Revised manuscript number MS #Polymers-414543 entitled “Synthesis and Properties of Plasma-Polymerized Methyl Methacrylate via Atmospheric Pressure Plasma Polymerization Technique” by C.-S. Park et al.

First of all, the authors really appreciate the reviewers’ valuable comments for the paper. Based on the reviewers’ comments, the descriptions for the experimental results are clarified in the revised manuscript. In addition, the related explanations and discussions are compensated and intensified in the revised manuscript. As a result, the X-ray photoelectron spectroscopy (XPS) results in Figure 2 with Table 1, the time of flight-secondary ion mass spectrometry (ToF-SIMS) results in Figure 3, and the film thickness results in Figure 5 are modified in the revised manuscript to express the experimental data more clearly as per the reviewers’ recommendations. In addition, new 13 references [14-24, 50, 51], including the published papers of the group of ‘Morent and De Geyter’, are also provided in the revised manuscript. Total figures, table, and references changed are given as follows.

<table>
<thead>
<tr>
<th>Old Manuscript</th>
<th>Revised Manuscript</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fig. 1</td>
<td>Fig. 1</td>
</tr>
<tr>
<td>Fig. 2</td>
<td>Fig. 2 [Modified]</td>
</tr>
<tr>
<td>Fig. 3</td>
<td>Fig. 3 [Modified]</td>
</tr>
<tr>
<td>Fig. 4</td>
<td>Fig. 4</td>
</tr>
<tr>
<td>Fig. 5</td>
<td>Fig. 5 [Modified]</td>
</tr>
<tr>
<td>Fig. 6</td>
<td>Fig. 6</td>
</tr>
<tr>
<td>Fig. 7</td>
<td>Fig. 7</td>
</tr>
<tr>
<td>Table 1.</td>
<td>Table 1. [Modified]</td>
</tr>
<tr>
<td>[Ref.] 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13</td>
<td>[Ref.] 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13</td>
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<tr>
<td>[Ref.] 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24 [New]</td>
<td>[Ref.] 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49</td>
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<td>[Ref.] 50, 51 [New]</td>
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</table>
III. Upon the reviewer #3’ comments

Authors of “Synthesis and Properties of Plasma-Polymerized Methyl Methacrylate via Atmospheric Pressure Plasma Polymerization Technique” proposed interesting new technology for deposition of transparent pinhole free coatings by plasma technique. The results will be interesting for researchers working in the field of functional layers and plasma polymers. The quality of the coatings in terms of optical properties and homogeneity seems to be very high and therefore these results are very promising. Nevertheless, although this work has satisfactory level of novelty and originality, it needs some revision and corrections.

► We appreciate your kind and detailed assessment of the work presented in this paper.

1. First of all the quality of XPS analysis is not sufficient. The peaks are too broad due to charging effect. The fitting has high error. Either authors must remeasure the spectra or please provide std deviations for all components. It might be useful to adjust the charge compensation during measurements.

► We appreciate your kind and detailed assessment of the work presented in this paper. Since the pPMMA samples and substrates were insulators, the XPS measurement were very difficult to obtain narrow spectra due to charging effect. Therefore, we used an additional electron gun to allow for surface neutralization to adjust the charge compensation during the measurements. According to the reviewer’ comment, to clearly describe this mechanism, the fitting procedure, program, relative sensitivity factors, and FWHM are newly provided in the revised manuscript, and the high-resolution peaks with deconvolutions of XPS spectrum of Fig. 2 are modified in the revised manuscript.

At lines 101-118 on page 3 in Experimental and Characterizations:

“The X-ray photoelectron spectroscopy (XPS) (ESCALAB 250XI surface analysis system, Thermo Fisher Scientific, Waltham, MA, USA) was used to investigate the surface chemical compositions and atomic concentration of the pPMMA films. In the XPS measurement, the voltage and current of the monochromatic Al Kα X-ray source (hv = 1486.7 eV) was 15 kV and 20 mA, respectively. The measurement angle was 60°.
and the measurement depth was estimated to range from 8 to 10 nm. The measurement area was 500 \( \mu \text{m} \times 500 \mu \text{m} \) and the pressure was about \( 10^{-8} \) Pa. The C 1s spectrum (285.0 eV) was used to calibrate the energy scale. Elements present on the deposited surfaces were identified from XPS survey scans and quantified with Thermo Avantage software (v.5.977, Waltham, MA, USA) using a Shirley background and applying the relative sensitivity factors provided by manufacturer of the instrument. The relative sensitivity factors of C 1s and O 1s were 1.0 and 2.8, respectively. For high-resolution spectra, the constant analyzer energy modes were used at 200 eV for survey scan and 50 eV pass energy for element scan, respectively. Since the pPMMA samples and substrates were insulators, we used an additional electron gun to allow for surface neutralization to adjust the charge compensation during the measurements. To curve fit the high-resolution C 1s and O 1s peaks, the deconvolution of C 1s and O 1s peaks was analyzed by the Thermo Avantage. The peaks were deconvoluted using Gaussian–Lorentzian peak shapes (constrained between 80 and 100% Gaussian) and the full-width at half maximum (FWHM) of each line shape was constrained between 2.0 and 3.0 eV.”
2. The binding energies chosen by authors are not fully correct. C1s The C-C component should be placed at BE=285 eV and not at 285.2 and C(O)O is at 289 and not 289.6 eV. O1 The BE of C-O can be around 533 eV but 533.7 eV is too high. It is generally the feature of bad charge compensation.

We appreciate your kind and detailed assessment of the work presented in this paper. The binding energies are re-calculated and modified based on your comment. In addition, according your comment, the fitting procedure, program, relative sensitivity factors, and FWHM are newly provided in the revised manuscript, and the high-resolution peaks with deconvolutions of XPS spectrum of Fig. 2 are modified in the revised manuscript.

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**Fig. 2 [Modified]**
3.
Page 8. Authors should correct their conclusion regarding the hydrophobic nature of coated surface, as \( \text{wca}=75 \) degrees is still hydrophilic angle and Hydrophobic property of the surface led to \( \text{wca}=90 \) or more. Thus they should correct the paragraph. However, authors must explain the increase of \( \text{wca} \) after \( \text{pPMMA} \) coating, because according to their analysis the surface has high density of hydrophilic group (C-O, O-C=O).

► The authors appreciate the in-depth comments from the reviewer. According to the reviewer’s comment, the use of ‘hydrophobic’ are changed to ‘increase of WCA’, and the related sentences are modified in the revised manuscript in order to avoid potential confusion.

At lines 189-191 on page 5 in Results and Discussion:
“...This means that our \( \text{pPMMA} \) surface becomes carbon-rich and eventually it is expected to increase the water contact angles (WCAs) after \( \text{pPMMA} \) coating [40,41].”

At lines 269-280 on page 11 in Results and Discussion:
“...To calculate the surface wettability or hydrophobicity of the newly proposed polymer, WCAs of \( \text{pPMMA} \) were measured [46-49]. The WCAs analysis is a simple, rapid, and direct method to evaluate the hydrophobic or hydrophilic feature of a surface. The variation in the WCAs on the pristine (bare) substrates and \( \text{pPMMA} \) thin films grown on glass and polyethylene terephthalate (PET) substrates when using proposed APP polymerization technique after 90 min deposition is shown in Figure 7. From Fig. 7, after deposition, we can see that the WCAs of the \( \text{pPMMA} \) thin films were gradually increased for both glass and PET substrates. The results of WCA tests showed that the \( \text{pPMMA} \) thin films could increase the WCAs after deposition. The increased WCAs of the \( \text{pPMMA} \) thin films were presumably due to the cleavage of hydrophilic groups and newly formed hydrophobic groups (C-C and C-H).”

4.
There are many small grammar and spelling errors:
Munster must be with capital M. The sentence on lines 203 and 204 has missing verb.
We appreciate your kind and detailed assessment of the work presented in this paper. According to the reviewer’s comment, some words, sentences, and phrases including your comments are modified to improve the English expression including grammar of our paper. In addition, typo error and sentence of manuscript were also double checked.

At lines 122-124 on page 3 in Experimental and Characterizations:

“The surface structure and composition of the pPMMA films were examined by the time of flight-secondary ion mass spectrometry (ToF-SIMS) V instrument (ION-TOF GmbH, Munster, Germany) with a bismuth primary-ion (Bi$_3^+$) gun source.”

At lines 219-221 on page 7 in Results and Discussion:

“The ions at m/z = 15, 27, 31, 39, 41, 55, 59, 69, 77, and 91 were assigned to CH$_3^+$, C$_2$H$_5^+$, CH$_3$O$^-$, C$_3$H$_7^+$, C$_4$H$_8^+$, C$_2$H$_3$O$_2^+$, C$_4$H$_7$O$^-$, C$_8$H$_9^+$, and C$_7$H$_7^+$, respectively.”

At lines 237-239 on page 8 in Results and Discussion:

“As shown in the SEM results of Figure 4, the pPMMA film had a deposition rate of about 0.023 μm·min$^{-1}$, and had no pits and pin holes.”

5.
I recommend to revise this manuscript.

The authors really appreciate the reviewers’ valuable comments for the paper. Based on the reviewers’ comments, the descriptions for the experimental results are clarified in the revised manuscript. In addition, the related explanations and discussions are compensated and intensified in the revised manuscript. As a result, the XPS results in Figure 2 with Table 1, the ToF-SIMS results in Figure 3, and the film thickness results in Figure 5 are modified in the revised manuscript to express the experimental data more clearly as per the reviewers’ recommendations. In addition, new 13 references [14-24, 50, 51], including the published papers of the group of ‘Morent and De Geyter’, are also provided in the revised manuscript. Finally, the authors again thank the reviewer’s efforts for improving the context of this paper.