Food Engineering Reviews (2020) 12:346–363
https://doi.org/10.1007/s12393-020-09235-y

Strategies for Producing Improved Oxygen Barrier Materials Appropriate for the Food Packaging Sector

Maryam Zabihzadeh Khajavi[1] · **Arezoo Ebrahimi**[2] · **Mojtaba Yousefi**[3] · **Shervin Ahmadi**[4] · **Mehdi Farhoodi**[2] · **Adel Mirza Alizadeh**[2] · **Musarreza Taslikh**[2]

Received: 22 October 2019 / Accepted: 5 June 2020 /

Abstract

Flexible and transparent polymeric and bio-polymeric "super barrier" packaging materials have become increasingly important in recent years especially for oxygen-sensitive foods packaging. Different approaches and emerging technologies have been applied in order to improve oxygen barrier properties which can extend the shelf life and maintain the quality and freshness of food products during their determined shelf life. In this review, we summarize the diverse strategies for manufacturing improved oxygen barrier materials including: incorporation of nanoparticles into polymer matrix, fabrication of multilayer polymer, creation of new barrier methods such as development of crystals in polymer matrix, and cross-linking technique. The structure, preparation, and gas barrier properties of obtained polymers via mentioned approaches are discussed in general along with detailed examples drawn from the scientific literature.

Keywords Oxygen barrier · Food packaging · Nanoparticles · Nanotechnology

Introduction

There is a significant demand for packaging that exhibit excellent oxygen barrier properties for food and medicine sectors. It is vital for some food packaging to allow maintenance of the product in an appropriate condition, for instance, limited oxygen transference between packed food and atmosphere, in order to prolong product's shelf life [13, 36, 55]

High oxygen permeability of food packaging not only contributes to limited shelf life of packed food, but also leads to deterioration of food during storage and handling and reduces consumer compliance [56]. In case of food items which are rich in lipid or oil amount, holding under high O_2 concentration leads to possible chemical changes in food, for example, lipid oxidation, discoloration, and off-flavor that affect quality of packed food ([73, 102]. Oxygen can also induce deterioration of food quality by microbial spoilage which not only

✉ Shervin Ahmadi
sh.ahmadi@ippi.ac.ir

✉ Mehdi Farhoodi
Farhoodi@sbmu.ac.ir

Maryam Zabihzadeh Khajavi
Mary.khajavi72@gmail.com

Arezoo Ebrahimi
aefood89@gmail.com

Mojtaba Yousefi
m.yousefi@semums.ac.ir

Adel Mirza Alizadeh
alizadeh.sbmu@gmail.com

Musarreza Taslikh
taslikh@sbmu.ac.ir

[1] Student Research Committee, Department of Food Science and Technology, National Nutrition and Food Technology Research Institute, Faculty of Nutrition Sciences, Food Science and Technology, Shahid Beheshti, University of Medical Sciences, P.O. Box19395-4741, Tehran, Iran

[2] Department of Food Science and Technology, National Nutrition and Food Technology Research Institute, Faculty of Nutrition Sciences, Food Science and Technology,, Shahid Beheshti University of Medical Sciences, P.O. Box19395-4741, Tehran, Iran

[3] Food Safety Research Center (Salt), Semnan University of Medical Sciences, Semnan, Iran

[4] Iran Polymer and Petrochemical Institute (IPPI), PO BOX: 14965-115, Tehran, Iran

cause mechanical or chemical damage, for instance, accelerating fruit softening and aging, but also bacterial or fungal infection, which cause potential safety risks of some ready-to-eat fresh products. Furthermore, oxygen can accelerate generation of ethylene and the rate of respiration in fruits and vegetables [26, 47, 66]. Food packaging materials with high oxygen barrier properties can ensure quality maintenance of the packed product during the determined shelf life and guarantee its safety [45]. The food packaging industry requires flexible films that not only have high oxygen barrier properties but also should be transparent. In these cases, some polymers like PE, PP, PLA, PHA, PCL, PVAL, etc., have the potential to be applied for production of transparent, light, and flexible films which are appropriate for utilization in the food packaging sector. Nevertheless, their poor barrier properties toward oxygen limit their application for food packaging. Improving these poor oxygen barrier properties would be a challenge in the packaging industry. Recently, many efforts have been made to produce packaging with the least permeability to oxygen, including fabrication of a new hybrid structure, for instance, application of plastic additives and blending different polymers; introducing new crystal into the matrix of polymer; creation of a multilayer structure with an appropriate oxygen barrier material; creation of cross-linked polymer; creation of nanocomposites via incorporation of various nanoparticles, nanosheet, and tubular nano into the polymer matrix; and production of multilayer film with enhanced oxygen barrier properties [163]. The focus of this article is to represent unique new approaches for producing oxygen superior barrier performance packaging compared to the current commercial food packaging.

Principle of Oxygen Mass Transport

Barrier materials can be defined as the materials which have the ability to inhibit or slow down the passage of gases, water vapor, and organic vapor through their borders [99].

Two factors have a critical role in determining the oxygen transmission or permeability rate through the packaging material, including solubility of oxygen in the polymer matrix and its diffusion rate through the polymer matrix. The solubility of oxygen is dependent upon the chemical relationship and also the affinity between the oxygen molecule and the polymer and the rate of diffusion is affected by the size of the permeant and also polymer characteristics, especially the proportion of crystalline and amorphous regions of the polymer [39].

Oxygen transmission rate (OTR) is expressed as cubic centimeters of oxygen, which pass through a square meter of packaging material when oxygen pressure is 1 atm greater than that on the other side of the packaging during 24 h, at a specified temperature. OTR can be calculated as follows [99]:

$$\frac{\Delta mgas}{\Delta t} = P\,\frac{A\,\Delta p}{L}$$

where $\frac{\Delta mgas}{\Delta t}$ is transmission rate of oxygen, P is permeability of the packaging material, A is area contact of the packaging material, ΔP is partial pressure difference across the packaging material, and L is the thickness of the packaging material. OTR is expressed in cubic centimeters (STP)/square meters/day.

In cases that we have laminated or coated film, OTR can be calculated using the “laminate” equation [99]:

$$\text{OTR Coating} = \frac{\text{ORT}_{\text{Uncoated}} \times \ \text{ORT}_{\text{Coated}}}{\text{ORT}_{\text{Uncoated}} - \text{ORT}_{\text{Coated}}}$$

Factors Affecting Oxygen Permeability

Many factors can affect oxygen permeability rate through packaging material which can be divided into two major groups including polymer and biopolymer characteristics and environment as demonstrated in Fig. 1 [103, 125, 158].

Polymer Characteristics

Many parameters influence molecular organization of the polymer which contribute to creation of specific properties of the polymer matrix; these parameters will be discussed in the following part:

Pendent Chain

Pendent chains or side chains are oligomeric or polymeric branches which extend from the backbone chain of a polymer. Pendent chains significantly affect characteristics of the polymer, predominantly its crystallinity and density (Ryan [147]). Ghasemnejad-Afshar et al. [46] have studied the effect of side branch on gas separation performance of polyimide branched with the side groups C4H9, C3H7, CH3, and CF3. Results illustrated that membranes with larger side branch groups are more rigid. As a consequence of restriction in chain mobility, the free volume in the membrane’s structure increased which accordingly enhanced the permeation of gases into the membrane. The findings indicated that the membrane with C4H9, as the largest side branch, has the greatest diffusivity and permeation [46].

Degree of Crystallinity

The polymer matrix is consisted of two distinguishable regions: crystalline and amorphous. In crystalline regions, polymer chains are arranged in a regular, periodic manner with

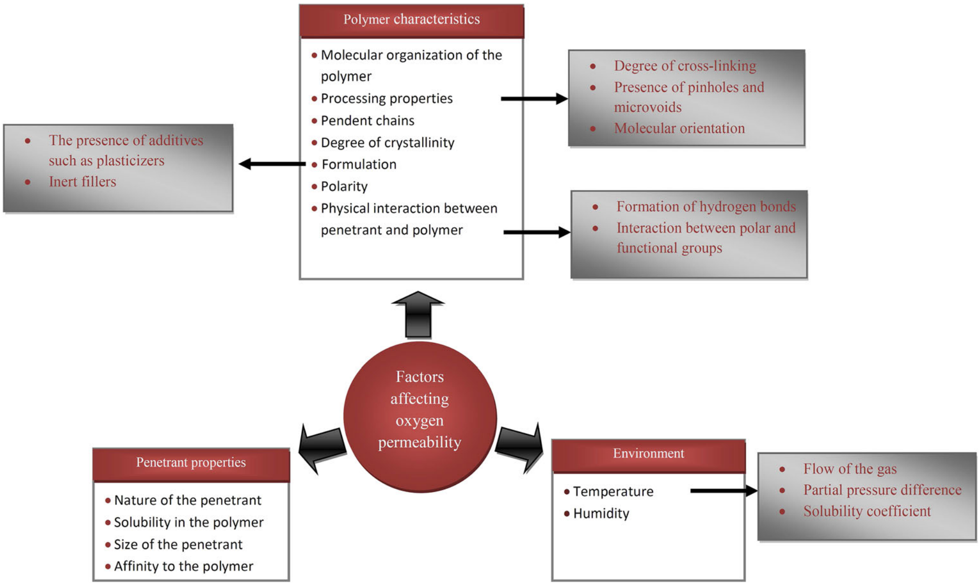

Fig. 1 Factors affecting oxygen permeability of packaging materials

strong interactions, while amorphous regions behave quite differently; in these regions, polymer chains are arranged irregularly and have less density and present more free volumes compared to crystalline regions. Since the crystallites are impermeable, permeant molecule must seek out amorphous zones in order to pass through the polymer matrix. Therefore, a polymer with higher degree of crystallinity compared to the polymer with fewer degree of crystallinity provide less amount of permeation [152]. It has also been demonstrated that the arrangement of crystalline lamellae in semicrystalline polymers can significantly influence gas barrier characteristics. In particular, when the arrangements of the lamellae stacks are perpendicular to the direction of gas diffusion, the highest barrier properties will be achieved. In a study, the self-assembly nucleator approach was applied to design the arrangement of crystalline lamellae in the PLA matrix. Octamethylenedicarboxylic dibenzoylhydrazide (TMC-300) and graphene were used as active nucleators for PLA. A multilayer sheet consisting of PT (PLA+TMC-300) alternating with PTG (PLA+TMC-300+graphene) was prepared by multilayer coextrusion. Under isothermal processing as a result of the induced impact of graphene, the melted TMC-300 self-assembles into solid-state fibrils that were perpendicular to PT/PTG layered interface. As a result, a reduction of 85.4% in O2 permeability was achieved compared to the blended control sample [88–90]. Gupta et al. [52] have studied the effect of modified chitosan on the oxygen permeability of PDLA/PLLA blend and pristine PLA. The results indicated that incorporation of 3wt% modified chitosan into blended polymer increased the degree of crystallinity. Consequently, oxygen permeability reductions have been found to be nearly 56 and 84%, respectively, compared to blended PDLA/PLLA and pristine PLA ([52](.

Formulation

It should be taken into consideration that formulation of the compounds has a key role in molecular arrangement and properties of final produced polymer particularly in the case of biomaterials such as polysaccharide- and protein-based materials, which have poor oxygen barrier properties [15, 27, 98, 111, 166]. For instance, the presence of additives such as plasticizers can affect permeability of packaging. For some polymers, it is essential to use plasticizer in order to achieve desired properties, but plasticizers generally increase the permeability of the polymer. Zhang et al. [164] have investigated the impacts of glycerol and sorbitol as plasticizer on the oxygen permeability of edible film based on gum ghatti (GG). The results revealed that oxygen permeability of films marginally increased with increasing glycerol concentration. Whereas, increasing sorbitol concentration had no significant effect on oxygen permeability. SEM micrograph showed that the structure of glycerol-GG films was more regular and smoother compared to sorbitol-GG films. It was also indicated that crystallinity degree decreased with increasing glycerol concentration [164]. On the other hand, incorporation of some filler like nanoparticles into the polymer matrix can enhance its barrier properties. Especially, the insertion of fillers that have high adhesion and compatibility with the polymer matrix can improve barrier properties of the packaging [122].

Processing Properties

The results of processing conducted during production of polymer can noticeably influence its permeability properties. For instance, creation of pinholes and microvoids contributed to the higher transmission rate of gases, since gas molecules require free volume for movement and diffusion through the polymer matrix [75]. Processing which initiates higher degree of cross-linking or increases molecular orientation in polymer will improve barrier properties, since these approaches further increase density and crystallinity of polymer, which make the path to permeate more difficult. As an illustration, graphene oxide nanosheet/cellulose nanofiber (GONS/CNF) nanocomposite film with ultra-low oxygen permeability was manufactured through layer-by-layer coating. In this procedure, GONS and CNF were dispersed into distilled water with ultrasonic treatment. The achieved mixture was repeatedly coated onto a glass plate until a film with a thickness of approximately 30–40 μm was manufactured. The images of SEM and GI-WAXS manifested that the exfoliated GONS and CNF were highly oriented along the film direction, which is the reason of ultra-low oxygen permeability (F. [137]).

Physical Interaction between Penetrant and Barrier Material

Some interactions between penetrant and barrier material can lead to trapping of diffusing molecule and slow down the permeation rate. For instance, presence of polar or functional groups that cause interaction between polymer and penetrant or hydrogen bonds formation will also trap diffusing molecules [31, 41]. Paine [118] investigated the oxygen uptake capacity of three polymers, including poly (cyclohexylmethyl methacrylate) (CHMA), poly (cyclohexenylmethyl acrylate (CHAA), and polyvinylidene fluoride (PVDF). The findings of this study showed that both polyCHMA and polyCHAA have greater capacity for oxygen uptake compared to PVDF. One possible explanation for this could be the existence of cyclic ring units, which are capable of scavenging oxygen molecules [118].

Environment

External factors such as temperature and humidity also can influence the value of permeability.

Temperature

Penetrant molecules in order to diffuse through the polymer matrix require enough energy to overcome the interactions that exist between polymer chains. On the other hand, increasing temperature causes more motion in polymer chains and creates free volume, which accelerate oxygen transmission rate. The transport equation (which stated was in section 2) is affected by increasing temperature in two ways, including the flow of gas and the partial pressure difference [99]. Öner et al. [113] have investigated the influence of temperature on oxygen-barrier performance of nanobiocomposites based on poly (3-hydroxybutyrate-*co*-3-hydroxyvalerate) and boron nitride (PHBV/BN). The PHBV nanocomposites showed an increase in oxygen permeability as temperature was increased, with an Arrhenius behavior, as was expected (M [113]).

Humidity

Since absorbed water could have a plasticizing effect on some packaging materials, high humidity can increase packaging permeability. Exposure of polymer to an environment with high level of humidity or product with high moisture content can also lead to swelling of the polymer which will increase permeability. In the case of polar polymers like cellophane and PVA, contact with high humidity has a plasticizing effect on them that disturbs their barrier properties [39]. For instance, chitosan-based films exhibit good oxygen barrier but are degraded when exposed to high humidity, due to their great water solubility [86]. In an investigation, the impact of moisture on the oxygen permeability of polyvinyl alcohol (PVOH) and PVOH–kaolin dispersion barrier coatings was examined. The oxygen permeability was measured at different humidity levels, and the material properties were characterized under the same conditions, including polymer crystallinity, kaolin concentration, and kaolin orientation. The experimental results indicated that the water is able to plasticize the PVOH material of the coatings, and the incorporation of kaolin as filler failed to inhibit or overcome such phenomenon substantially. It was also revealed that the crystallinity degree of the PVOH was affected dramatically by the humidity, since water melts polymer crystallites, which further increased oxygen permeability [112].

Collected Comparative Oxygen Barrier Properties of Plastic

In Fig. 2. oxygen and water vapor barrier properties of some commercial petro- and bio-based materials for food packaging have been illustrated, which were measured at 23 °C and 85% RH.

Novel Approaches for Producing High Oxygen Barrier Packaging Materials

Nanocomposite

In the last few decades, polymer nanocomposites, in the field of nanotechnology, has attracted enormous attention because of their unique characteristics, especially gas barrier properties [94, 108, 132]. It has been shown that incorporation of nanofillers into the polymer matrix improves oxygen barrier properties [5, 61], since these nanofillers are impermeable and do not permit diffusion of gas molecules and make them to follow a longer and more torturous pathway to pass through

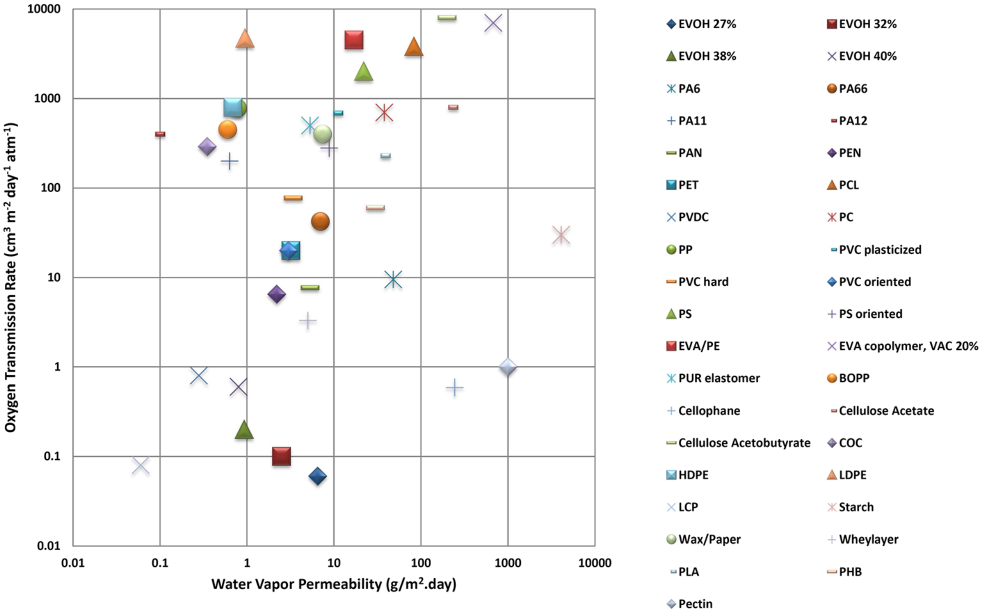

Fig. 2 Oxygen and water vapor barrier properties of some commercial petro- and bio-based materials for food packaging [14, 101]

the nanocomposite [34, 37, 100] as illustrated in Fig. 3. Furthermore, nanofillers insertion in the polymer matrix increases crystallinity degree which acts as an obstacle and does not allow the permeation of gas molecules through them. It has also been indicated that the molecular orientation of polymer chains that are around the nanofillers will be affected and contributed to formation of a restricted or rigid amorphous fraction which has lower permeability and further enhances barrier properties [68, 143]. Three parameters have a critical role in determining barrier properties of nanocomposite including filler characteristics (volume fraction, aspect ratio), the intrinsic barrier property of the polymer matrix, and the state of nanofillers dispersion in the polymer matrix (intercalation, exfoliation, flocculation or agglomeration, specific interface, free volume and in the case of nanosheets the orientation of nanoplatelets) [107]. The fundamental point of manufacturing nanocomposites is that we can use commercially available polymers for making high oxygen barrier films. Therefore, it is a cost-effective and practical procedure to reach uncomplicated and scale-up manufacturing and thus has attracted intensive attention from both academic and industrial fields (Y. [91]). In a study by Ramos et al. [134], nano-biocomposite films based on PLA were prepared by incorporating modified montmorillonite (D43B) at two different concentrations of 2.5 and 5 wt%. It was reported that D43B at both concentrations reduced the oxygen transmission rate by formation of intercalated nanocomposites [134]. In another study, the octadecylamine (ODA) modified graphene oxide (mGO-ODA)/maleic anhydride grafted polypropylene (MAPP) nanocomposites (mGO-ODA/MAPP) were prepared

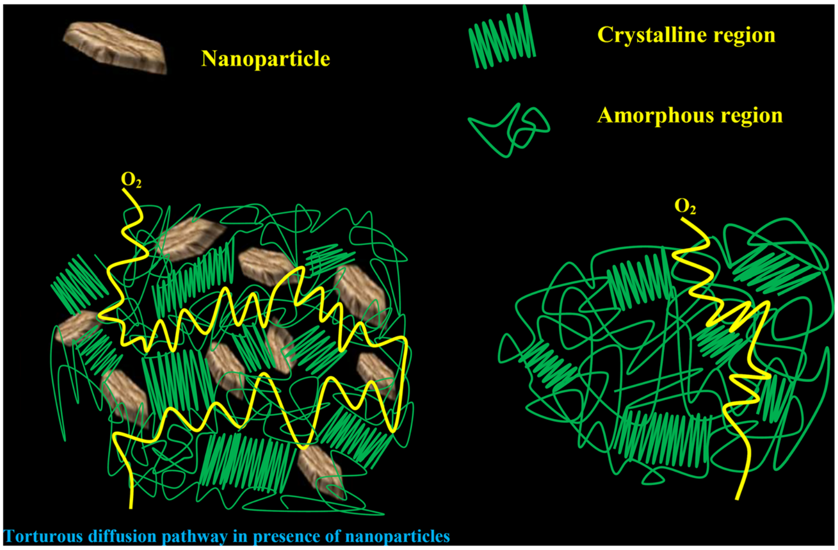

Fig. 3 Torturous pathway of diffusing molecules around nanoparticles

and applied successfully as gas barrier materials. The result indicated that 60% mGO-ODA/MAPP exhibited 94.1 and 95.0% reduction for H_2GTR and O_2GTR, respectively, compared to the pure nylon film [90]. In another study, PLA/titanium oxide nanocomposite films were prepared by the casting method. The effect of titanium oxide nanoparticles and its concentration (1, 3, and 5wt %) on oxygen transmission rate of nanocomposites were investigated, and it was shown that oxygen transmission rate was decreased when the concentration of titanium oxide was increased [1]. Also, in the case of PLA as a biodegradable polymer, it has been proved in many researches that incorporation of nanocellulose in the PLA matrix will enhance oxygen barrier properties. These kinds of bio-based hydric polymers have semicrystalline behavior, which makes them appropriate materials for food packaging [78, 92, 109].

Cheng et al. [23] manufactured biodegradable films as packaging materials that could act as barrier for oxygen, by mixing a guar gum (GG) solution with a nanocrystalline cellulose (NCC) dispersion using a novel circular casting technology [23]. In a study, an active nanocomposite of Halloysite nanotubes and PE was prepared as ethylene scavenging packaging, but it was also found that oxygen barrier properties of nanocomposite was improved compared to neat PE [146]. It was demonstrated that the antimicrobial nanocomposite film of HDPE/cu-nanofiber, which were prepared by the melt mixing method, also showed better barrier properties toward oxygen compared to neat HDPE [12]. Majeed et al. have prepared montmorillonite (MMT)/rice husk (RH) hybrid filler-filled LDPE nanocomposite films, containing 0, 2, 3, 4, 5, and 6 wt% MMT (based on the total weight) by extrusion blown film. The findings of this study showed that addition of MMT into the LDPE/RH system improved the O_2 barrier properties [97]. It was shown that the oxygen permeability coefficient of GONS/PVA nanocomposite at a low GONS loading of 0.72 vol% was declined about 98% (H.-D. [48, 64] have demonstrated that incorporation of different montmorillonite type (hydrophylic vs. organically modified) into chitosan/PVOH blends enhances its oxygen barrier properties. In an investigation, the effect of encapsulating the polymer within a nanoplatelet shell on oxygen permeability was examined. The Pickering suspension polymerization method was applied for manufacturing few-layered GO nanoplatelets encapsulated polystyrene (PS) microparticles. The oxygen permeability was reduced by 96 and 34% in obtained PS/GO composite film containing 2 wt% of GO, respectively, compared to the PS control film and the solution mixed PS/GO composite film [101]. Risyon et al. [138] have developed polylactic acid (PLA)/halloysite nanotubes (HNTs) bionanocomposite films for extending the shelf life of packaged cherry tomatoes. The PLA/HNTs bionanocomposite films were demonstrated to have great oxygen barrier properties especially at 3.0 wt% of HNTs and had the potential to prolong the shelf life of cherry tomatoes [138]. Vaezi et al. [149] examined the oxygen barrier properties of a bionanocomposite based on cationic starch (CS)/montmorillonite (MMT)/nanocrystalline cellulose (NCC). CS nanocomposites with 5 wt% NCC and MMT showed the best improvement in the barrier properties against oxygen and also water vapor [149]. Pengwu et al. have developed a high barrier polyhydroxyalkanoates (PHA) nanocomposite by compounding graphene oxide grafted by long alkyl chain quaternary salt (GO-g-LAQ). The GO-g-LAQ was capable of improving the interfacial adhesion between GO and PHA due to its hydrophobic nature. As a result of the condensed crystal structure of PHA and the impermeable property of GO sheets, PHA/GO-g-LAQ nanocomposite showed improved oxygen barrier property [157]. The impact of different types of boron nitride particles (BNPs) including silanized flake type BN (OSFBN) and silanized hexagonal disk type BN (OSBN) on oxygen permeability of poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) was examined. As a consequence of higher crystallinity degree in the presence of BN particles, all the obtained nanocomposites had lower oxygen permeability compared to the neat PHBV. The best barrier properties was obtained by incorporating 2 wt% OSFBN, for which a reduction of oxygen permeability up to 36% was observed in comparison to the neat PHBV (Mualla [114]). The influence of nanofillers on the oxygen barrier properties of some nanocomposites prepared by different methods is summarized in Table 1. It should be noted that the values of the permeability coefficients in the literature are often given in different units of measurement. To facilitate the comparison of gas permeability, the values of gas barrier properties reported in the literatures have been converted into the same units (m^2 s^{-1} Pa^{-1}). As evidenced, incorporation of scarce amount of graphene oxide has resulted in significant reduction in oxygen permeability compared to O-MMT and MMT. As an illustration, application of 0.4 wt% graphene oxide into the PLA matrix contributed to 68% reduction in oxygen permeability, while incorporation of 7.9 wt% O-MMT caused only 24% reduction, which indicated the critical role of the nanofiller type. In addition, apart from the polymer kind, in most of cases, by increasing nano amount, greater oxygen permeability reduction was achieved.

Multilayer Polymer

Multilayer polymer packaging had gained a lot of attention due to its excellent gas barrier properties. The first multilayer films were introduced in the food packaging industry by polymers coated with thin metal films, known as metalized plastics. Metalized plastics have been utilized in the food industry since the early 1970s [142]. The main problem associated with metalized packaging is their opaqueness, which limits their

Table 1 Oxygen permeability of polymer/nanocomposites

Polymer	Type of nano	Nano loading	Processing	Oxygen permeability (m^2 s^{-1} Pa^{-1})	Oxygen permeability reduction (%)	Reference
SBR	O-MMT, CF	16 phr	Melt intercalation	2.75×10^{-17}	46	[11]
PLA	O-MMT	7.9 wt%	Melt intercalation	2.85×10^{-18}	24	[123]
PLA, PLA/PCL	O-MMT	3 wt%	Melt intercalation	6.39×10^{-19}	26	[140]
PA	MMT	3 wt%	Melt intercalation	3.37×10^{-19}	14	[43]
PET	O-MMT	5 wt%	Melt intercalation	1.69×10^{-19}	55	[141]
PET	O-MMT	5 wt%	Melt intercalation	2.74×10^{-19}	69	[54]
PET	O-MMT	1 wt%	Melt intercalation	1.42×10^{-17}	45	[40]
PP	O-MMT	4 vol%	Melt intercalation	4.18×10^{-18}	46	[106]
PP	O-MMT	7.5 wt%	Melt intercalation	2.59×10^{-15}	56	[104]
PP	MMT, O-MMT	5 wt%	Melt intercalation	$\sim 6.00\times 10^{-18}$	77	[151]
HDPE	MMT, O-MMT	3 vol%	Melt intercalation	3.05×10^{-18}	35	[116]
LDPE	O-MMT	7 wt%	Melt intercalation	2.86×10^{-17}	24	[6]
LLDPE	O-MMT	5 phr	Melt intercalation	1.81×10^{-17}	55	[33]
LLDPE	VER, O-VER	3 wt%	Melt intercalation	1.07×10^{-20}	18	[18]
PET, MXD6	MMT	2 wt% 3 wt%	Mel intercalation	1.89×10^{-17} 6.89×10^{-19}	51.7 70.3	[153]
PET-PA/MXD6	O-MMT	3.5 wt%	Melt intercalation	2.27×10^{-19}	21	[32]
LLDPE PVC	MMT, O-MMT	5 wt% 10 wt%	Melt intercalation	1.25×10^{-17} 0.731×10^{-17}	58 65	[76]
Polyester	O-MMT	2.5 wt%	Melt intercalation	$\sim 7.83 \times 10^{-20}$	63	[10]
Paraffinic wax	O-MMT	2.5 wt%	Melt intercalation	3.14×10^{-18}	99.7	[19]
EVOH, PLA	Kaolinite	4 wt% 4 wt%	Melt intercalation	$< 1 \times 10^{-21}$ 6×10^{-19}	75 45	[82]
PI	O-MMT	3 wt%	Solution intercalation	2.33×10^{-18}	30	[63]
XG	MMT	20 wt%	Solution intercalation	5.79×10^{-22}	89	[79]
PVA, NFC	MMT	50 wt%	Solution intercalation	5.71×10^{-22}	99.8	[144]
NFC	MMT	50 wt%	Solution intercalation	4.00×10^{-19}	80	[93]
Chitosan	MMT	5 wt%	Solution intercalation	$\sim 1.1 \times 10^{-18}$	~50	[60]
PP/EPDM	O-MMT	5 wt%	Solution intercalation	2.55×10^{-17}	62	[120]
Epoxy	O-MMT	5 vol%	Solution intercalation	4.79×10^{-20}	75	[117]
EP	O-MMT	7 wt%	In situ polymerization	2.34×10^{-18}	86.1	[30]
PS	MMT, O-MMT	16.7 wt%	In situ polymerization	5.03×10^{-18}	64	[110]
PLA	GONS	1.37 vol%		1.145×10^{-18}	45	[65]

Table 1 (continued)

Polymer	Type of nano	Nano loading	Processing	Oxygen permeability (m^2 s^{-1} Pa^{-1})	Oxygen permeability reduction (%)	Reference
			Solution intercalation			
Cellulose	GNPs	5 wt%	Solution intercalation	$\sim 0.8 \times 10^{-18}$	~27	[96]
HDPE	DA-GO, DA-RGO	1 wt%	Solution intercalation	1.75×10^{-18}	67	[136]
PLA	Graphene oxide, GNP	0.4 wt%	Solution intercalation	1.2×10^{-18}	68	[124]
PVA	GONS	0.72 vol%	Solution intercalation	0.24×10^{-19}	98.9	[64]
EVOH	TRG	0.5 wt%	Solution intercalation	8.517×10^{-19}	99.98	[160, 161]
PVA	Graphene oxide	0.07 vol%	Solution intercalation Recrystallization	$< 5.0 \times 10^{-24}$	> 99.94	[22]
PU	RGO	2 wt%	Melt intercalation	0.309×10^{-18}	90.5	[44]
XNBR	Graphene oxide	1.9 vol%	Latex co-coagulation	$\sim 1.5 \times 10^{-17}$	55	[77]
PLA	Graphene oxide	0.1 wt%	solution intercalation	2.513×10^{-18}	33	[150]
PLA	Graphene oxide	0.3 wt%	solution intercalation	2.627×10^{-18}	30	[150]
PLA	Graphene oxide	0.5 wt%	solution intercalation	3.4×10^{-18}	9	[150]
PLA	Silk nano-disc	0.5 wt%	Melt intercalation	1.68×10^{-18}	50	[121]
PLA	Silk nano-disc	1 wt%	Melt intercalation	0.92×10^{-18}	72.4	[121]
PLA	Silk nano-disc	2 wt%	Melt intercalation	1.45×10^{-18}	56.9	[121]
PLA	Silk nano-disc	5 wt%	Melt intercalation	1.04×10^{-18}	68.9	[121]
Thermoplastic starch	Cellulose nanofibers	15 wt%	Solution intercalation	0.58×10^{-18}	48.68	[133]
Thermoplastic starch	Cellulose nanofibers/graphene oxide	15, 3 wt%	Solution intercalation	0.38×10^{-18}	66.45	[133]
Cassia-gum	C-CNCW	2 wt%	Solution intercalation	2×10^{-19}	5.58	[16]
Cassia-gum	C-CNCW	4 wt%	Solution intercalation	1.98×10^{-19}	19.21	[16]
Cassia-gum	C-CNCW	6 wt%	Solution intercalation	1.93×10^{-19}	18.68	[16]

application. Recently, the demand for transparent packaging materials with equivalent gas barrier properties to metalized plastics has increased significantly, since they allow content visibility. Furthermore, elimination of metallic components provides better recyclability and microwave compatibility [81]. In the following parts, we will discuss about different approaches of making transparent multilayer films.

Thin Metal Oxide Films

The first alternative for metalized plastics was transparent silicon oxide (SiO_x) coating which, besides providing oxygen barrier properties, also prevents from moisture ingress or aroma loss. In the 1980s, application of these thin films in the food packaging sector became very popular. Although thin metal oxide films exhibit good oxygen barrier properties, these coatings fall short of their predicted performance, mainly due to microscopic defects like pinholes that form during the deposition process and also their surface roughness. It has been shown that barrier trends logarithmically with surface roughness and smoother films illustrate a better oxygen barrier characteristic [29, 57].

Hybrid Coating

An innovative approach to achieving enhanced barrier properties of polymers can be presented by coating micro-layers of material with appropriate gas barrier properties on them. Various materials from natural or synthetic bases can be utilized for coating. For instance, in a study, modified whey protein was coated on PLA in order to improve its oxygen barrier properties. Comparison of neat PLA versus PLA coated with whey protein isolate showed improvement of about 90% in the oxygen barrier properties [155]. In another study, cellulose nanocrystals, which were obtained from cotton linters and Kraft pulp, were coated on PET to enhance PET oxygen barrier characteristics. The result illustrated that the oxygen permeability value of coated PET was hundreds of times lower than pure PET over a board range of time [135]. In a study, a biodegradable PLA-based hybrid coating material was obtained via coating PLA film by prepared PLA/SiO2 hybrids. In this study, 3-isocyanatopropyltriethoxysilane was employed as a silane coupling agent in order to promote adhesion between dissimilar materials (silica and PLA). Oxygen barrier properties of coated PLA improved by 69.7% compared to neat PLA [9]. High-strength regenerated cellulose/attapulgite (ATT) composite films (RC/ATT) with good oxygen barrier performance were prepared from cellulose/LiOH/urea solutions with different ATT contents ranging from 5 to 20 wt%. The RC/ATT composite films exhibited relative low oxygen permeability below 0.5 cm^3 μm/day m^2 kPa, which could even reach 0.32 cm^3 μm/day m^2 kPa with 20 wt% ATT content (C. [154]). In a study, functional antimicrobial LDPE films with coatings containing different amounts of pyrogallol (PGL), a natural phenolic substance, and polyurethane were prepared. Coatings with pyrogallol caused the barrier properties for water, and oxygen was increased from 0.78–0.32 to 470 ± 23.2–273 ± 57.1 (g mm)/(m^2 h kPa), respectively. These findings indicate that the barrier properties of the LDPE/PGL films were highly improved compared to those of neat LDPE [42, 25] have explored the mass transfer of renewable films based on gelatin (Ge), glycerol (Gly), and epoxidized soybean oil (ESO) for application in food packaging. The results illustrated that gelatin sample containing 20% Gly and 20% ESO present appropriate gas barrier properties [25]. In another study, the oxygen transfer rate (OTR) and water vapor permeability (WVP) of polyethylene terephthalate (PET) films were adjusted via coating of polyphenols and gelatin mixture (PGM) with different concentrations while maintaining the other properties of modified PET films. The results showed that OTR was decreased (63.5±0.02 to 38.1±0.03g/in^2/day) with respect to uncoated film (82 ± 3.5) [69]. Lu et al. [95] reduced the oxygen permeability of biaxially oriented PP/LDPE film by coating nanofibrillated cellulose (NC) solution on it. The result showed that oxygen transmission rate of the coated film was as low as 24.02 cc/m^2/day compared to non-coated film (67.03 cc/m^2/day) [95].

Layer by Layer Assembly

Layer by layer assembly has received extensive, worldwide attention over the past two decades, due to its relatively inexpensive deposition technique and typically for producing multifunctional thin film such as enhanced gas barrier properties film which has less than a micron thick ([4]; M A [130]). Generally, the following steps are done in order to prepare the LBL film as shown in Fig. 4: often the negatively charged substrate is immersed into a mixture of positively charged material (polyelectrolytes or nanoparticles) for a given amount of time, ranging from seconds to tens of minutes. The substrate is then removed from the mixture, rinsed with deionized water, optionally dried in a stream of filtered air (or nitrogen), and then immersed into a mixture of negatively charged material, rinsed and dried. This simple procedure is then repeated to deposit a given number of cationic and anionic pairs layers [131]. Individual layers can range from angstroms to hundreds of nanometers in thickness, although many factors can influence their thickness, including pH, buffer, ionic strength, temperature, the molecular weight of the deposition species, and the relative humidity of the fabrication environment [2, 51].

Layer by Layer Assembly of Polymers Much of the early LbL gas barrier work was done using only polymers, rather than particle-filled or nanostructural systems. Leväsalmi and McCarthy [87] showed that layered structural films which consisted of PAH (a strong poly cation) and PSS (a strong poly anion) have good gas barrier properties due to its dense ionically crosslinked structure [87]. Yang et al. [159] have also reported that the multilayer film of PEI and PAA form a dense network, which has super gas barrier performance (Y.-H. [159]). In a study, the LBL approach was applied to produce a biodegradable multilayer film of sodium alginate/PEI on biaxially oriented PLA. Oxygen permeability of the prepared film was found to be three orders of magnitude less than the uncoated biaxially oriented PLA film [50]. In another study, a multilayer film of fish gelatin/PLA was prepared in order to reduce oxygen permeability. It was found that the oxygen permeability value of the multilayer film reduced more than eightfold compared to pure PLA film [62]. Joo et al. have developed whey protein isolate (WPI)-coated multilayer films using PET film as a substrate. In order to improve the interfacial compatibility between PET film and water-based WPI coating solution, various surface pretreatments (corona discharge, plasma, and primer coating) were applied to PET. Oxygen transmission rates of surface-pretreated

Fig. 4 Procedure of layer by layer assembly in production of multilayer films

multilayer films with WPI coating layer [PET/WPI/nylon/LLDPE] were significantly lower, about 43–234 times, than the multilayer films without WPI film layer [74]. Apicella et al. [3] developed a multilayer film based on PET polymer, which indicated excellent barrier properties toward oxygen. In this investigation, an active layer of PET containing 10 wt% polymeric oxygen scavenger, named Amosorb DFC 4020E, were sandwiched between two layers of neat PET polymer. [3].

Layer by Layer Assembly of Polymers and Nanomaterials LbL films exhibit unique properties when incorporated with nanoparticles. Nanoparticle-based LbL films have super barrier gas properties [119]. In a study, a polymer/nanoclay multilayer ultrathin film with only 52 nm thickness was prepared via LBL assembly of 12 polymer and 4 clay layers. The obtained film presented oxygen permeability orders of magnitude even lower than EVOH and SiO_x [126, 127]. In another study, a quadlayer film which consisted of CH/CR/CH/MMT was deposited on PET film, using the LBL technique. This multilayered thin film was able to reduce the oxygen permeability value of PET by two orders of magnitude compared to pure PET film, under the same conditions (Galina [85]). In another study, a high barrier multilayer film based on PET substrate was achieved by LBL assembly of similarly cationic charged layers of PEI and successive anionic charged layer of clay. Transmission electron microscopy images of the prepared film illustrated a nanobrick wall structure of clay nanoplatelets within the polymeric mortar, which resulted in enhanced oxygen barrier properties [53]. The impact of deposition of multilayered hybrid thin film composed of cellulose nanocrystals (CNCs) and gibbsite nanoplatelets (GNPs) onto different selected substrates on the oxygen barrier properties was examined. The substrates were an uncoated kraft cardboard, a polyethylene-coated cardboard, a low density virgin PE, and a smart paper. The results revealed that the oxygen barrier properties of all the substrates were significantly improved after the deposition of thin multilayered hybrid film [20]. Other studies related to oxygen barrier properties of multilayer film are summarized in Table 2. As illustrated in majority of cases, the reduction in oxygen permeability was higher than 90% that reveals the efficiency of multilayer film in improving oxygen barrier properties. It is worth mentioning that multilayer films compromising of polymer and nanofiller demonstrated enhanced oxygen barrier characteristics, which are analogs to multilayer films containing SiOx coating.

Introducing New Crystals Phase or Shape in the Polymer Matrix

It has been shown that increasing the degree of crystallinity contributed to enhancement of gas barrier properties of packaging, since crystallites act as impermeable obstacles and make the diffusing molecules to take a longer pathway around them. It has been also illustrated that crystalline zones can influence their surrounding polymer chains and cause formation of a restricted or rigid amorphous fraction which has lower chain movement and improved oxygen barrier

Table 2 Oxygen permeability of multilayer polymer

Type of layers	Processing method	Oxygen permeability	Oxygen permeability or OTR reduction (%)	Reference
PE with SiO_x coating	Plasma enhanced chemical vapor deposition	$300\ cm^{3}\ m^{-2}\ day^{-1}\ atm^{-1}$	96.48	[38]
PA with SiO_x coating	Plasma enhanced chemical vapor deposition	$0.5\ cm^{3}\ m^{-2}\ day^{-1}\ atm^{-1}$	99.26	[38]
OPP with SiOx coating	Plasma enhanced chemical vapor deposition	$9\ cm^{3}\ m^{-2}\ day^{-1}\ atm^{-1}$	99.64	[97]
OPP with SiOx coating	Physical vapor deposition	$218\ cm^{3}\ m^{-2}\ day^{-1}\ atm^{-1}$	91.28	[105]
PET with SiOx coating	Physical vapor deposition	$2.64\ cm^{3}\ m^{-2}\ day^{-1}\ atm^{-1}$	97.8	[139]
PET with SiOx coating	Physical vapor deposition	$2.18\ cm^{3}\ m^{-2}\ day^{-1}\ atm^{-1}$	98.18	[80]
PET with SiOx coating	Plasma enhanced chemical vapor deposition	$1\ cm^{3}\ m^{-2}\ day^{-1}\ atm^{-1}$	99.16	[38]
PET with SiOx coating	Plasma enhanced chemical vapor deposition	$0.66\ cm^{3}\ m^{-2}\ day^{-1}\ atm^{-1}$	99.45	[70]
PET with SiOx coating	Plasma enhanced chemical vapor deposition	$0.44\ cm^{3}\ m^{-2}\ day^{-1}\ atm^{-1}$	99.63	[35]
PET with SiOx coating	Plasma enhanced chemical vapor deposition	$0.29\ cm^{3}\ m^{-2}\ day^{-1}\ atm^{-1}$	99.75	[72]
PET with SiOx coating	Plasma enhanced chemical vapor deposition	$0.1\ cm^{3}\ m^{-2}\ day^{-1}\ atm^{-1}$	99.9	[67]
PAAm, PAA with MMT nanofiller	Layer by layer assembly	$< 1 \times 10^{-25}\ (m^{2}\ s^{-1}\ Pa^{-1})$	99.8	[129]
PVAm, PEI, PAA with MMT nanofiller	Layer by layer assembly	$< 5 \times 10^{-26}\ (m^{2}\ s^{-1}\ Pa^{-1})$	99.94	[148]
Chitosan with MMT nanofiller	Layer by layer assembly	$3 \times 10^{-22}\ (m^{2}\ s^{-1}\ Pa^{-1})$	99.98	[84]
PVP with MMT nanofiller	Layer by layer assembly	$6.2 \times 10^{-22}\ (m^{2}\ s^{-1}\ Pa^{-1})$	64	[58]
PEI, PAA with 26.2 wt% MMT nanofiller	Layer by layer assembly	$< 0.9 \times 10^{-25}\ (m^{2}\ s^{-1}\ Pa^{-1})$	99.94	[126, 127]
PEI, PAA with LAP nanofiller	Layer by layer assembly	$8.3 \times 10^{-24}\ (m^{2}\ s^{-1}\ Pa^{-1})$	99.4	[156]
PEI, PAA with MMT nanofiller	Layer by layer assembly	$6.4 \times 10^{-25}\ (m^{2}\ s^{-1}\ Pa^{-1})$	93.2	[156]
PEI, PAA with VMT nanofiller	Layer by layer assembly	$3.3 \times 10^{-25}\ (m^{2}\ s^{-1}\ Pa^{-1})$	99.8	[156]
PEI, PAA with MMT nanofiller	Layer by layer assembly	$1.21 \times 10^{-25}\ (m^{2}\ s^{-1}\ Pa^{-1})$	99.94	[53]
PEI with more than 84 wt% nanofiller	Layer by layer assembly	$< 2.28\times 10^{-25}\ (m^{2}\ s^{-1}\ Pa^{-1})$	99.94	[126, 127]
PGD, PEI with MMT nanofiller	Layer by layer assembly	$3.50 \times 10^{-20}\ (m^{2}\ s^{-1}\ Pa^{-1})$	71	[59]
PEI with MMT nanofiller	Layer by layer assembly	$< 1 \times 10^{-25}\ (m^{2}\ s^{-1}\ Pa^{-1})$	99.94	[128]
PAM with MMT nanofiller	Layer by layer assembly	$< 6.51\times 10^{-19}\ (m^{2}\ s^{-1}\ Pa^{-1})$	99.94	[71]
BPEI Graphene oxide with nanofiller	Layer by layer assembly	$< 0.05\ cm^{3}\ m^{2}\ day^{-1}$	99.6	[21]
PEI with graphene oxide nanofiller	Layer by layer assembly	$0.05\ cc/m^{2}\ day$	99.4	[162]
PEI with Graphene oxide nanofiller	Layer by layer assembly	$0.12\ cc/m^{2}\ atm\ day$	99	[161]
Graphene oxide film sandwiched between two layers of PLA	Layer by layer assembly	$7.08 \times 10^{-20}\ (m^{2}\ s^{-1}\ Pa^{-1})$	97.2	[49]
Reduced graphene oxide film sandwiched between two layers of PLA	Layer by layer assembly	$2.63 \times 10^{-20}\ (m^{2}\ s^{-1}\ Pa^{-1})$	99%	[49]
Chitosan and MMT films coated on PLA	Layer by layer assembly	$2.97 \times 10^{-20}\ (m^{2}\ s^{-1}\ Pa^{-1})$	98	[145]
20 bilayers of BPEI and Nafion deposited on PLA	Layer by layer assembly	$1.32 \times 10^{-18}\ (m^{2}\ s^{-1}\ Pa^{-1})$	22.15	[17]

Table 2 (continued)

Type of layers	Processing method	Oxygen permeability	Oxygen permeability or OTR reduction (%)	Reference
4 quadlayers of BPEI/Nafion/BPEI/MMT deposited on PLA	Layer by layer assembly	0.7×10^{-18} ($m^2\ s^{-1}\ Pa^{-1}$)	59.73	[17]
6 quadlayers of BPEI/Nafion/BPEI/MMT deposited on PLA	Layer by layer assembly	0.24×10^{-18} ($m^2\ s^{-1}\ Pa^{-1}$)	85.91	[17]
8 quadlayers of BPEI/Nafion/BPEI/MMT deposited on PLA	Layer by layer assembly	4.57×10^{-20} ($m^2\ s^{-1}\ Pa^{-1}$)	97.31	[17]
10 quadlayers of BPEI/Nafion/BPEI/MMT deposited on PLA	Layer by layer assembly	2.28×10^{-20} ($m^2\ s^{-1}\ Pa^{-1}$)	98.66	[17]
4 hexalayers of $(BPEI/Nafion)_2$/BPEI/MMT deposited on PLA	Layer by layer assembly	0.12×10^{-18} ($m^2\ s^{-1}\ Pa^{-1}$)	92.62	[17]
6 hexalayers of $(BPEI/Nafion)_2$/BPEI/MMT deposited on PLA	Layer by layer assembly	2.97×10^{-20} ($m^2\ s^{-1}\ Pa^{-1}$)	98.25	[17]
8 hexalayers of $(BPEI/Nafion)_2$/BPEI/MMT deposited on PLA	Layer by layer assembly	3.99×10^{-20} ($m^2\ s^{-1}\ Pa^{-1}$)	97.65	[17]
10 hexalayers of $(BPEI/Nafion)_2$/BPEI/MMT deposited on PLA	Layer by layer assembly	2.4×10^{-20} ($m^2\ s^{-1}\ Pa^{-1}$)	98.6	[17]
BPEI/NFC film coated on PLA	Layer by layer assembly	0.1×10^{-18} ($m^2\ s^{-1}\ Pa^{-1}$)	92	[7]

properties compared to amorphous fraction as illustrated in Fig. 5 [143]. In a study, combined techniques of solution blending and isothermal recrystallization were applied for formation of film comprised of PVA and GO, in order to introduce new crystals into the polymer mortar. It was found that during isothermal recrystallization process, GO sheets act as nucleating agents and contributed to formation of PVA crystals around the GO sheets. Transmission electron microscopy images revealed that newly formed PVA crystals have filled the empty spaces that existed between GO sheets and have formed ultra huge impermeable fractions that inhibit oxygen diffusion through the polymer matrix [22]. In another study, the effect of crystallinity degree on oxygen permeability values of films prepared via in situ polymerization of L-lactide with silane-modified nanosilica and MMT and also pure PLA and commercial PLA was evaluated. The results showed that the difference in crystallinity between commercial PLA and pure polymer results in different oxygen permeability with commercial PLA having permeability 76% higher than pure PLA, which is the result of its lower crystallinity degree compared to pure PLA. These are two unmodified PLAs; therefore, the difference in permeability can be attributed only to the different crystalline content in these two samples. Also, it was demonstrated that the presence of nanoparticles in the polymer matrix increases crystallinity degree which positively affects permeability and contributed to improved oxygen barrier properties [115].

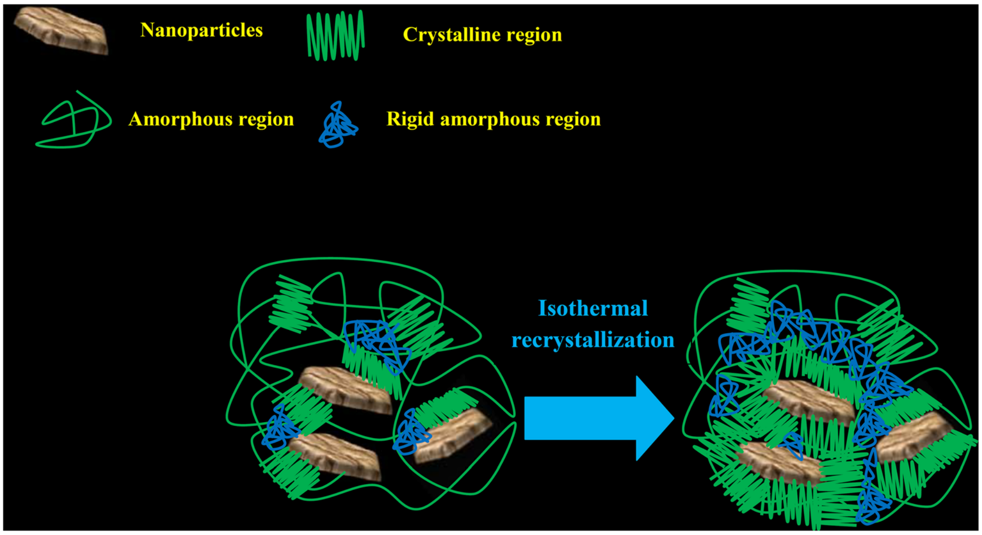

Fig. 5 Effect of isothermal recrystallization on crystallinity degree of nanocomposite matrix

Cross-Linked Polymers

Cross-linking can minimize the gasps and spaces that exist in the polymer matrix and by this way improves gas barrier performance [28]. In the case of nanocomposite, cross-linking can also connect neighboring nanoparticles and form huge impermeable obstacles for diffusing molecules and contribute to improved oxygen barrier properties [8]. In a study, borate was utilized as a cross-linking agent in order to enhance oxygen barrier properties of PVA/GO nanocomposite. It was demonstrated that cross-linking networks joined GO sheets with each other and formed large impermeable regions which contributed in a significant reduction in oxygen permeability of cross-linked nanocomposite [83]. In another similar study, boric acid induced cross-linking in PVA/GO nanocomposite which was also confirmed by FTIR through the formation of a B–O–C bond. Then, the prepared cross-linked nanocomposite was deposited on the nylon. Coated nylon film exhibits dramatic reduction of oxygen gas permeability compared to uncoated nylon film [89]. In another study by Lazar et al. [86], multilayer nanocoating which consisted of cross-linkable CH and PAA were deposited on PET substrate. In this study, glycidyl methacrylate was applied in order to functionalize CH via formation of acrylic functionalities within the film. After deposition, films were cross-linked by the use of a free radical initiator. Cross-linking of film was confirmed by FTIR results and also by the reduction in thickness of film after cross-linking. Moreover, the results of this study revealed a considerable reduction in oxygen permeability value [86]. In an investigation, a ternary polysaccharide polyelectrolyte complex (PPC) material based on crystalline nanocellulose (CNC), chitosan (CS), and carboxymethyl cellulose (CMC) was manufactured by an immediate high-shear homogenization procedure. CS and CMC were ionically cross-linked and formed a homogeneous continuous matrix. CNC was incorporated as nanoreinforcement into the formed network. The developed PPC film exhibited improved barrier properties, ascribed to the even distribution and good interfacial compatibility of CNC within the CS/CMC matrix [24]. Zhuang et al. [165] have prepared an antibacterial chitosan-citric biomembrane with enhanced oxygen barrier performance by in situ cross-linking. The obtained film exhibited a low oxygen transmission rate (below 0.1 $\mathrm{cm}^3/\mathrm{m}^{-2}/\mathrm{day}^{-1}$ at 40% RH), as a result of the increased diffusion length arising from the hydrogen-bonding, ionic, and covalent cross-linking [165].

Conclusion

In recent years by established new regulation for single use packaging materials and banding equal multilayer structure such as polymer-polymer, more and more approaches have emerged in the food packaging industry, and future developments hold great potential for food packaging material with excellent oxygen barrier properties intended to protect oxygen-sensitive food products from deterioration and prolong their shelf life. This review highlights many processes that were tried to prepare, “superbarrier” packaging toward oxygen including layer-by-layer assembly, coating, incorporation of nanoparticles, increasing ratio of crystalline to amorphous regions in polymer via introducing new crystals, and the combination of different methods. Compared to the pure polymer matrix, the gas barrier performance of polymer via these approaches was improved. The results of the scientific literature had illustrated that these prepared materials can also replace other preferred materials like aluminum foil and solve the opaqueness problem associated with metalized packaging. In this review, we highlighted the manufacturing methods of polymer with their respective improvement in gas barrier properties. This literature review showed that the barrier properties could be significantly improved with these techniques and the results could vary from case to case. Lastly, the percentages of oxygen permeability reduction by different strategies are also thoroughly discussed. Owing to increased demand on biopolymer in the past few years that typically have poor barrier characteristics toward oxygen compared to conventional polymer, it would be necessary to conduct more research on improving biopolymer oxygen barrier properties by applying techniques, which would make them capable of competing with synthetic polymers.

Acknowledgments This study is related to the project NO. 1398/10200 from the Student Research Committee, Shahid Beheshti University of Medical Sciences, Tehran, Iran. We also appreciate “Student Research Committee” and “Research & Technology Chancellor” in Shahid Beheshti University of Medical Sciences for their financial support of this study.

Compliance with ethical standards

Conflicts of interest The authors declare that they have no conflicts of interest.

Nomenclature BOPP, Biaxially oriented polypropylene; BPEI, Branched poly(ethylenimine); C-CNCW, carboxylated cellulose nano crystal whisker; CF, Carbon fiber; CH, Chitosan; CNW, Cellulose nanowhiskers; COC, Cyclic olefin copolymer; CR, Carrageenan; DA-GO, Dodecyl amine-functionalized graphene oxide; DA-RGO, Dodecyl amine-functionalized reduced graphene oxide; EP, Epoxy resin; EPDM, Ethylene–propylene–diene rubber; EVA, Ethylene-vinyl acetate; EVOH, Ethylene vinyl alcohol; FTIR, Fourier transform infrared spectroscopy; GNPs, Graphite nanoplatelets; GO, Graphene oxide; GONS, Graphene oxide nanosheet; HDPE, High density polyethylene; HEC, Hydroxyethyl cellulose; ICN, n-Octadecyl isocyanate; IIR, Poly (isobutylene- isoprene) rubber; LAP, Laponite; LCP, Liquid-crystal polymer; LDPE, Low density polyethylene; Li-Hec, Lithium fluoro-hectorite; LLDPE, Linear low density polyethylene; MMT, Montmorillonite; MXD6, Poly(m-xylylene adipamide); Nafion, hydrophobic fluorinated polymer; NFC, Nanofibrillated cellulose; O-MMT, Organo-modified montmorillonite; OPP, Oriented polypropylene; O-VER, Organo-vermiculite; PA, polyamide; PAA, Poly(acrylic acid); PAAm, Poly(allyl amine); PAH, Poly(allyl amine hydrochloride); PAI, Poly(amide-imide); PAM, Polyacrylamide; PAN, Polyacrylonitrile; PC, Polycarbonate; PCL, Polycaprolactone; PEI, Polyethylenimine; PEN, Polyethylene naphthalate; PEO, Polyethylene oxide; PET, Polyethylene terephthalate; PETG, Polyethylene terephthalate glycol; PGD, Polyglycidol; phr, Weight parts per 100 weight parts polymer; PHB, Polyhydroxyalkanoate; PI, polyimide; PLA, Poly lactic acid; PP, Polypropylene; PS, polystyrene; PSS, Polystyrene sulfonate; PU, polyurethane; PVA or PVOH, Polyvinyl alcohol; PVAm, Polyvinylamine; PVC, Polyvinyl chloride; PVDC, Poly (vinylidine) chloride; PVP, Polyvinylpyrrolidone; PUR, Polyurethane; RGO, Reduced graphene oxide; RH, Relative humidity; SBR, Styrene butadiene rubber; VAC, Vinyl acetate; VMT, Vermiculite; XG, Xyloglucan; XNBR, Carboxylated acrylonitrile butadiene rubber

References

1. Ali NA, Noori FTM (2014) Gas barrier properties of biodegradable polymer nanocomposites films. *Chem Mater Res 6*(1)
2. Apaydin K, Laachachi A, Ball V, Jimenez M, Bourbigot S, Toniazzo V, Ruch D (2013) Polyallylamine–montmorillonite as super flame retardant coating assemblies by layer-by layer deposition on polyamide. *Polym Degrad Stab 98*(2):627–634. https://doi.org/10.1016/J.POLYMDEGRADSTAB.2012.11.006
3. Apicella A, Scarfato P, Di Maio L, Incarnato L (2018) Transport properties of multilayer active PET films with different layers configuration. *React Funct Polym 127*:29–37
4. Ariga K, Hill J (2007) Layer-by-layer assembly as a versatile bottom-up nanofabrication technique for exploratory research and realistic application. *Phys Chem Chem Phys 9*(19):2319–2340 Retrieved from https://pubs.rsc.org/en/content/articlehtml/2007/cp/b700410a
5. Arrieta MP, Peponi L, López D, López J, Kenny JM (2017) An overview of nanoparticles role in the improvement of barrier properties of bioplastics for food packaging applications. In Food Packaging (pp. 391–424). Elsevier
6. Arunvisut S, Phummanee S, Somwangthanaroj A (2007) Effect of clay on mechanical and gas barrier properties of blown film LDPE/clay nanocomposites. *J Appl Polym Sci 106*(4):2210–2217
7. Aulin C, Karabulut E, Tran A, Waišgberg L, Lindström T (2013) Transparent nanocellulosic multilayer thin films on polylactic acid with tunable gas barrier properties. *ACS Appl Mater Interfaces 5*(15):7352–7359. https://doi.org/10.1021/am401700n
8. Azeredo HMC, Waldron KW (2016) Crosslinking in polysaccharide and protein films and coatings for food contact–A review. *Trends Food Sci Technol 52*:109–122
9. Bang G, Kim S (2012) Biodegradable poly (lactic acid)-based hybrid coating materials for food packaging films with gas barrier properties. *J Ind Eng Chem* 18:1063–1068 Retrieved from https://www.sciencedirect.com/science/article/pii/S1226086X11003650
10. Bharadwaj RK, Mehrabi AR, Hamilton C, Trujillo C, Murga M, Fan R, Chavira A, Thompson AK (2002) Structure-property relationships in cross-linked polyesterclay nanocomposites. *Polymer 43*(13):3699–3705
11. Bhattacharya M, Biswas S, Bhowmick AK (2011) Permeation characteristics and modeling of barrier properties of multifunctional rubber nanocomposites. *Polymer 52*(7):1562–1576
12. Bikiaris DN, Triantafyllidis KS (2013) HDPE/Cu-nanofiber nanocomposites with enhanced antibacterial and oxygen barrier properties appropriate for food packaging applications. *Mater Lett 93*: 1–4. https://doi.org/10.1016/j.matlet.2012.10.128
13. Brockgreitens J, Abbas A (2016) Responsive food packaging: Recent progress and technological prospects. *Compr Rev Food Sci Food Saf 15*(1):3–15
14. Bugnicourt E, Schmid M, Nerney OM, Wildner J, Smykala L, Lazzeri A, Cinelli P (2013) Processing and validation of whey-protein-coated films and laminates at semi-industrial scale as novel recyclable food packaging materials with excellent barrier properties. *Adv Mater Sci Eng* 2013:*2013*–2010. https://doi.org/10.1155/2013/496207
15. Calva-Estrada SJ, Jiménez-Fernández M, Lugo-Cervantes E (2019) Protein-based films: advances in the development of biomaterials applicable to food packaging. *Food Eng Rev 11*(2):78–92
16. Cao L, Ge T, Meng F, Xu S, Li J, Wang L (2020) An edible oil packaging film with improved barrier properties and heat sealability from cassia gum incorporating carboxylated cellulose nano crystal whisker. *Food Hydrocoll 98*:105251
17. Carosio F, Colonna S, Fina A, Rydzek G, Hemmerlé J, Jierry L, Schaaf P, Boulmedais F (2014) Efficient gas and water vapor barrier properties of thin poly(lactic acid) packaging films: functionalization with moisture resistant Nafion and clay multilayers. *Chem Mater 26*(19):5459–5466. https://doi.org/10.1021/cm501359e
18. Carvalho JWC, Sarantópoulos C, Innocentini-Mei LH (2010) Nanocomposites-based polyolefins as alternative to improve barrier properties. *J Appl Polym Sci 118*(6):3695–3700. https://doi.org/10.1002/app.32507
19. Chaiko DJ, Leyva AA (2005) Thermal transitions and barrier properties of olefinic nanocomposites. *Chem Mater 17*(1):13–19
20. Chemin M, Heux L, Guérin D, Crowther-Alwyn L, Jean B (2019) Hybrid gibbsite nanoplatelets/cellulose nanocrystals multilayered films coatings for oxygen barrier improvement. *Front Chem 7*:507
21. Chen J, Fu Y, An Q, Lo S, Huang S, Hung W (2013) Tuning nanostructure of graphene oxide/polyelectrolyte LbL assemblies by controlling pH of GO suspension to fabricate transparent and super gas barrier films. Nanoscale. Retrieved from https://pubs.rsc.org/en/content/articlehtml/2013/nr/c3nr02845c
22. Chen J-T, Fu Y-J, An Q-F, Lo S-C, Zhong Y-Z, Hu C-C, Lee KR, Lai J-Y (2014a) Enhancing polymer/graphene oxide gas barrier film properties by introducing new crystals. *Carbon 75*:443–451. https://doi.org/10.1016/J.CARBON.2014.04.024
23. Cheng S, Zhang Y, Cha R, Yang J, Jiang X (2016) Water-soluble nanocrystalline cellulose films with highly transparent and oxygen barrier properties. *Nanoscale 8*(2):973–978. https://doi.org/10.1039/c5nr07647a
24. Chi K, Catchmark JM (2018) Improved eco-friendly barrier materials based on crystalline nanocellulose/chitosan/carboxymethyl cellulose polyelectrolyte complexes. *Food Hydrocoll 80*:195–205
25. Ciannamea EM, Castillo LA, Barbosa SE, De Angelis MG (2018) Barrier properties and mechanical strength of bio-renewable, heat-sealable films based on gelatin, glycerol and soybean oil for sustainable food packaging. *React Funct Polym 125*:29–36. https://doi.org/10.1016/j.reactfunctpolym.2018.02.001
26. Cirillo G, Curcio M, Spataro T, Picci N, Restuccia D, Iemma F, Spizzirri UG (2018) Antioxidant polymers for food packaging. In Food Packaging and Preservation (pp. 213–238). Elsevier
27. Coma V, Bartkowiak A (2019) Potential of chitosans in the development of edible food packaging. Chitin and Chitosan: Properties and Applications, 349–369
28. Compton OC, Kim S, Pierre C, Torkelson JM, Nguyen ST (2010) Crumpled graphene nanosheets as highly effective barrier property enhancers. *Adv Mater 22*(42):4759–4763. https://doi.org/10.1002/adma.201000960
29. da Silva Sobrinho AS, Czeremuszkin G, Latrèche M, Wertheimer MR (2000) Defect-permeation correlation for ultrathin transparent barrier coatings on polymers. *J Vac Sci Technol A 18*(1):149–157. https://doi.org/10.1116/1.582156
30. Dai C-F, Li P-R, Yeh J-M (2008) Comparative studies for the effect of intercalating agent on the physical properties of epoxy

resin-clay based nanocomposite materials. *Eur Polym J 44*(8): 2439–2447

31. Dobrucka R, Przekop R (2019) New perspectives in active and intelligent food packaging. *J Food Process Preserv 43*(11): e14194
32. Donadi S, Modesti M, Lorenzetti A, Besco S (2011) PET/PA nanocomposite blends with improved gas barrier properties: effect of processing conditions. *J Appl Polym Sci 122*(5):3290–3297
33. Durmucs A, Woo M, Kacsgöz A, Macosko CW, Tsapatsis M (2007) Intercalated linear low density polyethylene (LLDPE)/clay nanocomposites prepared with oxidized polyethylene as a new type compatibilizer: structural, mechanical and barrier properties. *Eur Polym J 43*(9):3737–3749
34. Enescu D, Cerqueira MA, Fucinos P, Pastrana LM (2019) Recent advances and challenges on applications of nanotechnology in food packaging. A literature review. *Food Chem Toxicol* 110814
35. Erlat AG, Spontak RJ, Clarke RP, Robinson TC, Haaland PD, Tropsha Y, Harvey NG, Vogler EA (1999) SiOx gas barrier coatings on polymer substrates: morphology and gas transport considerations. *J Phys Chem B 103*(29):6047–6055. https://doi.org/10.1021/jp990737e
36. Fang Z, Zhao Y, Warner RD, Johnson SK (2017) Active and intelligent packaging in meat industry. *Trends Food Sci Technol 61*:60–71
37. Farhoodi M (2016) Nanocomposite materials for food packaging applications: characterization and safety evaluation. *Food Eng Rev 8*(1):35–51
38. Felts JT, Grubb AD (1992) Commercial-scale application of plasma processing for polymeric substrates: from laboratory to production. *J Vac Sci Technol A 10*(4):1675–1681. https://doi.org/10.1116/1.577768
39. Finch CA (1985) Encyclopedia of polymer science and engineering, 2nd edition. *Brit Polymer J 17*(4):377–377. https://doi.org/10.1002/pi.4980170412
40. Frounchi M, Dourbash A (2009) Oxygen barrier properties of poly (ethylene terephthalate) nanocomposite films. *Macromol Mater Eng 294*(1):68–74
41. Gaikwad KK, Singh S, Lee YS (2018) Oxygen scavenging films in food packaging. *Environ Chem Lett 16*(2):523–538. https://doi.org/10.1007/s10311-018-0705-z
42. Gaikwad KK, Singh S, Lee YS (2019) Antimicrobial and improved barrier properties of natural phenolic compound-coated polymeric films for active packaging applications. *J Coat Technol Res 16*(1):147–157. https://doi.org/10.1007/s11998-018-0109-9
43. Garc'ia A, Eceolaza S, Iriarte M, Uriarte C, Etxeberria A (2007) Barrier character improvement of an amorphous polyamide (Trogamid) by the addition of a nanoclay. *J Membr Sci 301*(1–2):190–199
44. Gavgani JN, Adelnia H, Gudarzi MM (2014) Intumescent flame retardant polyurethane/reduced graphene oxide composites with improved mechanical, thermal, and barrier properties. *J Mater Sci 49*(1):243–254
45. Ghaani M, Cozzolino CA, Castelli G, Farris S (2016) An overview of the intelligent packaging technologies in the food sector. *Trends Food Sci Technol 51*:1–11
46. Ghasemnejad-Afshar E, Amjad-Iranagh S, Zarif M, Modarress H (2020) Effect of side branch on gas separation performance of triptycene based PIM membrane: a molecular simulation study. Polymer Testing 106339
47. Ghoshal G (2019) Recent development in beverage packaging material and its adaptation strategy. In Trends in Beverage Packaging (pp. 21–50). Elsevier
48. Giannakas A, Vlacha M, Salmas C, Leontiou A, Katapodis P, Stamatis H, Barkoula N M, Ladavos A (2016) Preparation, characterization, mechanical, barrier and antimicrobial properties of chitosan/PVOH/clay nanocomposites. Carbohydr Polym 140: 408–415
49. Goh K, Heising JK, Yuan Y, Karahan HE, Wei L, Zhai S, Koh JX, Htin NM, Zhang F, Wang R, Fane AG, Dekker M, Dehghani F, Chen Y (2016) Sandwich-architectured poly(lactic acid)-graphene composite food packaging films. *ACS Appl Mater Interfaces 8*(15):9994–10004. https://doi.org/10.1021/acsami.6b02498
50. Gu C-H, Wang J-J, Yu Y, Sun H, Shuai N, Wei B (2013) Biodegradable multilayer barrier films based on alginate/polyethyleneimine and biaxially oriented poly(lactic acid). *Carbohydr Polym 92*(2):1579–1585. https://doi.org/10.1016/J.CARBPOL.2012.11.004
51. Guin T, Krecker M, Hagen DA, Grunlan JC (2014) Thick growing multilayer nanobrick wall thin films: super gas barrier with very few layers. *Langmuir 30*(24):7057–7060. https://doi.org/10.1021/la501946f
52. Gupta A, Mulchandani N, Shah M, Kumar S, Katiyar V (2018) Functionalized chitosan mediated stereocomplexation of poly(lactic acid): influence on crystallization, oxygen permeability, wettability and biocompatibility behavior. *Polymer 142*:196–208. https://doi.org/10.1016/j.polymer.2017.12.064
53. Hagen DA, Box C, Greenlee S, Xiang F, Regev O, Grunlan JC (2014a) High gas barrier imparted by similarly charged multilayers in nanobrick wall thin films. *RSC Adv 4*(35):18354–18359. https://doi.org/10.1039/C4RA01621A
54. Hamzehlou SH, Katbab AA (2007) Bottle-to-bottle recycling of PET via nanostructure formation by melt intercalation in twin screw compounder: improved thermal, barrier, and microbiological properties. *J Appl Polym Sci 106*(2):1375–1382
55. Han J W, Ruiz-Garcia L, Qian J P, Yang X T (2018) Food packaging: A comprehensive review and future trends. Compr Rev Food Sci Food Saf 17(4): 860–877
56. Han'guk Sikp'um Kwahakhoe, J.-W. (2007). *Food science and biotechnology*. *Food Science and Biotechnology* (Vol. 16). Korean Society of Food Science and Technology. Retrieved from http://www.dbpia.co.kr/Journal/ArticleDetail/NODE01728955
57. Hanika M, Langowski H-C, Moosheimer U, Peukert W (2003) Inorganic layers on polymeric films—influence of defects and morphology on barrier properties. *Chem Eng Technol 26*(5): 605–614. https://doi.org/10.1002/ceat.200390093
58. Holder KM, Priolo MA, Secrist KE, Greenlee SM, Nolte AJ, Grunlan JC (2012) Humidity-responsive gas barrier of hydrogen-bonded polymer-clay multilayer thin films. *J Phys Chem C 116*(37):19851–19856. https://doi.org/10.1021/jp306002p
59. Holder KM, Spears BR, Huff ME, Priolo MA, Harth E, Grunlan JC (2014) Stretchable gas barrier achieved with partially hydrogen-bonded multilayer nanocoating. *Macromol Rapid Commun 35*(10):960–964. https://doi.org/10.1002/marc.201400104
60. Hong SI, Lee JH, Bae HJ, Koo SY, Lee HS, Choi JH, Kim DH, Park SH, Park HJ (2011) Effect of shear rate on structural, mechanical, and barrier properties of chitosan/montmorillonite nanocomposite film. *J Appl Polym Sci 119*(5):2742–2749
61. Hosseini SF, Gómez-Guillén MC (2018) A state-of-the-art review on the elaboration of fish gelatin as bioactive packaging: special emphasis on nanotechnology-based approaches. *Trends Food Sci Technol 79*:125–135
62. Hosseini SF, Javidi Z, Rezaei M (2016) Efficient gas barrier properties of multi-layer films based on poly(lactic acid) and fish gelatin. *Int J Biol Macromol 92*:1205–1214. https://doi.org/10.1016/J.IJBIOMAC.2016.08.034
63. Huang H-Y, Huang T-C, Yeh T-C, Tsai C-Y, Lai C-L, Tsai M-H, Yeh JM, Chou Y-C (2011) Advanced anticorrosive materials prepared from amine-capped aniline trimer-based electroactive polyimide-clay nanocomposite materials with synergistic effects

of redox catalytic capability and gas barrier properties. *Polymer 52*(11):2391–2400

64. Huang H-D, Ren P-G, Chen J, Zhang W-Q, Ji X, Li Z-M (2012) High barrier graphene oxide nanosheet/poly(vinyl alcohol) nanocomposite films. *J Membr Sci* 409–410:156–163. https://doi.org/10.1016/j.memsci.2012.03.051
65. Huang H-D, Ren P-G, Xu J-Z, Xu L, Zhong G-J, Hsiao BS, Li Z-M (2014) Improved barrier properties of poly (lactic acid) with randomly dispersed graphene oxide nanosheets. *J Membr Sci 464*: 110–118
66. Idumah CI, Zurina M, Ogbu J, Ndem JU, Igba EC (2020) A review on innovations in polymeric nanocomposite packaging materials and electrical sensors for food and agriculture. *Compos Interf 27*(1):1–72
67. Inagaki N, Tasaka S, Hiramatsu H (1999) Preparation of oxygen gas barrier poly(ethylene terephthalate) films by deposition of silicon oxide films plasma-polymerized from a mixture of tetramethoxysilane and oxygen. *J Appl Polym Sci 71*(12):2091–2100. https://doi.org/10.1002/(SICI)1097-4628(19990321)71:12<2091::AID-APP20>3.0.CO;2-A
68. Incarnato L, Scarfato P, Russo GM, Di Maio L, Iannelli P, Acierno D (2003) Preparation and characterization of new melt compounded copolyamide nanocomposites. *Polymer 44*(16): 4625–4634
69. Ishtiaque S, Naz S, Ahmed J, Faruqui A (2018) Barrier properties analysis of polyethylene terephthalate films (PET) coated with natural polyphenolic and gelatin mixture (PGM). *Defect Diffusion Forum 382 DDF*:38–43. https://doi.org/10.4028/www.scientific.net/DDF.382.38
70. Izu M, Dotter B, Ovshinski SR (1993). In High performance clear coat barrier film , 36 th Annual Technical Conference Proceedings , Society of Vacuum Coaters (p. 333). Dallas
71. Jang W, Rawson I, Grunlan J (2008) Layer-by-layer assembly of thin film oxygen barrier. *Thin Solid Films* Retrieved from https://www.sciencedirect.com/science/article/pii/S0040609007015581 516:4819–4825
72. Jiang J, Benter M, Taboryski R, Bechgaard K (2010) Oxygen barrier coating deposited by novel plasma-enhanced chemical vapor deposition. *J Appl Polym Sci 115*(5):2767–2772. https://doi.org/10.1002/app.30222
73. Johnson DR, Decker EA (2015) The role of oxygen in lipid oxidation reactions: a review. *Annu Rev Food Sci Technol 6*(1):171–190. https://doi.org/10.1146/annurev-food-022814-015532
74. Joo E, Chang Y, Choi I, Lee SB, Kim DH, Choi YJ, Yoon CS, Han J (2018) Whey protein-coated high oxygen barrier multilayer films using surface pretreated PET substrate. *Food Hydrocoll 80*: 1–7. https://doi.org/10.1016/j.foodhyd.2018.01.027
75. Júnior LM, Cristianini M, Padula M, Anjos CAR (2019) Effect of high-pressure processing on characteristics of flexible packaging for foods and beverages. *Food Res Int 119*:920–930
76. Kalendová A, Merinska D, Gerard JF, Slouf M (2013) Polymer/clay nanocomposites and their gas barrier properties. *Polym Compos 34*(9):1418–1424
77. Kang H, Zuo K, Wang Z, Zhang L, Liu L, Guo B (2014) Using a green method to develop graphene oxide/elastomers nanocomposites with combination of high barrier and mechanical performance. *Compos Sci Technol 92*:1–8
78. Khosravi A, Fereidoon A, Khorasani MM, Naderi G, Ganjali MR, Zarrintaj P, Saeb MR, Gutiérrez TJ (2020) Soft and hard sections from cellulose-reinforced poly (lactic acid)-based food packaging films: a critical review. *Food Packag Shelf Life 23*:100429
79. Kochumalayil JJ, Bergenstrahle-Wohlert M, Utsel S, Wagberg L, Zhou Q, Berglund LA (2012) Bioinspired and highly oriented clay nanocomposites with a xyloglucan biopolymer matrix: extending the range of mechanical and barrier properties. *Biomacromolecules 14*(1):84–91
80. Krug T (1990) In Transparent barriers for food packaging , 33rd Annual Technical Conference Proceedings , Society of Vacuum Coaters (p. 163). New Orleans
81. Krug T, Ludwig R, Steiniger G (1993) In New developments in transparent barrier coatings , 36 th Annual Technical Conference Proceedings , Society of Vacuum Coaters (p. 302). Dallas
82. Lagaron JM, Cabedo L, Cava D, Feijoo JL, Gavara R, Gimenez E (2005) Improving packaged food quality and safety. Part 2: Nanocomposites. *Food Addit Contam 22*(10):994–998
83. Lai C-L, Chen J-T, Fu Y-J, Liu W-R, Zhong Y-R, Huang S-H, Hung WS, Lue SJ, Hu CC, Lee K-R (2015) Bio-inspired crosslinking with borate for enhancing gas-barrier properties of poly(-vinyl alcohol)/graphene oxide composite films. *Carbon 82*:513–522. https://doi.org/10.1016/J.CARBON.2014.11.003
84. Laufer G, Kirkland C, Cain AA, Grunlan JC (2012) Clay-chitosan nanobrick walls: completely renewable gas barrier and flame-retardant nanocoatings. *ACS Appl Mater Interfaces 4*(3):1643–1649. https://doi.org/10.1021/am2017915
85. Laufer G, Kirkland C, Cain AA, Grunlan JC (2013) Oxygen barrier of multilayer thin films comprised of polysaccharides and clay. *Carbohydr Polym 95*(1):299–302. https://doi.org/10.1016/J.CARBPOL.2013.02.048
86. Lazar S, Garcia-Valdez O, Kennedy E, Champagne P, Cunningham M, Grunlan J (2019) Crosslinkable-chitosan-enabled moisture-resistant multilayer gas barrier thin film. *Macromol Rapid Commun 40*(6):1800853. https://doi.org/10.1002/marc.201800853
87. Leväsalmi J-M, McCarthy TJ (1997) Poly(4-methyl-1-pentene)-supported polyelectrolyte multilayer films: preparation and gas permeability. *Macromolecules 30*(6):1752–1757. https://doi.org/10.1021/ma961245s
88. Li C, Jiang T, Wang J, Peng S, Wu H, Shen J, Guo S, Zhang X, Harkin-Jones E (2018a) Enhancing the oxygen-barrier properties of polylactide by tailoring the arrangement of crystalline lamellae. *ACS Sustain Chem Eng 6*(5):6247–6255
89. Li X, Bandyopadhyay P, Guo M, Kim NH, Lee JH (2018b) Enhanced gas barrier and anticorrosion performance of boric acid induced cross-linked poly(vinyl alcohol-co-ethylene)/graphene oxide film. *Carbon 133*:150–161. https://doi.org/10.1016/J.CARBON.2018.03.036
90. Li X, Bandyopadhyay P, Nguyen TT, Park O, Lee JH (2018c) Fabrication of functionalized graphene oxide/maleic anhydride grafted polypropylene composite film with excellent gas barrier and anticorrosion properties. *J Membr Sci 547*:80–92
91. Li Y, Zhang K, Nie M, Wang Q (2020) Application of compatibilized polymer blends in packaging. In Compatibilization of Polymer Blends (pp. 539–561). Elsevier
92. Lin N, Tang J, Dufresne A, Tam MKC (2019) Advanced functional materials from nanopolysaccharides. Springer
93. Liu A, Walther A, Ikkala O, Belova L, Berglund LA (2011) Clay nanopaper with tough cellulose nanofiber matrix for fire retardancy and gas barrier functions. *Biomacromolecules 12*(3): 633–641
94. López-Rubio A, Fabra MJ, Mart'inez-Sanz M (2019) Food packaging based on nanomaterials. Multidisciplinary Digital Publishing Institute
95. Lu P, Guo M, Xu Z, Wu M (2018) Application of nanofibrillated cellulose on BOPP/LDPE film as oxygen barrier and antimicrobial coating based on cold plasma treatment. *Coatings 8*(6). https://doi.org/10.3390/coatings8060207
96. Mahmoudian S, Wahit MU, Imran M, Ismail AF, Balakrishnan H (2012) A facile approach to prepare regenerated cellulose/graphene nanoplatelets nanocomposite using room-temperature ionic liquid. *J Nanosci Nanotechnol 12*(7):5233–5239
97. Majeed K, Hassan A, Bakar AA, Jawaid M (2016) Effect of montmorillonite (MMT) content on the mechanical, oxygen

barrier, and thermal properties of rice husk/MMT hybrid filler-filled low-density polyethylene nanocomposite blown films. *J Thermoplast Compos Mater 29*(7):1003–1019

98. Mangaraj S, Yadav A, Bal LM, Dash SK, Mahanti NK (2019) Application of biodegradable polymers in food packaging industry: a comprehensive review. *J Packaging Technol Res 3*(1):77–96
99. Massey LK (2003) Permeability properties of plastics and elastomers: a guide to packaging and barrier materials. William Andrew
100. Mei L, Wang Q (2020) Advances in using nanotechnology structuring approaches for improving food packaging. *Annu Rev Food Sci Technol 11:339–364*
101. Merritt S, Wemyss AM, Farris S, Patole S, Patias G, Haddleton DM, … Wan C (2020) Gas barrier polymer nanocomposite films prepared by graphene oxide encapsulated polystyrene microparticles. ACS Applied Polymer Materials
102. Min D B, Boff J M (2002) Chemistry and reaction of singlet oxygen in foods. Compr Rev Food Sci Food Saf 1(2):58–72
103. Minelli M, Sarti GC (2017) Elementary prediction of gas permeability in glassy polymers. *J Membr Sci 521*:73–83
104. Mirzadeh A, Kokabi M (2007) The effect of composition and draw-down ratio on morphology and oxygen permeability of polypropylene nanocomposite blown films. *Eur Polym J 43*(9):3757–3765
105. Misiano C, Simonetti E, Cerolini P, F S (1991) In Silicon oxide barrier improvement on plastic substrate , 33 rd Annual Technical Conference Proceedings , Society of Vacuum Coaters (p. 105). Philadelphia
106. Mittal V (2007) Gas permeation and mechanical properties of polypropylene nanocomposites with thermally-stable imidazolium modified clay. *Eur Polym J 43*(9):3727–3736
107. Möller MW, Kunz DA, Lunkenbein T, Sommer S, Nennemann A, Breu J (2012) UV-cured, flexible, and transparent nanocomposite coating with remarkable oxygen barrier. *Adv Mater 24*(16):2142–2147. https://doi.org/10.1002/adma.201104781
108. Moustafa H, Youssef AM, Darwish NA, Abou-Kandil AI (2019) Eco-friendly polymer composites for green packaging: future vision and challenges. Compos B: Eng
109. Mu R, Hong X, Ni Y, Li Y, Pang J, Wang Q, Xiao J, Zheng Y (2019) Recent trends and applications of cellulose nanocrystals in food industry. *Trends Food Sci Technol* 93:136–144
110. Nazarenko S, Meneghetti P, Julmon P, Olson BG, Qutubuddin S (2007) Gas barrier of polystyrene montmorillonite clay nanocomposites: effect of mineral layer aggregation. *J Polym Sci B Polym Phys 45*(13):1733–1753
111. Nešić A, Cabrera-Barjas G, Dimitrijević-Branković S, Davidović S, Radovanović N, Delattre C (2020) Prospect of polysaccharide-based materials as advanced food packaging. *Molecules 25*(1):135
112. Nyflött Å, Meriçer Ç, Minelli M, Moons E, Järnström L, Lestelius M, Baschetti MG (2017) The influence of moisture content on the polymer structure of polyvinyl alcohol in dispersion barrier coatings and its effect on the mass transport of oxygen. *J Coat Technol Res 14*(6):1345–1355
113. Öner M, Çöl AA, Pochat-Bohatier C, Bechelany M (2016) Effect of incorporation of boron nitride nanoparticles on the oxygen barrier and thermal properties of poly (3-hydroxybutyrate-co-hydroxyvalerate). *RSC Adv 6*(93):90973–90981
114. Öner M, Keskin G, Kizil G, Pochat-Bohatier C, Bechelany M (2019) Development of poly (3-hydroxybutyrate-co-3-hydroxyvalerate)/boron nitride bionanocomposites with enhanced barrier properties. *Polym Compos 40*(1):78–90
115. Ortenzi MA, Basilissi L, Farina H, Di Silvestro G, Piergiovanni L, Mascheroni E (2015) Evaluation of crystallinity and gas barrier properties of films obtained from PLA nanocomposites synthesized via “in situ” polymerization of l-lactide with silane-modified nanosilica and montmorillonite. *Eur Polym J 66*:478–491. https://doi.org/10.1016/J.EURPOLYMJ.2015.03.006
116. Osman MA, Atallah A (2004) High-density polyethylene microand nanocomposites: effect of particle shape, size and surface treatment on polymer crystallinity and gas permeability. *Macromol Rapid Commun 25*:1540–1544
117. Osman MA, Mittal V, Morbidelli M, Suter UW (2004) Epoxy-layered silicate nanocomposites and their gas permeation properties. *Macromolecules 37*(19):7250–7257
118. Paine AC (2016) Cyclic oxygen scavenging polymers for barrier applications
119. Park J, Won JK, Park J-G, Yu S, An J, Hong J, Kim C, Baeg KJ, Kim M-G (2019) High throughput bar-coating processed organic-inorganic hybrid multi-layers for gas barrier thin-films. *J Nanosci Nanotechnol 19*(7):4299–4304. https://doi.org/10.1166/jnn.2019.16333
120. Patra SK, Swain SK (2012) Effect of organoclays on the thermal, mechanical, and oxygen barrier properties of poly (methylmethacrylate-co-acrylonitrile)/clay nanocomposites. *Polym Compos 33*(5):796–802
121. Patwa R, Kumar A, Katiyar V (2018) Effect of silk nano-disc dispersion on mechanical, thermal, and barrier properties of poly(lactic acid) based bionanocomposites. *J Appl Polym Sci 135*(38). https://doi.org/10.1002/app.46671
122. Paul DR, Robeson LM (2008) Polymer nanotechnology: nanocomposites. *Polymer 49*(15):3187–3204
123. Picard E, Espuche E, Fulchiron R (2011) Effect of an organo-modified montmorillonite on PLA crystallization and gas barrier properties. *Appl Clay Sci 53*(1):58–65
124. Pinto AM, Cabral J, Tanaka DAP, Mendes AM, Magalhães FD (2013) Effect of incorporation of graphene oxide and graphene nanoplatelets on mechanical and gas permeability properties of poly (lactic acid) films. *Polym Int 62*(1):33–40
125. Prasad K, Nikzad M, Sbarski I (2018) Permeability control in polymeric systems: a review. *J Polym Res 25*(11):232
126. Priolo MA, Gamboa D, Grunlan JC (2010a) Transparent clay−polymer nano brick wall assemblies with tailorable oxygen barrier. *ACS Appl Mater Interfaces 2*(1):312–320. https://doi.org/10.1021/am900820k
127. Priolo MA, Gamboa D, Holder KM, Grunlan JC (2010b) Super gas barrier of transparent polymer-clay multilayer ultrathin films. *Nano Lett 10*(12):4970–4974. https://doi.org/10.1021/nl103047k
128. Priolo MA, Holder KM, Gamboa D, Grunlan JC (2011) Influence of clay concentration on the gas barrier of clay–polymer nanobrick wall thin film Assemblies. *Langmuir 27*(19):12106–12114. https://doi.org/10.1021/la201584r
129. Priolo MA, Holder KM, Greenlee SM, Stevens BE, Grunlan JC (2013) Precisely tuning the clay spacing in nanobrick wall gas barrier thin films. *Chem Mater 25*(9):1649–1655
130. Priolo MA, Holder KM, Guin T, Grunlan JC (2015) Recent advances in gas barrier thin films via layer-by-layer assembly of polymers and platelets. *Macromol Rapid Commun 36*(10):866–879. https://doi.org/10.1002/marc.201500055
131. Quinn JF, Johnston APR, Such GK, Zelikin AN, Caruso F (2007) Next generation, sequentially assembled ultrathin films: beyond electrostatics. *Chem Soc Rev 36*(5):707–718. https://doi.org/10.1039/b610778h
132. Rai M, Ingle AP, Gupta I, Pandit R, Paralikar P, Gade A, Chaud MV, dos Santos CA (2019) Smart nanopackaging for the enhancement of food shelf life. *Environ Chem Lett 17*(1):277–290
133. Ramezani H, Behzad T, Bagheri R (2019) Synergistic effect of graphene oxide nanoplatelets and cellulose nanofibers on mechanical, thermal, and barrier properties of thermoplastic starch. Polym Adv Technol
134. Ramos M, Jiménez A, Peltzer M, Garrigós MC (2014) Development of novel nano-biocomposite antioxidant films based on poly (lactic acid) and thymol for active packaging. *Food Chem 162*:149–155

135. Rampazzo R, Alkan D, Gazzotti S, Ortenzi MA, Piva G, Piergiovanni L (2017) Cellulose nanocrystals from lignocellulosic raw materials, for oxygen barrier coatings on food packaging films. *Packag Technol Sci 30*(10):645–661. https://doi.org/10.1002/pts.2308
136. Ren P-G, Wang H, Huang H-D, Yan D-X, Li Z-M (2014) Characterization and performance of dodecyl amine functionalized graphene oxide and dodecyl amine functionalized graphene/high-density polyethylene nanocomposites: a comparative study. *J Appl Polym Sci 131*(2)
137. Ren F, Tan W, Duan Q, Jin Y, Pei L, Ren P, Yan D (2019) Ultra-low gas permeable cellulose nanofiber nanocomposite films filled with highly oriented graphene oxide nanosheets induced by shear field. *Carbohydr Polym 209*:310–319
138. Risyon NP, Othman SH, Basha RK, Talib RA (2020) Characterization of polylactic acid/halloysite nanotubes bionanocomposite films for food packaging. *Food Packag Shelf Life 23*:100450
139. Roberts AP, Henry BM, Sutton AP, Grovenor CRM, Briggs GAD, Miyamoto T, Kano M, Tsukahara Y, Yanaka M (2002) Gas permeation in silicon-oxide/polymer (SiOx/PET) barrier films: role of the oxide lattice, nano-defects and macro-defects. *J Membr Sci 208*(1–2):75–88. https://doi.org/10.1016/S0376-7388(02)00178-3
140. Sabet SS, Katbab AA (2009) Interfacially compatibilized poly (lactic acid) and poly (lactic acid)/polycaprolactone/organoclay nanocomposites with improved biodegradability and barrier properties: effects of the compatibilizer structural parameters and feeding route. *J Appl Polym Sci 111*(4):1954–1963
141. Sanchez-Garcia MD, Gimenez E, Lagaron JM (2007) Novel PET nanocomposites of interest in food packaging applications and comparative barrier performance with biopolyester nanocomposites. *J Plastic Film Sheet 23*(2):133–148. https://doi.org/10.1177/8756087907083590
142. Schiller C, Strunz W (2001) The evaluation of experimental dielectric data of barrier coatings by means of different models. *Electrochim Acta* Retrieved from https://www.sciencedirect.com/science/article/pii/S0013468601006442 46:3619–3625
143. Sonchaeng U, Iniguez-Franco F, Auras R, Selke S, Rubino M, Lim L-T (2018) Poly (lactic acid) mass transfer properties. *Prog Polym Sci* 86:85–121
144. Spoljaric S, Salminen A, Dang Luong N, Lahtinen P, Vartiainen J, Tammelin T, Seppälä J (2014) Nanofibrillated cellulose, poly (vinyl alcohol), montmorillonite clay hybrid nanocomposites with superior barrier and thermomechanical properties. *Polym Compos 35*(6):1117–1131
145. Svagan AJ, Åkesson A, Cárdenas M, Bulut S, Knudsen JC, Risbo J, Plackett D (2012) Transparent films based on PLA and montmorillonite with tunable oxygen barrier properties. *Biomacromolecules 13*(2):397–405. https://doi.org/10.1021/bm201438m
146. Tas CE, Hendessi S, Baysal M, Unal S, Cebeci FC, Menceloglu YZ, Unal H (2017) Halloysite nanotubes/polyethylene nanocomposites for active food packaging materials with ethylene scavenging and gas barrier properties. *Food Bioprocess Technol 10*(4):789–798. https://doi.org/10.1007/s11947-017-1860-0
147. Tomlinson R, Klee M, Shane Garrett S-H, Heller J, Duncan R, Brocchini S (2001) Pendent chain functionalized polyacetals that display pH-dependent degradation: a platform for the development of novel polymer therapeutics. *Macromolecules 35*(2):473–480. https://doi.org/10.1021/MA0108867
148. Tzeng P, Maupin CR, Grunlan JC (2014) Influence of polymer interdiffusion and clay concentration on gas barrier of polyelectrolyte/clay nanobrick wall quadlayer assemblies. *J Membr Sci 452*:46–53. https://doi.org/10.1016/j.memsci.2013.10.039
149. Vaezi K, Asadpour G, Sharifi H (2020) Bio nanocomposites based on cationic starch reinforced with montmorillonite and cellulose nanocrystals: fundamental properties and biodegradability study. *Int J Biol Macromol* 146:374–386
150. Valapa RB, Pugazhenthi G, Katiyar V (2015) Effect of graphene content on the properties of poly (lactic acid) nanocomposites. *RSC Adv 5*(36):28410–28423
151. Villaluenga JPG, Khayet M, Lopez-Manchado MA, Valentin JL, Seoane B, Mengual JI (2007) Gas transport properties of polypropylene/clay composite membranes. *Eur Polym J 43*(4):1132–1143
152. Wan T, Chen L, Chua YC, Lu X (2004) Crystalline morphology and isothermal crystallization kinetics of poly (ethylene terephthalate)/clay nanocomposites. *J Appl Polym Sci 94*(4):1381–1388
153. Wang Y, Jabarin SA (2013) Novel preparation method for enhancing nanoparticle dispersion and barrier properties of poly (ethylene terephthalate) and poly (m-xylylene adipamide). *J Appl Polym Sci 129*(3):1455–1465
154. Wang C, Shi J, He M, Ding L, Li S, Wang Z, Wei J (2018) High strength cellulose/ATT composite films with good oxygen barrier property for sustainable packaging applications. *Cellulose 25*(7):4145–4154. https://doi.org/10.1007/s10570-018-1855-7
155. Weizman O, Dotan A, Nir Y, Ophir A (2017) Modified whey protein coatings for improved gas barrier properties of biodegradable films. *Polym Adv Technol 28*(2):261–270. https://doi.org/10.1002/pat.3882
156. Xiang F, Tzeng P, Sawyer JS, Regev O, Grunlan JC (2013) Improving the gas barrier property of clay-polymer multilayer thin films using shorter deposition times. *ACS Appl Mater Interfaces 6*(9):6040–6048
157. Xu P, Yang W, Niu D, Yu M, Du M, Dong W et al (2020) Multifunctional and robust polyhydroxyalkanoate nanocomposites with superior gas barrier, heat resistant and inherent antibacterial performances. *Chem Eng J 382*:122864
158. Yampolskii Y (2017) Permeability of polymers. Membrane Materials for Gas and Vapor Separation, 1–15
159. Yang Y-H, Haile M, Park YT, Malek FA, Grunlan JC (2011) Super gas barrier of all-polymer multilayer thin films. *Macromolecules 44*(6):1450–1459. https://doi.org/10.1021/ma1026127
160. Yang J, Bai L, Feng G, Yang X, Lv M, Zhang C et al (2013a) Thermal reduced graphene based poly (ethylene vinyl alcohol) nanocomposites: enhanced mechanical properties, gas barrier, water resistance, and thermal stability. *Ind Eng Chem Res 52*(47):16745–16754
161. Yang Y-H, Bolling L, Priolo MA, Grunlan JC (2013b) Super gas barrier and selectivity of graphene oxide-polymer multilayer thin films. *Adv Mater 25*(4):503–508. https://doi.org/10.1002/adma.201202951
162. Yu L, Lim Y, Han J, Kim K, Kim J, Choi S, Shin K (2012) A graphene oxide oxygen barrier film deposited via a self-assembly coating method. *Synth Met* Retrieved from https://www.sciencedirect.com/science/article/pii/S0379677912000707 162:710–714
163. Zabihzadeh Khajavi M, Mohammadi R, Ahmadi S, Farhoodi M, Yousefi M (2019) strategies for controlling release of plastic compounds into foodstuffs based on application of nanoparticles and its potential health issues. *Trends Food Sci Technol* 90:1–12
164. Zhang P, Zhao Y, Shi Q (2016) Characterization of a novel edible film based on gum ghatti: Effect of plasticizer type and concentration. *Carbohydr Polym 153*:345–355
165. Zhuang L, Zhi X, Du B, Yuan S (2020) Preparation of elastic and antibacterial chitosan–citric membranes with high oxygen barrier ability by in situ cross-linking. ACS Omega