Algorithm programming and modeling developed in the MatLab software.

These calculations were implemented by means of a program built in MatLab R2014a. The outline of the calculation algorithm is presented in Figure 4.

## 5. Simulation Results

After the simulation under conditions previously established, the values provided by each nanobiosensor are obtained, obtaining data at different points of the package, averaging them by WNSNs. In Figures 5–7, the results of the AA migration analysis of the impregnated films at 7, 9, and 12 MPa are observed. According to the results given by the WNSNs, we can see that the higher the impregnation pressure, the higher the concentrations of AA in the polymer matrices of the AP, while the number of days of antimicrobial protection increases. In Figure 5, the equilibrium is reached approximately in 50 days; “Equilibrium” can be defined as when little or no mass transfer of AA exists. Nanobiosensor 1 presents the higher migration providing a concentration of  $4.8 \text{ mg/dm}^2$ . According to WNSNs, on day 11 it presents excellent protection against AA; days 11 to 30, it has good protection; after day 30, it has little protection against AA in the polymeric matrix of the container.

The results of Figure 6 equilibrium are reached approximately in 90 days, presenting a far greater migration provided by nanobiosensor 1 to give at this time a concentration in

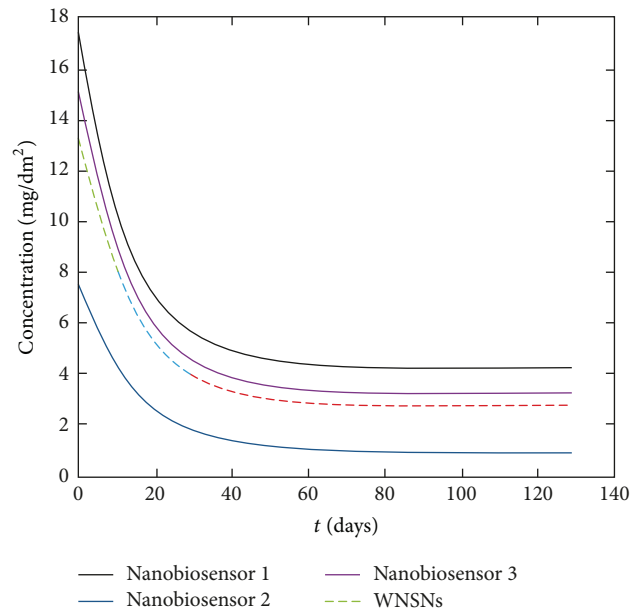


FIGURE 5: AA concentration values given by the WNSNs impregnated containers at  $313.15^\circ\text{K}$  and 7 MPa.

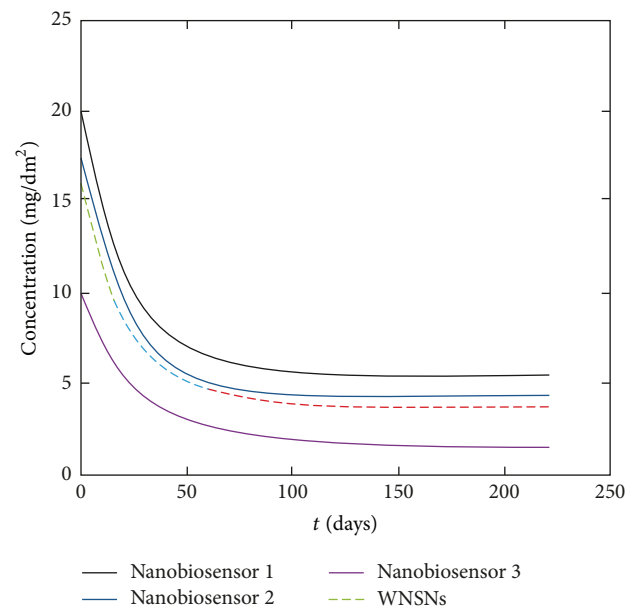


FIGURE 6: AA concentration values given by the WNSNs impregnated containers at  $313.15^\circ\text{K}$  and 9 MPa.

balancing  $6 \text{ mg/dm}^2$ . According to WNSNs, on day 21 it presents excellent protection of the AA; days 21 to 60, it has good protection; after day 60, the container has poor protection against AA in the polymeric matrix.

In Figure 7, equilibrium is reached approximately in 110 days, presenting a far greater migration provided by nanobiosensor 3 to yield to that time a concentration in the balance of  $8.9 \text{ mg/dm}^2$ . According to WNSNs, on day 31 it has excellent protection against AA; days 31 to 88, it has good protection; after 88 days, little protection is provided against AA in the polymeric matrix of the container.



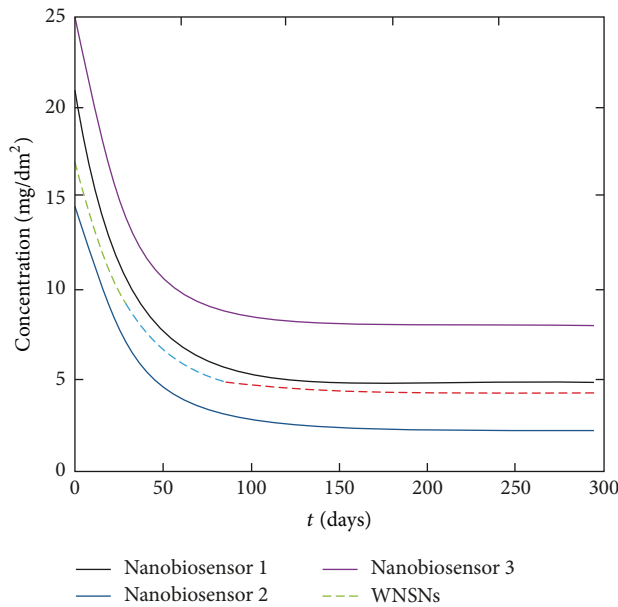


FIGURE 7: AA concentration values given by the impregnated WNSNs containers at 313.15°K and 12 MPa.

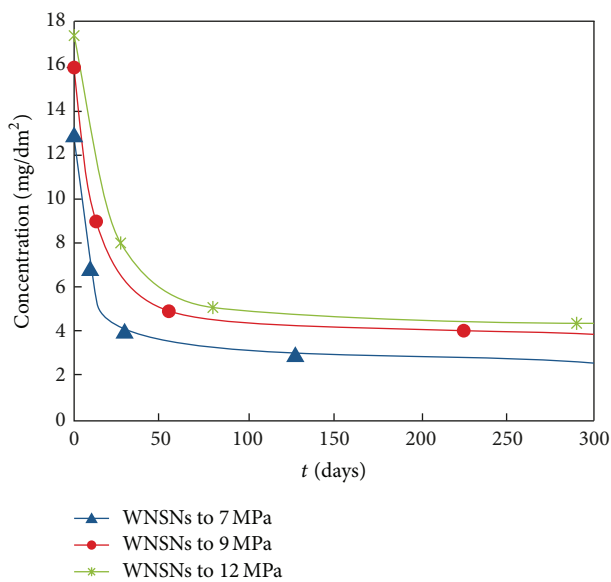


FIGURE 8: AA stability in time. Values given by the WNSNs.

The results obtained by the WNSNs in Figure 8 show in all cases that the concentration of AA in the containers decreases with time, but even at day 300 AA is still present in its composition, about 25% of the initial concentration at 7 MPa, 24% at 9 MPa, and 19% at 12 MPa, indicating that the release of AA from the films takes place gradually.

Figure 2 shows an example of how the red indicator (change package) is activated in the shopping cart, when a threshold concentration of 10% is assumed.

## 6. Conclusions

In this paper we have proposed a theoretical monitoring system in which the information of the FP condition is given, based on a WNSN. Across the nanotechnology two new packaging technologies are integrated, AP and IP, which use nanobiosensor as main detection method, and the relationship between initial and final AA concentration on different time frames was successfully obtained. WNSNs are able to measure the concentration in different spots of the package along the whole supply chain to the final consumer; as standard of judgment the measures given by the nanobiosensor were used. The WNSNs average out the results given by the nanobiosensor sorting them in three ranks: excellent, good, and lacking with their own colors green, cyan, and red.

For the purpose of confirming the system's utility, different simulations were made, all these with the same constant temperature of 313.15°K with three different pressure values 7, 9, and 12 MPa. AA is impregnated into gaps of the polymer, and its initial concentration distribution is different throughout the polymer matrix. Therefore concentration on the surface of the polymer will change in a different manner when it is exposed to the same pressure change as is shown in Figures 5–7. The concentration measurements provided by the NM vary on each simulation.

The first experiment used 7 MPa. In this condition, the mass transference velocity was fast, slightly running out the initial AA concentration that was in the package, and the expiration date was about 130 days. The second simulation had a pressure of 9 MPa. In this condition, mass transfer was not so fast as the first one, increasing the expiration date to 225 days. The third simulation took place using a pressure of 12 MPa. In this condition, the mass transference was slow in contrast to the other two conditions, running out, by day 294, AA's initial concentration on the package. According to results given by the WNSNs, it is possible to notice that the higher the embedded pressure, the higher the AA concentration in the AP's polymeric castes; at the same time the number of days of antimicrobial protection rises. The simulations show that system can detect in a very fast and accurate way the condition of antimicrobial protection existing in FP through time. In the future, this new monitoring method can integrate the current methods: temperature indicators, integrity indicators, fresh indicators, and radiofrequency indicators. To do so the WNSNs must be able to integrate different NM, even including new variables in the proposed mathematic model, or simply to broaden the information supplied by the IP. Currently these networks are in an early phase of research and development. The interconnection of nanosensors broadens the abilities of only one nanosensor; therefore the IP that uses WNSNs to monitor its packaging system will have a huge impact in the food safety.

## Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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