Response to reviewer comments
Title: Nascent Titanium/Silicon-Containing Particle Formation in Corona Discharge Assisted Combustion
Journal: Aerosol Research
Ref: ar-2025-41

Reviewer #2: Referee's comments:
The work is an experimental study on the effect of corona discharge plasma parameters on metal oxide nanoparticle production in flames.

Response: We thank the reviewer for their comments. We have addressed the comments and questions raised by the reviewer to the best of our abilities and look forward to positive feedback on our revision.

Comment 1: *The introduction section is quite long (nearly 3 pages). This discussion provides relevant and thorough background, but the long discussion makes the thrust of the current work more difficult to follow. Perhaps the goals and objectives of the current work can be made more clear throughout this intro section.*

Response: We thank the reviewer for their feedback. We agree that the manuscript would benefit from a shorter introduction section. Because the knowledge of particle formation and growth in combustion systems is well known, we cut portions of the introduction section that primarily talks about studies which look at particle formation and growth in combustion only systems.

The updated introduction section talks about the demand for nanomaterials, benefits and drawbacks of plasma assisted combustion, and particle formation in plasma assisted combustion systems.

Comment 2: *pg 5 line 142, Why does the stoichiometric flame condition reduce the flame temperature effect of adding precursor?*

Response: The concern that we are trying to address here is that introducing precursors and oxidizing them to form TiO_2 and SiO_2 particles might consume more oxygen that it might alter flame properties, which may impact particle formation and growth. With the measurement of the flame temperature, we can show that even at the highest precursor feed rates used in this study, the flame equivalence ratio still stays close to stochiometric conditions. We have added the following description to the manuscript to clarify this.

"To check if the flame was altered by the consumption of oxygen towards the oxidation of TTIP and TEOS, we calculated the flame equivalence ratio with precursors at their highest feed rates. If the equivalence ratio (ϕ), the fuel-to-oxidizer ratio to the stochiometric fuel-to-oxidizer ratio, shifts significantly towards fuel-rich conditions ($\phi > 1$), we would see the shift reflected in a significant change in flame temperature."

Comment 3: *Is the HRDMA also a TSI instrument?*

Response: The HRDMA is from SEADM Inc. The DMA cell was purchased from SEADM, while the rest of the components like the heat radiator were built in-house. The voltage across the central electrode of HRDMA was applied by the power source Prodisc 20DC and a pump driven by a brushless motor by DOMEL Incorporation provided the sheath flow rate ranging from 100 to 700 lpm. The following description has been added to the manuscript to give the reader more details on the HRDMA:

"The particles formed from the flame are extracted and studied using a hole-in-a-tube (HiaT) type dilution sampling probe (Zhao et al., 2003), SEADM HRDMA, and a TSI 3068A electrometer. Only the DMA cell in HRDMA was purchased from SEADM, while the rest of the components were built in house as described in Wang et al., (2014)"

References:

Wang, Y., Fang, J., Attoui, M., Chadha, T. S., Wang, W. N., and Biswas, P.: Application of Half Mini DMA for sub 2nm particle size distribution measurement in an electrospray and a flame aerosol reactor, J. Aerosol Sci., 71, 52–64, https://doi.org/10.1016/j.jaerosci.2014.01.007, 2014.

Zhao, B., Yang, Z., Wang, J., Johnston, M. V., and Wang, H.: Analysis of soot nanoparticles in a laminar premixed ethylene flame by scanning mobility particle sizer, Aerosol Science and Technology, 37, 611–620, https://doi.org/10.1080/02786820300908, 2003.

***Comment 4:** Why is air used for dilution of the metal oxide aerosol? Is the probe inlet temperature low enough to prevent further oxidation by air in the dilution probe flow?*

Response: Since we used a dilution ratio of 152, the temperature after mixing is sufficiently low that it can reduce the further oxidation by air in the dilution probe. Assuming the flow is well mixed in the sampling probe, the resulting flow temperature will be 302.6 K, which is relatively low for further reactions. We also measured the size distributions of the particles to ensure that the normalized size distributions do not change as we further increase the dilution ratios, meaning that further reactions and particle dynamics are quenched in the sampling probe (Wang, 2017; Zhao et al., 2003). The following description about probe inlet temperature has been added to the methods section of the manuscript:

"Due to dilution, the temperature after mixing is sufficiently low that it can reduce further oxidation by air in the dilution probe. We also measured the size distribution of particles to ensure that the normalized size distributions do not change as we further increase the dilution ratios, meaning that further reactions and particle dynamics are quenches in the sampling probe (Wang, 2017; Zhao et al., 2003)."

References:

Wang, Y.: Sub 2 nm Particle Characterization in Systems with Aerosol Formation and Growth, Washington University in St. Louis, 2017.

Zhao, B., Yang, Z., Wang, J., Johnston, M. V., and Wang, H.: Analysis of soot nanoparticles in a laminar premixed ethylene flame by scanning mobility particle sizer, Aerosol Science and Technology, 37, 611–620, https://doi.org/10.1080/02786820300908, 2003.

***Comment 5:** What are details of the thermocouple probe? Type? Shape? Size? Could there be energy losses to the temperature probe affecting the interpretation of the temperature readings?*

Response: For temperature measurements in this study, we used a type S thermocouple (Pt – 10% Rh) with a spherical bead of bead diameter 0.7 mm. There are radiative energy losses at the thermocouple surface. We have corrected for the energy losses at the thermocouple and the temperature that we have reported in Fig. 1b shows the corrected temperature.

The following description has been added to the manuscript to reflect this:

"For flame temperature measurement, we used a type S thermocouple (Pt – 10% Rh) with an exposed uncoated spherical bead of diameter 0.7 mm. In Fig. 1b, we can see the temperature profile which takes into account the radiative heat losses on the thermocouple bead surface."

Comment 6: *Is the thermocouple grounded or protected from plasma interference?*

Response: The thermocouple, with an exposed junction, is not grounded and is protected from plasma's interference. When measuring the temperature of the flame, we ensure to place the thermocouple at a higher distance away from the discharge electrodes than the distance between the discharge electrodes. This way we ensure that there is no arcing or any other type of plasma's interference on the thermocouple.

Comment 7: *pg 8, How is the flame height of 6.35 mm defined?*

Response: The flame height was measured based on an image where the O.D of inner tube was used as a reference length. The inner tube has an O.D of 6.35 mm. In previous version of our manuscript that we submitted, we incorrectly reported the flame height as 6.35 mm. After verifying our measurements, we obtained a flame height of 4.38 mm. The image of the flame with scale has been added to Fig. 1 of the manuscript.

"The flame height was measured optically using the images shown in Fig. 1b. The length for reference in these images is the width of the inner stainless-steel tube, which has an outer diameter of 6.35 mm."

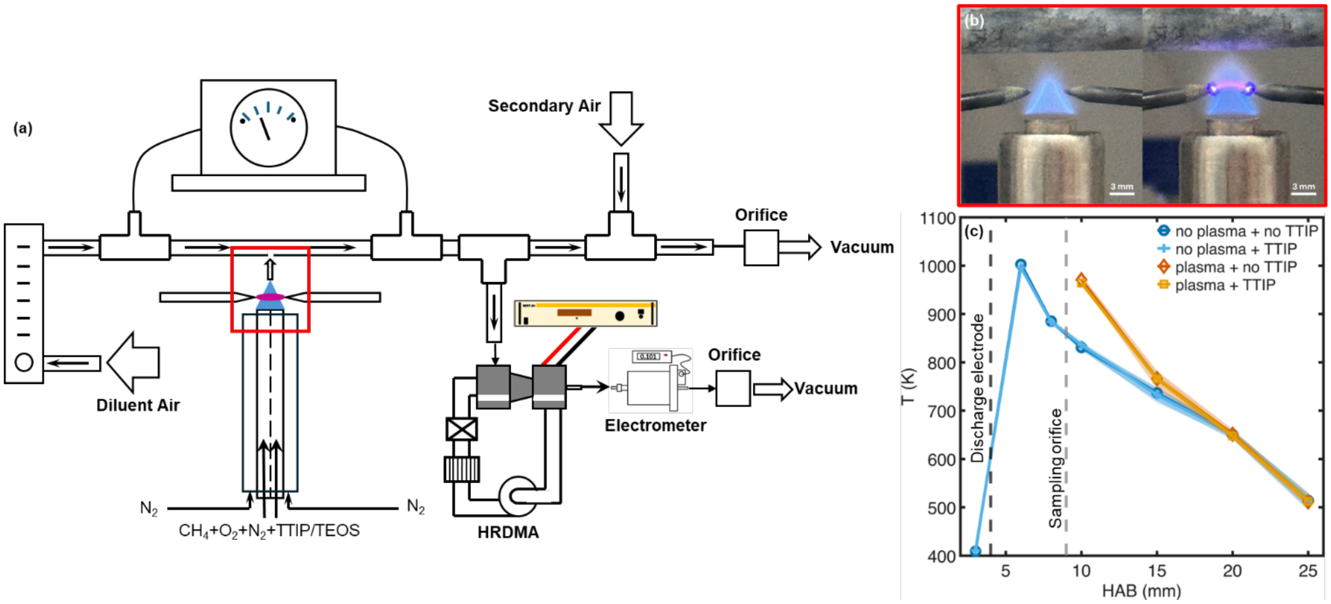

Figure 1: (a) Schematic of the experimental apparatus with a red box highlighting the flame and corona discharge region. (b) Image of flame with and without a corona discharge in the highlighted region. (c) Temperature profile of flames under different plasma and synthesis precursor conditions. The feed rate of TTIP is 19 mg h^{-1} for temperature profile measure, and the plasma power is 125 W.

Comment 8: *In industrial applications, the precursor concentrations are in the heavily loaded regime, would the currently observed plasma affects apply by simply increasing the ion concentration to match the heavy precursor concentration?*

Response: Based on the results of our study, in the heavily loaded regime, increasing ion concentration to match the precursor concentration should give similar results. This result is also supported by our previous work done on characteristic charging time and coagulation time (Wang et al., 2017). We have added the following description to the discussion and conclusion sections:

“In industrial applications, the precursor concentrations are in the heavily loaded regime. Based on the results of this study, in the heavily loaded regime, increasing ion concentration to match the precursor concentration should give similar results. This result is also supported by our previous work done on characteristic charging and coagulation time (Wang et al., 2017a).”

“Based on the results of this study, for industrial applications in the heavily loaded regime, increasing ion concentration to match the precursor concentration should give similar results.”

References:

Wang, Y., Sharma, G., Koh, C., Kumar, V., Chakrabarty, R., and Biswas, P.: Influence of flame-generated ions on the simultaneous charging and coagulation of nanoparticles during combustion, Aerosol Science and Technology, 51, 833–844, https://doi.org/10.1080/02786826.2017.1304635, 2017.

We thank the reviewer for these helpful comments. This has improved our manuscript and we look forward to the paper being accepted for publication.