Supplement of: Direct Measurement of the Viscosity of Ternary Aerosol Mixtures

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Design and Performance Characteristics of the Thermal Conditioner

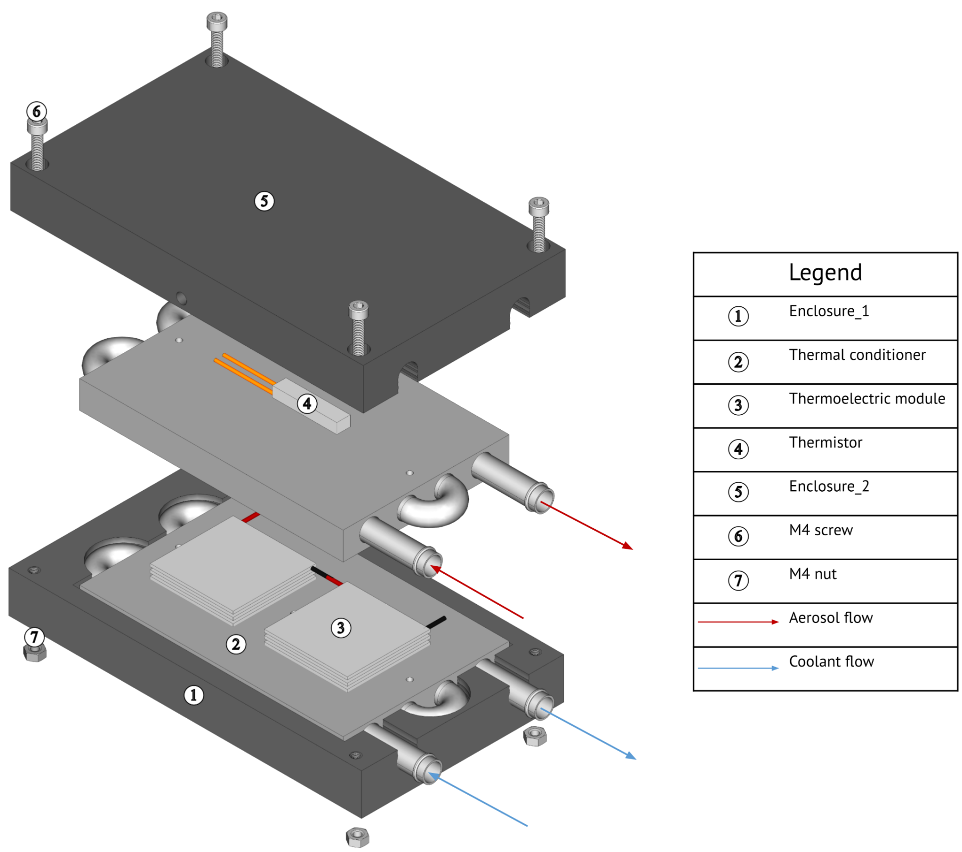

Figure S1. Schematic design of the revised thermal conditioner.

A schematic of the design of the thermal conditioner is provided in Figure S1. The thermal conditioner consists of two heat exchanger loops that were removed from a commercial air-cooled Peltier-thermoelectric liquid cooler (TE Technology, LC-061). The top loop is used to thermally condition the aerosol sample flow. The bottom loop is used to heatsink two 48W two-stage thermoelectric modules (CustomThermoelectric, 25412-5L31-07CQQ). The bottom loop is cooled with fluid from a chilled bath circulator (PolyScience, Model 9102) operated at

5 °C. Liquid cooling was used to avoid unnecessary heat sources inside the thermally insulated box (Figure 1, main manuscript). Temperature was measured by mounting a 15 kΩ thermistor (TE Technology, MP-3193) to the outside of the metal plate. The thermoelectric modules are operated in series and powered using a 24V DC 12.5 A power supply and controlled using a bipolar proportional-integral-derivative temperature controller (TE Technology, TC-36-25-RS232). Cover plates were custom designed and printed using a 3D extrusion printer using polyethylene terephthalate glycol (PETG) plastic. The cover plates provide thermal insulation and structural stability of the device. This thermal conditioner can be heated up to 100 °C and cooled ~50 °C below the temperature of the heatsink. A different thermistor is needed if cooled below −20 °C.

Figure S2 shows an example temperature scan using this device. Unlike Figure 3 in the main manuscript, there is no thermal lag. This conditioner was used for the ternary mixture experiments.

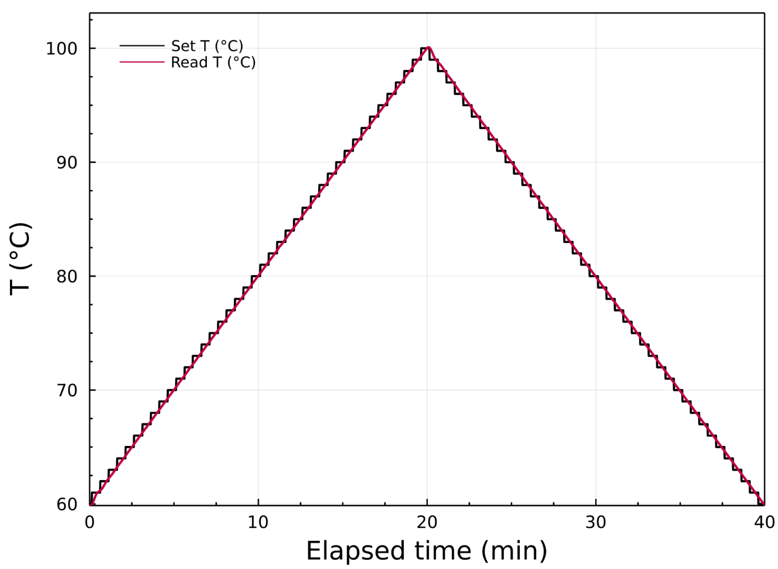

Figure S2. Set point and read temperature of an example scan obtained with the revised thermal conditioner. Steps of the setpoint temperature are due to an unintentional rounding to the nearest integer when writing the output data file. The rounding is caused by a typo in the data acquisition code, which specified %3.f versus the intended %.3f as string format specifier.

Table S1. Summary of all experiments. Numbers in columns (a)-(c) denote the fractional concentration (weight), date and time indicate the timestamp of the start of the scan, D_p indicates the mobility diameter selected by the DMA in nm, $RH \pm \sigma_{RH}$ denote the mean and standard deviation of the relative humidity in % of the sample flow, $T_s \pm \sigma_{Ts}$ denotes mean and standard deviation the temperature inside the enclosure in °C. A_1 and A_2 denote the μ_{PH} before and after transition for the fitted model (Eq. 2). T_t and σ_{Tt} denote the fitted transition temperature and standard deviation for each experiment in °C.

Suc[a]	Cit[b]	Tar[c]	Date	Time	D_p	RH	σ_{RH}	T_s	σ_{Ts}	a	b	T ramp	A_1	A_2	T_t	σ_{Tt}
[-]	[-]	[-]	[yyyymmdd]	[h:m:s]	[nm]	[%]	[%]	[°C]	[°C]	[-]	[-]	[°C]	[-]	[-]	[°C]	[°C]
1	0	0	20220825	16:29:30	250	-	-	-	-	160	480	60-100	359.3	376.1	83.7	4.9
1	0	0	20220829	14:22:00	300	-	-	-	-	160	480	60-100	374.5	439.7	84.8	2.8
1	0	0	20220826	10:57:50	350	-	-	-	-	160	480	60-100	367.4	442.7	86.9	1.8
1	0	0	20220826	11:46:40	400	-	-	-	-	320	960	60-100	794.0	853.5	87.1	1.5
1	0	0	20220829	12:42:00	450	-	-	-	-	600	1040	60-100	912.5	944.8	88.2	1.5
1	0	0	20220826	13:30:00	500	-	-	-	-	600	1040	60-100	915.8	966.8	88.7	1.6
1	0	0	20220826	14:19:20	550	-	-	-	-	720	1440	60-100	1228.7	1301.9	88.8	1.3
1	0	0	20220829	15:03:00	600	-	-	-	-	800	1600	60-100	1380.8	1490.8	89.4	1.3
1	0	0	20220826	16:26:00	650	-	-	-	-	800	2000	60-100	1694.3	1838.1	89.2	1.6
1	0	0	20220826	17:12:00	700	-	-	-	-	1200	2400	60-100	2042.8	2215.5	89.8	1.3
Experiments below this line used the setup in Figure 1, above this line the setup in Section 2.3.																
1	0	0	20220926	15:26:00	300	3.1	0.04	5.6	0.02	250	500	60-100	410.0	476.9	88.3	1.8
0.8	0.2	0	20220928	14:37:10	300	1.6	0.03	8.0	0.05	250	500	40-80	448.2	474.6	70.1	1.6
0.6	0.4	0	20220928	17:34:00	300	2.0	0.04	7.6	0.05	250	500	35-75	442.0	474.7	56.3	4.2
0.4	0.6	0	20221012	16:25:15	300	1.4	0.03	7.8	0.07	250	500	25-65	458.6	475.7	44.7	3.6
0.2	0.8	0	20220929	12:28:05	300	2.0	0.05	8.0	0.06	250	500	15-55	454.2	470.0	39.1	2.9
0	1	0	20220928	16:29:30	300	1.9	0.04	7.6	0.02	250	500	5-45	458.3	470.5	27.6	2.9
0.8	0	0.2	20221007	11:53:55	300	1.2	0.03	8.5	0.41	250	500	45-85	410.0	476.9	72.1	2.7
0.6	0	0.4	20221007	12:40:43	300	1.2	0.03	7.9	0.04	250	500	35-75	442.2	466.0	59.0	2.7
0.4	0	0.6	20221007	13:32:05	300	1.3	0.05	7.7	0.01	250	500	25-65	447.1	471.9	47.2	7.4
0.2	0	0.8	20221007	14:15:35	300	1.3	0.03	7.7	0.02	250	500	15-55	452.1	474.5	38.5	2.6
0	0	1	20220929	16:41:45	300	2.4	0.04	8.5	0.03	250	500	5-45	454.7	471.3	26.9	2.5
0	0.8	0.2	20220929	13:20:15	300	2.1	0.04	8.6	0.02	250	500	15-45	455.4	467.8	27.6	1.0
0	0.6	0.4	20220929	14:13:05	300	2.2	0.04	8.6	0.02	250	500	5-45	458.3	470.5	25.6	0.8
0	0.4	0.6	20220929	14:58:00	300	2.3	0.03	7.7	0.02	250	500	5-45	456.9	467.4	27.1	2.4
0	0.2	0.8	20220929	15:43:45	300	2.3	0.04	7.7	0.02	250	500	5-45	456.1	465.8	26.6	1.7
0.6	0.2	0.2	20221008	11:57:25	300	1.1	0.06	8.7	0.46	250	500	35-75	456.0	466.7	56.0	3.4
0.4	0.4	0.2	20221007	16:18:45	300	1.4	0.03	7.7	0.02	250	500	25-65	455.3	465.8	47.2	1.8
0.4	0.2	0.4	20221007	17:05:00	300	1.4	0.02	7.7	0.02	250	500	25-65	455.4	467.8	46.4	3.4
0.2	0.6	0.2	20221008	13:00:26	300	1.2	0.02	7.7	0.02	250	500	15-55	460.0	470.7	37.2	4.7
0.2	0.4	0.4	20221008	13:43:22	300	1.3	0.05	7.6	0.02	250	500	15-55	453.8	471.2	34.9	5.2
0.2	0.2	0.6	20221008	14:26:30	300	1.3	0.05	7.5	0.01	250	500	15-55	452.3	471.6	37.7	3.2

[a]Sucrose [b]Citric acid [c]Tartaric acid

Viscosity Calculation

Rothfuss and Petters (2016)[1] showed that the particle geometry factor ξ is related to the angle of attachment θ given by Pokluda et al. (1997)[2] via

$$\xi = (1 + cos(\theta))^2 \quad \text{(S1)}$$

Pokluda et al. (1997)[2], Table 1, gives simulated values "dimensionless neck" vs. "dimensionless time". Dimensionless neck is defined as x/a where x is the radius of the neck and a is the radius of the partially coalesced sphere and $\sin(\theta) = x/a$. Dimensionless time is defined as

$$t = \frac{\tau\sigma}{a_{mono}\eta}, \quad \text{(S2)}$$

where τ tau is the residence time, σ is the interfacial tension, a_{mono} is the radius of the monomer, and η is the viscosity.

Solving equation S1 for θ allows calculation of x/a from the geometry factor via

$$x/a = sin(acos(\sqrt{\xi} + 1)) \quad \text{(S3)}$$

Using linear interpolation between x/a and t from Table 1 in Pokluda et al. (1997) provides the appropriate value t_ξ. Solving Eq. (S3) fo η then yields the estimated viscosity from numerical integration

$$\eta = \frac{\tau\sigma}{a_{mono} t_\xi} \quad \text{(S4)}$$

For a D = 300 nm sphere equivalent particle, a_{mono} = 119 nm. For ξ = 2.5, τ = 11s, σ = 0.084 J m-2, and a_{mono} = 119 nm, the viscosity is 6.85 x 10^6 Pa s. A spreadsheet with the appropriate calculations is included as part of the final data repository.

References

(1) Rothfuss, N. E.; Petters, M. D. Coalescence-Based Assessment of Aerosol Phase State Using Dimers Prepared through a Dual-Differential Mobility Analyzer Technique. *Aerosol Sci. Technol.* **2016**, *50* (12), 1294–1305. https://doi.org/10.1080/02786826.2016.1221050.

(2) Pokluda, O.; Bellehumeur, C.; Vlachopoulos, J. Modification of Frenkel's Model for Sintering. *AIChE J.* **1997**, *43*, 3253–3256. https://doi.org/10.1002/aic.690431213.