Study on the Weathering Performance of Polypropylene by Artificial Accelerated Aging and Natural Aging

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ABSTRACT

The weathering performance of polypropylene (PP) was studied in an artificial accelerated weathering chamber of 340 nm ultraviolet light and natural conditions in Turpan, China. This work aimed to provide a method for calculating the aging failure of PP under natural conditions by the means of artificial accelerated aging. SEM showed that large crack widths of approximately 1.25 µm and 1.63 µm appealed on the PP surface after 15 days artificial accelerated aging and 150 days natural aging, respectively. XPS data revealed that the number of C-O and C=O bonds on the PP surface were generally the same after artificial accelerated aging and natural aging of PP for 3 and 30 days, 6 and 60 days, or 9 and 90 days, respectively. Moreover, the data of mechanical properties and capillary rheology also exhibited similar results. It was concluded that the aging rate of artificial accelerated aging was 10 times faster than that of natural aging in Turpan.

KEYWORDS: *Polypropylene, Weathering performance, Artificial accelerated aging, Natural aging.*

1. INTRODUCTION

The weathering performance and the mechanical and thermal properties of polymers are the most important topics in the applications of polymer materials.[1] Polypropylene (PP) is one of the five most widely used general resins in the world, which is white granules, odorless and non-toxic.[2] However, PP has poor resistance to photo-oxidation and temperature.[3] The deterioration

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of PP's properties is caused by several factors such as solar radiation, temperature, rain, wind, humidity, environmental pollutants, bacteria, and so on during natural service conditions.[4] Thus, the service life of polymer materials is shortened.[5] Specifically, ultraviolet (UV) radiation exposure would play an important role on the degradation of PP and consequently weaken its mechanical properties when exposed to direct sunlight.[6] Knowledge on the aging behavior of PP significantly helps its anti-aging research. Therefore, various studies have been conducted to assess the weathering performance of PP accurately. François-Heude et al.[7] investigated the effect of temperature, UV irradiation intensity, and wavelength of the thermal oxidative degradation of PP products. Gallo et al.[8] analyzed the changes in the thick and thin isotactic PP samples subjected to natural aging. Previous works mainly focused on investigating the results of PP degradation to identify the main factors that affect its long-term service performance in the natural conditions.

Artificial accelerated aging and natural aging have been commonly used to predict the weathering performance of PP.[9] The artificial accelerated aging is more efficient by intensifying UV radiation but the results are difficult to interpret with respect to the actual performances of PP in outdoors applications,[10] whereas the natural aging is more reliable and effective for evaluating its aging failure but is more time consuming.[11] Artificial accelerated aging is used to establish the method of artificial simulation of natural materials using, in order to supplement or even replace natural aging and predict the failure time of materials used in natural conditions, of which the cycle is short, not affected by regional climate, but often can not accurately simulate varied natural climate conditions.[12] Yang et al.[13] investigated the possibility of using the results from the accelerated weathering tests to predict the actual degradation of PP under outdoor weathering conditions. Rjeb et al.[14] used DSC and TG technology to investigate the natural and artificial aging of PP. Their researches mainly focused on the correlation of results from artificial accelerated aging to natural aging.

The energy of C-C bonds in PP and other polymer resins is 347.9 kJ/mol, which is equivalent to the ultraviolet light energy of 342 nm wavelength.[15] In this work, PP was placed under the ultraviolet light at 340 nm for artificial accelerated aging, which could cause the macromolecular chain fracture and chemical structure change of PP, and the molecular chain was degraded to different degrees, finally making the mechanical properties of materials worse. In addition, the simulated ultraviolet irradiation would also cause unstable free radicals to be produced in the matrix of weather-resistant materials, which would oxidize under the action of trace oxygen, and then cause the crystallinity of materials to decrease, which was manifested as an increase in C=O bonds.[16] China is vast country with the large climate difference, so aging behavior of the same materials vary in multiple regions due to different climatic conditions. When polymer materials are exposed to sunlight, it will undergo a series of reactions, mainly photochemical reactions according to the first and second laws of photochemical reactions.[17] Natural aging is a complete depolymerization which will cause the flaring, discoloration, foaming, cracking, falling off and so on for polymer materials.

Wavelength sensitivity of polymer materials is an important factor affecting natural aging. Natural aging sites should be selected because of the various climate types existing in the natural conditions. Elinor et al.[18] investigated the effects of the light intensity of the geographical location (Manila, Philippines) on PP weathering. Consequently, the aging extent of PP aging varies across different climatic regions.

However, few reports on the natural aging of PP had been conducted in Turpan which is located in an arid and semiarid area northwest of China. The place has a typical dry desert climate, abundant sunshine, hot and dry weather, strong UV radiation, and large temperature differences between day and night. Research on the long-term weathering performance of PP in such an environment is interesting for the development of weather-resistant materials and applications therein.

In this study, the weathering performance of the long-term natural aging for PP was compared with the exposing samples subjected to artificial accelerated aging under 340 nm ultraviolet light. Therefore, this work provided a method to calculate the failure of PP in natural conditions through artificial accelerated aging using the data of SEM, XPS, mechanical properties and capillary rheology within a certain time.

2. EXPERIMENTAL

2.1 Materials

PP (T30S: wire-drawing homopolymer, melt flow rate= 1.8~ 3.0 g·(10 min)$^{-1}$, M_w = 100,000 g/mol) was purchased from China Petroleum Dushanzi Petrochemical Company. The granules of PP after vacuum drying were fused and extruded, granulated and injected by plastic injection molding machine (XL-400 VI, Ningbo Gaoxin Co., Ltd., China) to obtain the standard strip splines for impact test and dumbbell splines for tensile test under 8×10^6 Pa at 190°C and screw speed of 30 r/min.

2.2 Artificial Accelerated Aging

Artificial accelerated aging of the standard strip splines and dumbbell splines of PP was done with an accelerated weathering apparatus (Atlas Material Testing Technology LLC.US), which is shown in Figure 1. In parallel to the natural aging, the irradiance value was at 340 nm of 0.68 W/m^2 (same to the

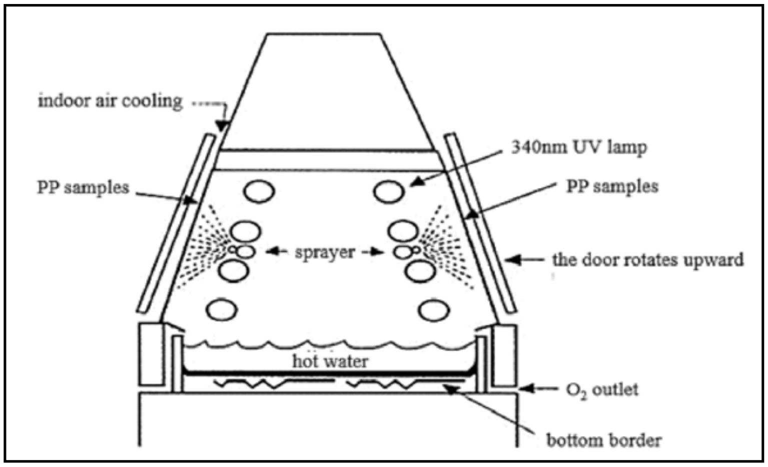


Figure 1. Artificial accelerated aging apparatus

average intensity of sunlight during the summer at 12:00). Main parameters of the experimental apparatus were that: equipment power supply was 220 V/50 Hz; total power was 560 W, of which the air heater was 300 W; the fan of 100 W; light source was UVA-340 fluorescent ultraviolet lamp tube imported from the US, two of which were installed in the apparatus; blackboard temperature: the highest was 70°C, the lowest was 10°C, which could produce about 60°C temperature difference; the two ends of the device were equipped with a blower (adjustable), which could generate 3-5 levels of wind to reduce the temperature of the blackboard. First, UV radiation was conducted for 3 h at the temperature of 60 ± 3°C, then cooled at 50 ± 3°C without UV radiation for 3 h, at last wind blew 10 min. The whole process of 370 minutes was repeatedly cycled for totally 15 days. The PP standard splines for tensile and impact test were sampled at intervals of 3, 6, 9, 12 and 15 days, respectively. The process flow is shown in Figure 2.

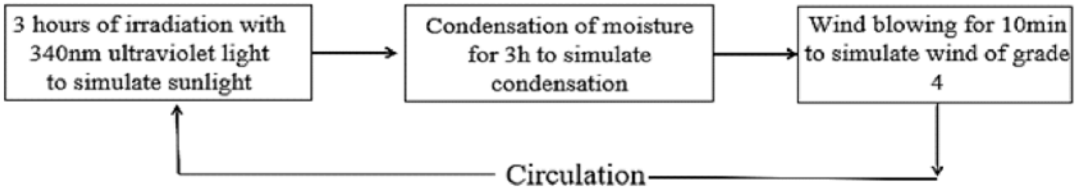


Figure 2. The process flow of artificial accelerated aging

2.3 Natural Aging

The PP samples of the standard strip splines and dumbbell splines were exposed in the world's second largest dry heat test station, Turpan natural environment research center of Xinjiang Uygur Autonomous Region, China (42°56' north, 89°12' east, 61.5 m from sea level). The PP standard splines for tensile and impact test were exposed to natural conditions for a period of time up to 5 months from June to October in 2019. The structure and mechanical properties of the samples were tested every month. In addition, the meteorological data of natural aging period were provided by the Turpan Weather Center, which was listed in Table 1.

TABLE 1. Weather conditions during natural aging

Period of Weather tests	Average Sunshine hours (h)	Average Intensity of Solar Irraditation (W-h/m^2)	Average Intensity of UV Irradiation (W-h/m^2)	Average Temperature (°C)	Average Humidity (%)
June	179.08	632.87	27.12	35.35	19.18
July	362.08	708.10	30.12	37.16	16.56
August	293.84	635.61	24.99	33.91	19.35
September	255.06	605.35	21.68	24.43	30.64
October	257.77	625.82	23.06	16.82	25.81

2.4 Mechanical Property Test of Samples Before and After Aging

2.4.1 Determination of tensile strength

The tensile specimens were tested according to ISO 527:2012 at room temperature (23°C) with a cross head speed of 100 mm/min using a computer-controlled universal testing machine (type CMT6104, China). The spline spacing is 30 mm, the width is 4 mm and the thickness is 2 mm. σ can be calculated by Equation (1):

$$\sigma = p/bd \qquad (1)$$

Iin which σ is tensile strength in MPa; P is maximum load and b and d are the width and thickness of the sample in mm, respectively.

The elongation at break (E, %) can be calculated by Equation (2):

$$E = L - L_0 / lL_0 \text{ x } 100\% \qquad (2)$$

where L_0 and L are the elongation length in mm before and after test, respectively.

2.4.2 Determination of impact strength

According to ISO 179:2010, an electronic impact testing machine (type XJJD-50, China) was used for Izod impact test with a 7.5 J pendulum at room temperature. The length of the spline is 80 mm, the width is 10 mm and the thickness is 4 mm, and the impact velocity is 3.8 m/s. Impact strength is calculated as follows:

$$\alpha = w / bh \text{ x } 10^3 \qquad (3)$$

in which α is the impact strength in kJ/m^2; W is the corrected energy to break the sample in J; b and h are the width and the thickness of the sample, respectively.

2.5 Structure Characterization

2.5.1 Surface electron morphology analysis

The surface morphology of PP before and after aging was investigated using the scanning electron microscopy (SEM, JSM-5600LV, JEOL, Ltd. Japan). The scanning was performed on the surface of the samples which were sputter-coated with gold prior to scanning. The acceleration voltage is 20 kv.

2.5.2 Capillary rheology analysis

The capillary rheology analysis was performed with a micro mixing rheometer (MiniLab II, Haake Germany). 5.0 g of PP granules before and after aging were added to the cylinder at a screw rotation speed of 20 r/min. The barrel temperature was set as 200°C and screw rod rotating was set as internal rotation. Screw speed changed from low to high at regular 10-second intervals, in order, 1, 2, 5, 10, 20, 50 r/min. Experimental data were collected after 3 s of the speed changes. The relationship between the torque and the shear viscosity of PP was obtained.

2.5.3 X-ray photoelectron spectroscopy (XPS)

XPS (ESCALAB 250Xi, Thermo Fisher. US) was used to investigate the surface composition of PP before and after aging. X-ray photoelectron spectra were recorded on a Thermo spectrometer with a monochromatized Al K Alpha X-ray source (1486.6 eV photons).

3. RESULTS AND DISCUSSION

3.1 Mechanical Properties Analyses of PP before and after Aging

For the PP samples subjected to artificial accelerated aging and natural aging, the data of the mechanical properties at different aging

TABLE 2. Mechanical properties data of PP before and after artificial accelerated aging

Mechanical properties	Without aging	Artificial accelerated aging time/day					
		3	6	9	12	15	20
Tensile Strength /Mpa	40.1	41.7	41.9	42.1	40.9	36.9	28.4
Elongation at break /%	492.1	240.1	209.5	186.3	168.5	147.4	75.2

TABLE 3. Mechanical properties data of PP before and after natural aging

Mechanical properties	**Without aging**	**Natural aging time/day**					
		15	30	60	90	120	150
Tensile Strength /Mpa	40.1	41.7	40.4	41.5	41.9	42.8	37.8
Elongation at break /%	492.1	310.3	291.3	196.6	161.8	81.2	23.9

times (Table 2 and Table 3) showed that the tensile strength of the specimens slightly increased and then decreased obviously. The retention rate of elongation at break of the artificial accelerated aging samples after 3 days of aging was 48.8%, indicating that the samples had basically lost its use value at this time, and the retention rate of elongation at break after artificial accelerated aging for 20 days was 3.1%, which had completely lost its value. Compared with the specimens without aging, the specimens that were artificially aged for 12 days and naturally aged for 120 days showed increased tensile strength by 6.0% and 7.3%, respectively. This was because there were some amorphous regions and incomplete parts in PP molecular chain, that was, defected or imperfect regions between crystals. The molecular chains and segments would be rearranged and the regularity of molecules would be improved during aging,[19] so that the tensile strength would improve. Compared with the specimens without aging, the tensile strength of the specimens naturally aged for 150 days was reduced by 5.3%. This difference was the result of the surface cracks on materials.

Figure 3 showed the consistent aging law between artificial accelerated aging and natural aging for 3 and 30 days, 6 and 60 days, or 9 and 90 days. In the artificial accelerated aging,

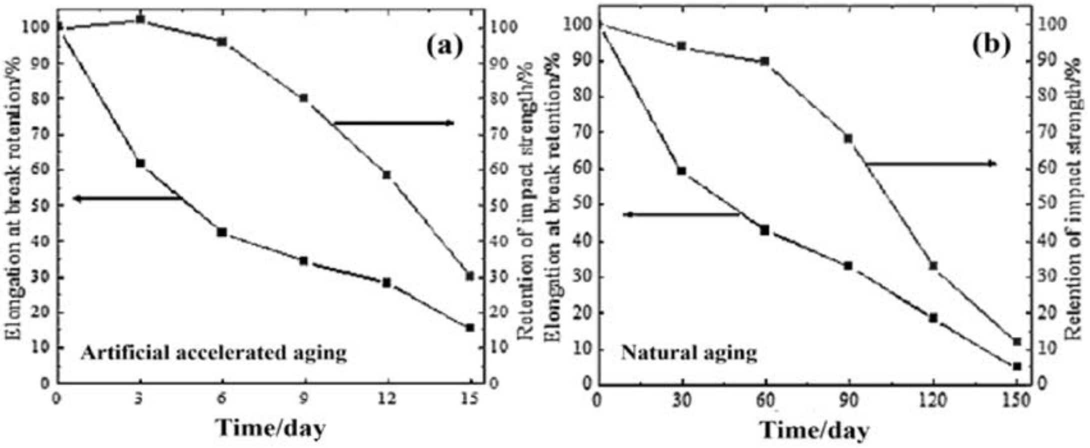


Figure 3. The mechanical properties of PP samples after different aging time:
(a) artificial accelerated aging; (b) natural aging

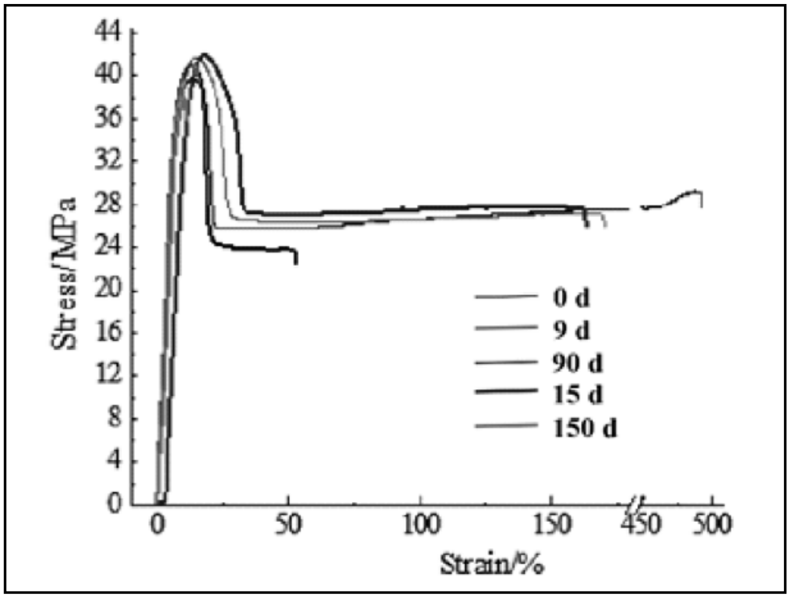


Figure 4. Tensile stress-strain curves of PP after different time aging

the breaking elongation retention rate after 3, 6, and 9 days was 61.5%, 42.2%, and 34.3%, respectively, compared with samples without aging. In the natural aging, the specimens aged for 30, 60, and 90 days had the breaking elongation retention rate of 59.2 %, 42.6 %, and 32.9 %, respectively. The degree of samples aged in natural conditions was lower than that of aged by artificially and resilient fracture on the PP samples became brittle fracture as shown in Figure 4. The breaking elongation retention rate of samples subjected to natural aging for 120 days was twice more than that of samples subjected to artificial accelerated aging for 12 days, three times for 150 days, which had totally lost them using value. This was because the samples were affected by the worse outdoors environment in the process of natural aging in Turpan, and their molecular chains were seriously degraded, and cracks were more likely to appear on the surface.

Figure 5 showed the scanning electron microscopy (SEM) images at ×1000 magnification of PP aged artificially and naturally. As the aging time increased, the surface of the samples gradually became uneven and rough. The surface of the PP samples, which were aged artificially for 3 and 6 days and naturally for 30 and 60 days, showed that the topography was smooth. The analysis of SEM showed that the samples were significantly affected by environmental factors and a large number of uneven rough surfaces began to show corrosion after 9 and 90 days of artificial and natural aging, respectively. Meanwhile, artificial accelerated aging produced several crack widths of approximately 1.25 μm after 15 days on PP surface. The surface of the PP samples exposed to the dry and heat conditions in Turpan exhibited a large amount of cracks with widths of approximately 1.63 μm after 150 days natural aging.

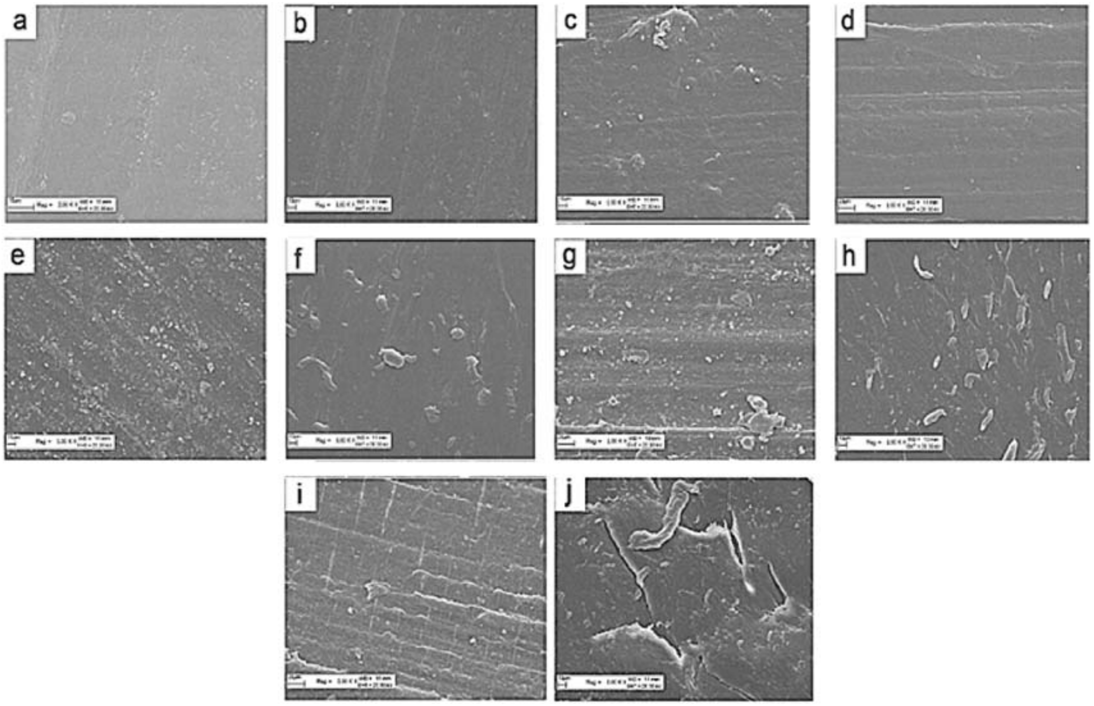


Figure 5. SEM images of PP samples before and after aging in accelerated and natural conditions: a, c, e, g, and i: PP samples after natural aging for 30, 60, 90, 120, and 150, days, respectively; b, d, f, h, and j: PP samples after artificial accelerated aging of 3, 6, 9, 12, and 15 days, respectively

3.3 Capillary Rheometer Analysis

Capillary rheometer analysis investigates the changes in polymer resin which is subjected above its molding temperature. Under the influence of small amount of oxygen during the oxidative degradation of the molecular chains, branched or crosslinked polymer shear viscosity and torque are induced in a closed chamber.[20]

This experiment used a 60 s short reflux capillary rheometer test to avoid oxidative degradation of PP samples by heat or cutting by the barrel shear action to compare the real data of PP samples under artificial ultraviolet-accelerated aging and outdoor aging, which caused polymer degradation by shear viscosity and torque variation. Figure 6(a) showed that the shear rate of polymer melt had a minimal effect on the different degrees of samples torque during PP aging when the speed of the screw was low at 1 rpm. The torque was increased between sample gaps as the speed increased. When the speed increased from 1 rpm to 50 rpm, the torque also increased from 0.039, 0.027, and 0.022 Nm to 0.52, 0.18, and 0.13 Nm after the samples were aged for 0 and 15 days artificially, and 150 days naturally, respectively. This result could be attributed to polymer molecular weight reduction during aging and the viscosity decreased and the torque decreased at the same shear rate.[21]

As shown in Figure 6(b), the measured shear viscosity value of different samples was gradually reduced when the screw speed was 1 rpm at lower melt shear rates. Compared with specimens without aging, the shear viscosity retention rates were 89.7%, 74.7%, 70.1%, 56.2%, and 42.7% after artificial accelerated aging for 3, 6, 9, 12, and 15 days, respectively. Moreover, the values of shear viscosity retention rates were 87.2%, 74.1%, 68.9%, 52.9 %, and 29.2 %, respectively, after natural aging for 30, 60, 90, 120, and 150 days. The artificial accelerated aging and natural aging for 3 and 30 days, 6 and 60 days, or 9 and 90 days had similar shear viscosity values, thereby indicating the consistent extent of artificial and natural aging of pre-aged PP. The natural aging had more serious effects than that inartificial accelerated chamber. When the samples were aged for 15 and 150 days compared with the specimens without aging, the shear viscosity decreased to 86646 Pa/s and 107023 Pa/s, respectively.

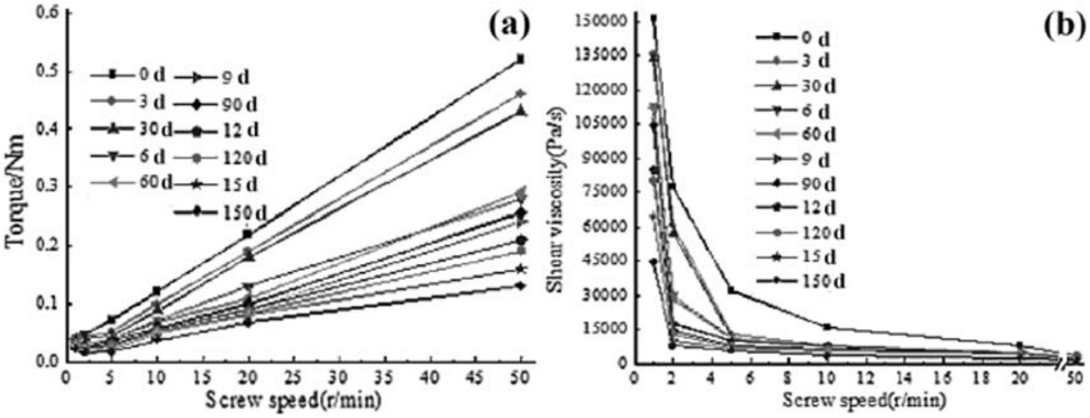


Figure 6. Capillary rheometer analysis of PP before and after aging: (a) relationship between dynamic torque and rotational speed, (b) relationship between surface shear viscosity and rotational speed

3.4 X-ray Photoelectron Spectroscopy Analysis

X-ray photoelectron spectroscopy (XPS) characterized the elemental composition and chemical changes in an aged sample. This analysis has an important role in determining the sample reaction mechanism of aging and in analyzing the indoor and outdoor aging processes that cause structural changes.[22]

The XPS data shown in Figure 7 and Table 4 revealed that the surfaces contained three functional groups, which were C–C, C–O, and C=O bonds, of elemental carbon after the artificial accelerated aging and natural aging of PP. As the aging time increased, C–C bonds on the samples surface were gradually reduced, whereas C-O and C=O bonds were gradually increased. The C=O bonds of PP was 3.06% (caused by thermal aging during injection and molding), 4.4%, and 5.01% after aging for 0 day, 3 days artificially, and 30 days naturally. The binding energy range of C-C bonds was 285. 2 eV to 288.4 eV. However, the C=O binding energy peak value decreased with the aging time. The binding energy peak of C=O bonds was 284.6 eV for artificial and accelerated aging for 15 and 150 days, respectively, because of the degradation of PP molecular chains which their reduced their interaction, thereby reducing the binding energy. Simultaneously, the C=O functional group was increased by 4.73% and 13.46% for artificial and accelerated aging for 15 and 150 days compared with PP without aging. Analysis of XPS data revealed that, after artificial and accelerated of PP for 3 and 30 days, 6 and 60 days, or 9 and 90 days, the number of C-O and C=O bonds on the PP surface were generally the same. The number of C-O and C=O bonds gradually increased with the aging time; the tertiary hydrogen structures of PP produce hydrogen peroxide under heat and oxygen light. Aging becomes serious, such that more hydrogen peroxide groups occur and most of which are transformed into acetone, acetic acid, and methanol.

The PP structure contains a tertiary hydrogen, which can easily produce hydrogen peroxide, thereby speeding up the formation of hydrogen peroxide in the main chain.[23] The presence of peroxide *o*-β hydrogen facilitates the following process:

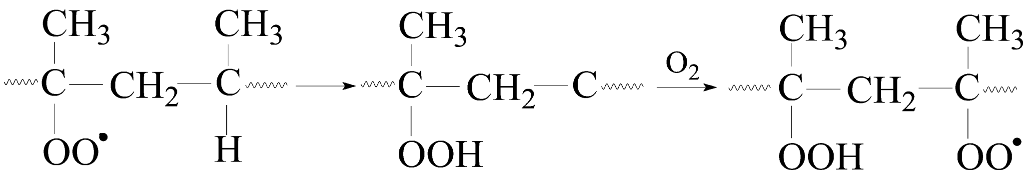


Figure 7. XPS of PP subjected to artificial accelerated and natural aging

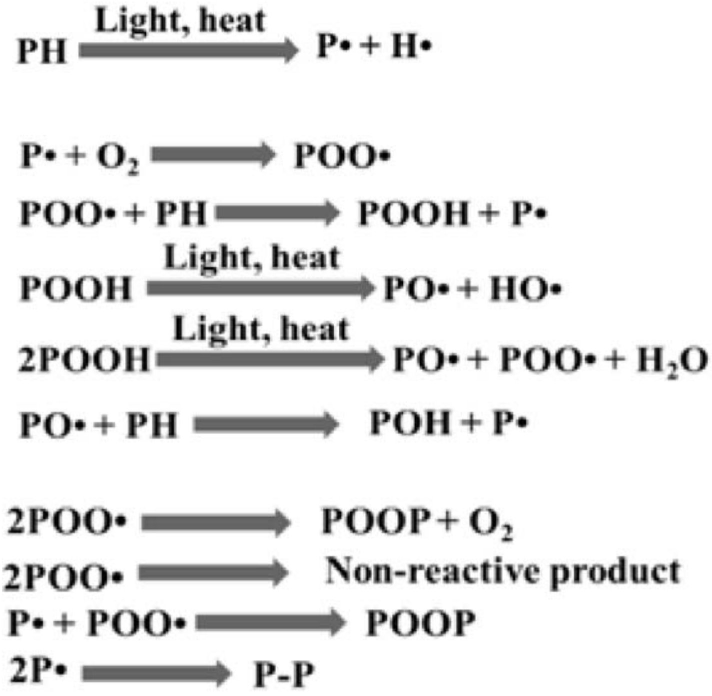
The reaction continues along the chain direction, thereby generating more hydrogen peroxide radicals; thus, chain terminal methyl ketone and volatile products are preferentially formed.[23]

TABLE 4. Changes in the relative molecular mass and distribution of PP samples after artificial and natural aging

Bond	Artificial accelerated aging time/day						Natural aging time/day				
	None	3	6	9	12	15	30	60	90	120	150
C-C/%	96.94	91.88	91.36	90.61	88.28	86.77	91.33	90.87	89.89	82.26	72.44
C-O/%	3.06	4.40	4.55	5.24	7.33	8.56	5.01	5.06	5.73	10.19	14.09
C=O/%	0	3.71	4.09	4.18	4.39	4.73	3.66	4.07	4.41	7.56	13.46

3.5 Aging Mechanism of PP

The factors leading to polymer degradation can be divided into internal and external factors. The internal factors are mainly related to the molecular structure of the polymer, such as the C-F bond energy of polytetrafluoroethylene is 500 kJ/mol, and the C-H bond energy of polypropylene is 410 kJ/mol. In addition, in the polymer oxidation reaction, the rate of hydrogen abstraction from the carbon atom in the oxidation reaction depends on the type of C-H bonds, and the C-H intensity is arranged as follows: primary hydrogen > secondary hydrogen > tertiary hydrogen. The C-H bond in the PP molecule belongs to the tertiary hydrogen, so it is easily oxidized. The molecular structure of PP is related to the synthesis process. Generally, its antioxidant activity is mainly effected by external factors. Among the external factors, the most important is thermal oxygen or photo oxygen degradation. In the oxidative degradation reaction, there are two periodic reactions. During aging, the high molecular polymer is continuously oxidized to form a large number of low molecular compounds, and the reaction process is generally divided into three stages: chain initiation, chain growth (or chain transfer, chain branching), and chain termination:



Chain trigger :

$$PH \xrightarrow{\text{Light, heat}} P\bullet + H\bullet$$

Chain growth:

$$P\bullet + O_2 \longrightarrow POO\bullet$$

$$POO\bullet + PH \longrightarrow POOH + P\bullet$$

$$POOH \xrightarrow{\text{Light, heat}} PO\bullet + HO\bullet$$

$$2POOH \xrightarrow{\text{Light, heat}} PO\bullet + POO\bullet + H_2O$$

$$PO\bullet + PH \longrightarrow POH + P\bullet$$

Chain termination:

$$2POO\bullet \longrightarrow POOP + O_2$$

$$2POO\bullet \longrightarrow \text{Non-reactive product}$$

$$P\bullet + POO\bullet \longrightarrow POOP$$

$$2P\bullet \longrightarrow P\text{-}P$$

In the above processing, PH represents a polymer, and P· represents a polymer macromolecular radical. Firstly, a chain initiation reaction and an oxidation reaction mainly occur. Then, a chain growth reaction (increase in free radicals) occurs mainly. Thus, the macromolecular chain undergoes decomposition, and the molecular weight is greatly reduced, resulting in a decrease in physical and mechanical properties. On the other hand, in the process of reaction, due to disordered cross-linking, disorder control is often formed. Finally, The network structure increases the molecular weight and causes the polymer to become brittle, hard, and elastic.

4. CONCLUSIONS

This work studied the weathering performance of PP by artificial accelerated aging for 15 days and natural aging for 150 days. SEM analysis showed that large crack widths of approximately 1.25 μm and 1.63 μm appealed on the PP surface after 15 days artificial accelerated aging under 340 nm UV lamp and 150 days natural aging in Turpan, respectively. XPS data revealed that the number of C-O and C=O bonds on the PP surface were generally the same after artificial accelerated aging and natural aging of PP for 3 and 30 days, 6 and 60 days, or 9 and 90 days. Moreover, the data of mechanical properties and capillary rheology also exhibited similar laws. Thus, the experimental conditions of the artificial accelerated aging showed that the aging rate was 10 times faster than that in the natural aging conditions of Turpan. By analyzing the surface micro-morphology, element composition, mechanical properties and capillary rheological properties of PP before and after aging, this method of artificial accelerated aging could provide a method to calculate the failure of PP in natural conditions within a certain time (about 5 months). When the aging time continues to increase, this method might no longer be applicable because PP would suffer more severe meteorological environments such as wind, heat and temperature difference when it aged in the natural conditions for a long time. The formulation of artificial accelerated aging method suitable for hot and dry climate could quickly detect and calculate the service life of wire-drawing grade PP and guide the production process such as drip tape and other applications. Formula optimization had important scientific significance and social and economic benefits.

5. ACKNOWLEDGMENTS

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