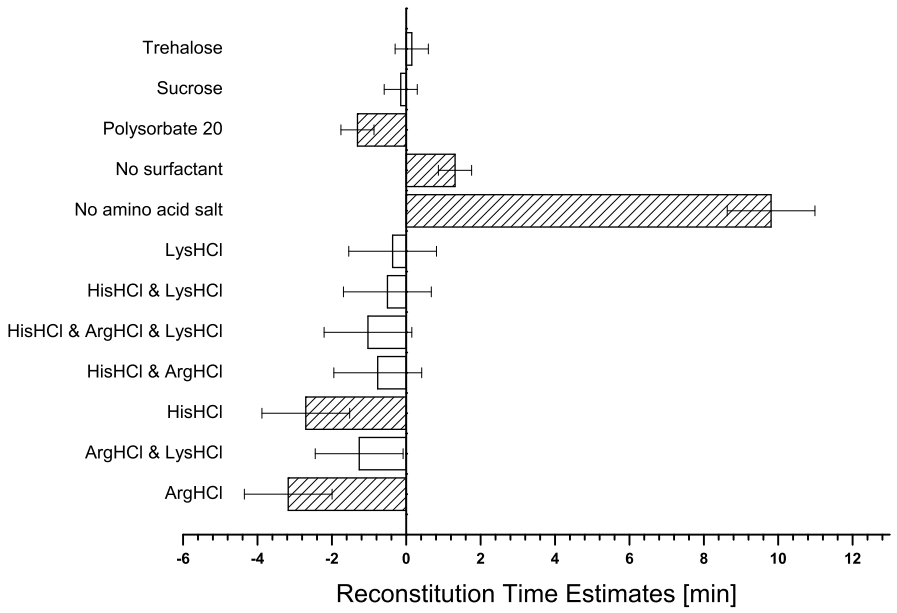
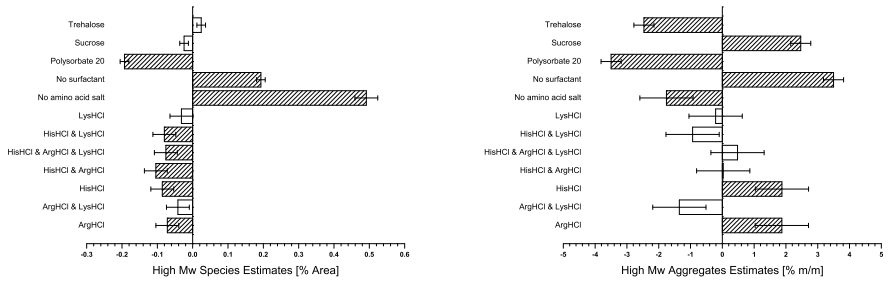


Figure 1: Expanded parameter estimates plot for the reconstitution time response of the spray dried formulations summarised in Table 2. The model was calculated using the values measured (n = 3) for samples stored at 5 °C. Open bars identify non-significant parameters, while parameters that are statistically significant at the 0.05 level are identified by shaded bars. Error bars depict the estimated standard error for each of the estimated parameters. Intercept estimate = 18.69 ± 0.45 (SE) minutes.



(a) (b)

Figure 2: Expanded parameter estimates plot for the (a) high Mw species with the intercept estimate equalling 5.19 ± 0.01 (SE) % Area and (b) high Mw aggregates response with the intercept estimate equalling 6.33 ± 0.32 (SE) % m/m of the spray dried and reconstituted formulations summarised in Table 2. The models were calculated using the values measured (n = 3) for samples stored at 5 °C. Open bars identify non-significant parameters, while parameters that are statistically significant at the 0.05 level are identified by shaded bars. Error bars depict the estimated standard error for each of the estimated parameters.

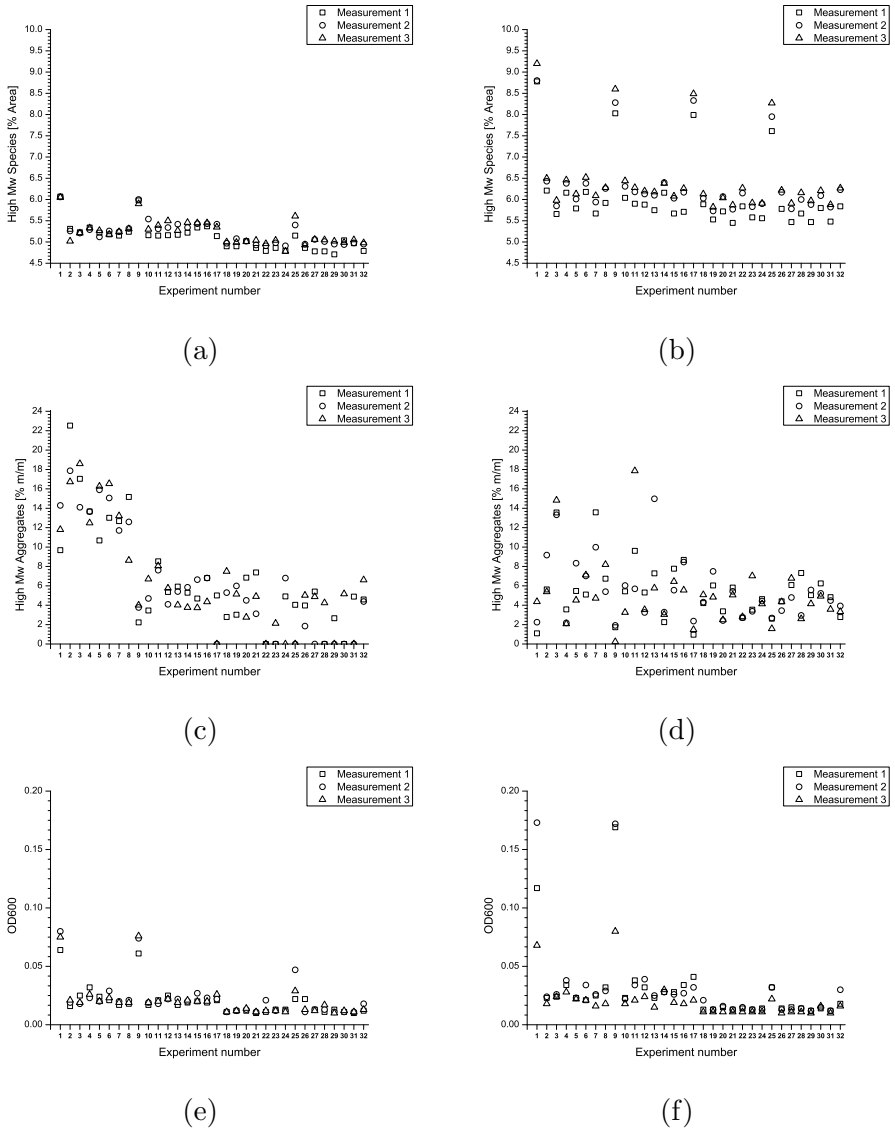


Figure 3: Stability responses measured for the formulations summarised in Table 2 after spray drying, storage and reconstitution. (a) High Mw species values measured for samples stored at 5 °C; (b) High Mw species values measured for samples stored at 40 °C during 4 weeks;(c) High Mw aggregates values measured for samples stored at 5 °C; (d) High Mw aggregates values measured for samples stored at 40 °C during 4 weeks; (e) OD600 values measured for samples stored at 5 °C; (f) OD600 values measured for samples stored at 40 °C during 4 weeks.

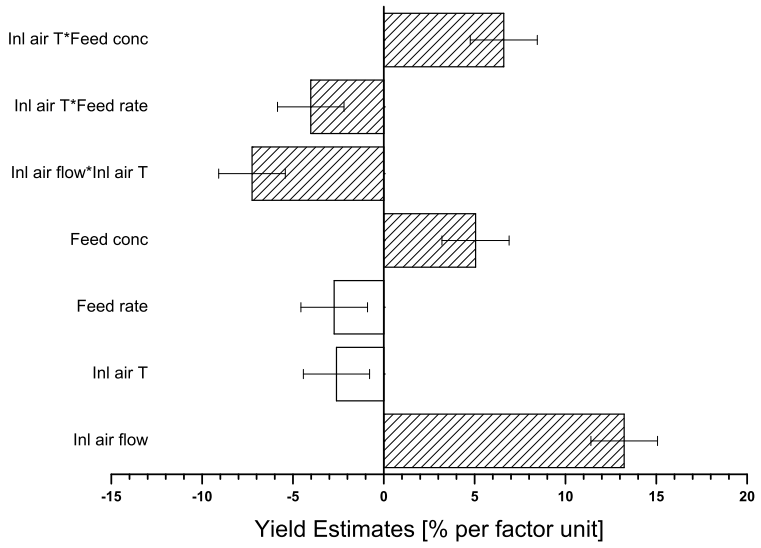


Figure 4: Parameter estimates plot of the model fitted for the yield response (n=1) of the SD process parameter design (Table 4). Open bars identify non-significant parameters, while parameters that are statistically significant at the 0.05 level are identified by shaded bars. Error bars depict the estimated standard error for each of the estimated parameters. Intercept estimate = 30.88 $\pm$ 1.74 (SE) %.

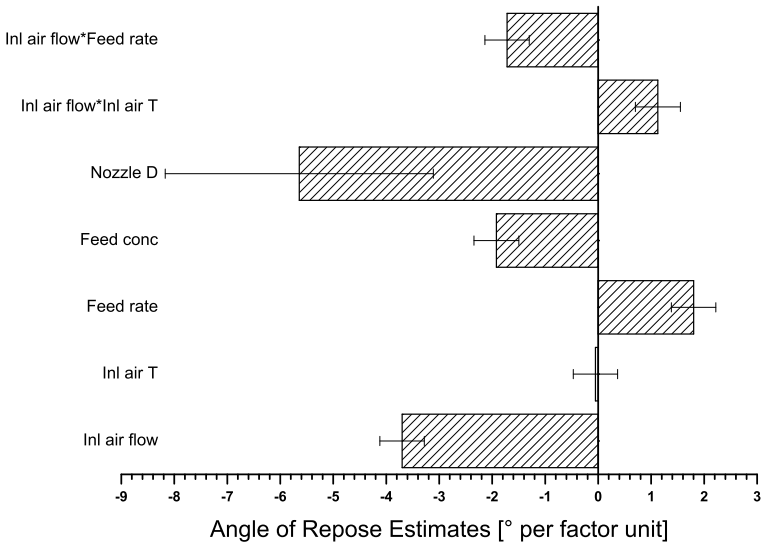


Figure 5: Parameter estimates plot of the model fitted for the angle of repose response of the SD process parameter design (Table 4). The model was calculated using the values measured (n = 3) for samples stored at 5 °C. Open bars identify non-significant parameters, while parameters that are statistically significant at the 0.05 level are identified by shaded bars. Error bars depict the estimated standard error for each of the estimated parameters. Intercept estimate = 48.30 ± 1.55 (SE) °.

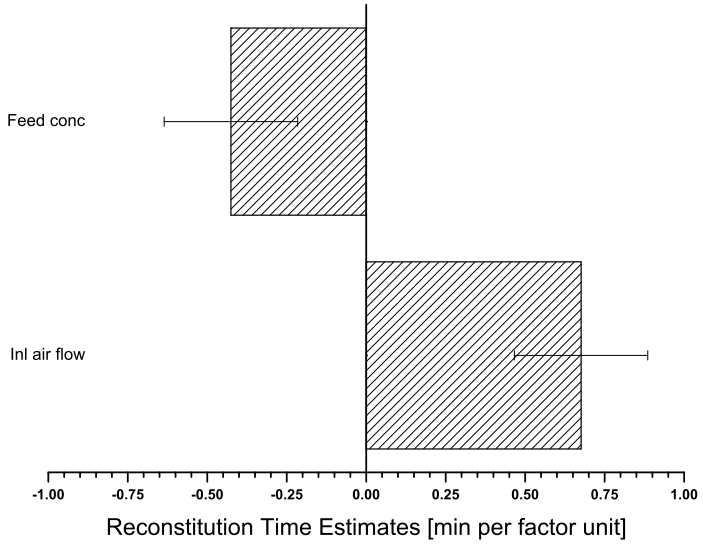


Figure 6: Parameter estimates plot of the model fitted for the reconstitution time response of the SD process parameter design (Table 4). The model was calculated using the values measured (n = 2) for samples stored at 5 °C. Open bars identify non-significant parameters, while parameters that are statistically significant at the 0.05 level are identified by shaded bars. Error bars depict the estimated standard error for each of the estimated parameters. Intercept estimate = 5.60 ± 0.20 (SE) minutes.

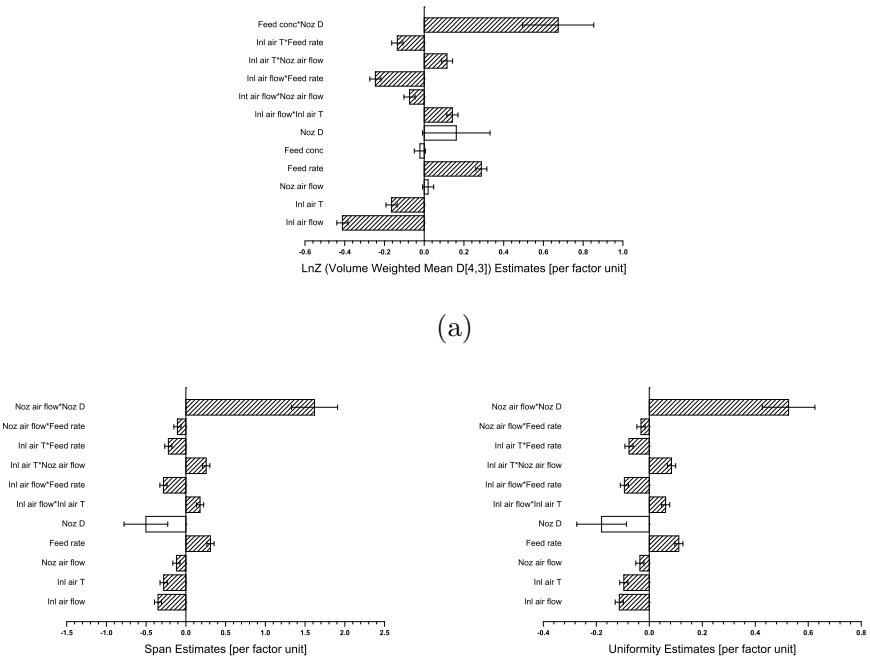


Figure 7: Parameter estimates plot of the models fitted for the particle size distribution responses of the SD process parameter design (Table 4). The model was calculated using the values measured (n = 3) for samples stored at 5 °C. Open bars identify non-significant parameters, while parameters that are statistically significant at the 0.05 level are identified by shaded bars. Error bars depict the estimated standard error for each of the estimated parameters. (a) Intercept estimate = 2.16 ± 0.10 (SE); (b) Intercept estimate = 2.27 ± 0.17 (SE); (c) Intercept estimate = 0.75 ± 0.06 (SE).

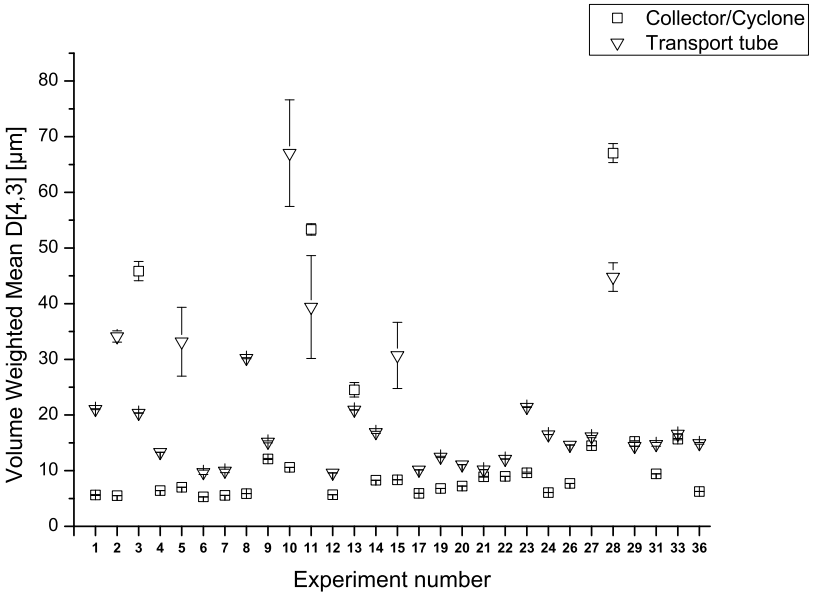


Figure 8: Volume weighted mean D[4,3] of collector/cyclone and transport tube samples (n=3) of the powders resulting from the SD process parameter design (Table 4). Particle sizes for experiments 16, 18, 25, 30, 32, 34 and 35 could not be measured because the quantity of powder recovered from the transport tube was insufficient. Standard deviations are depicted by the error bars.

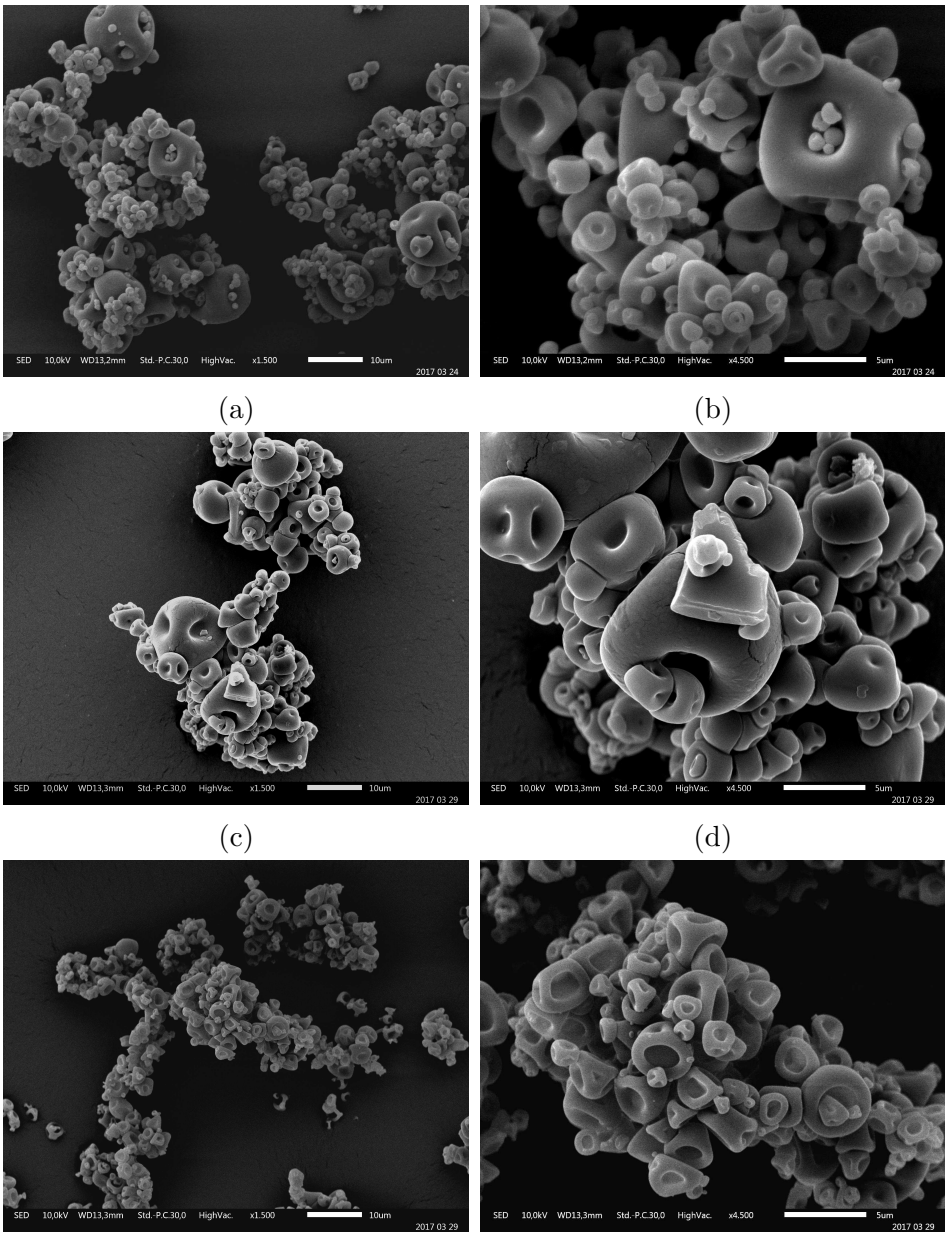
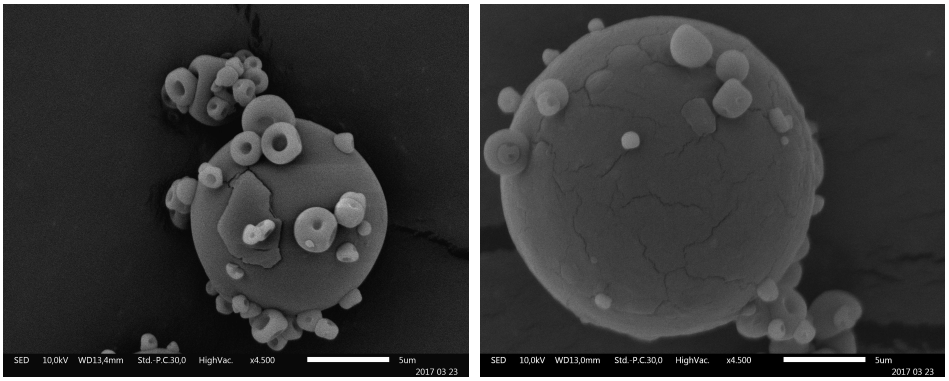


Figure 9: SEM images of selected powders from the SD process parameter design (Table 4) with comparable values for their average (n=3) volume weighted mean D[4,3]. (a) & (b) SEM images of the powder resulting from experiment 20 with average (n=3) volume weighted mean D[4,3] = 7.235 ± 0.010 (SD) μm; (c) & (d) SEM images of the powder resulting from experiment 26 with average (n=3) volume weighted mean D[4,3] = 7.701 ± 0.004 (SD) μm; (e) & (f) SEM images of the powder resulting from experiment 32 with average (n=3) volume weighted mean D[4,3]= 7.052 ± 0.393 (SD) μm.



(a) (b)

Figure 10: SEM images of spherical particles present in selected powders from the SD process parameter design (Table 4). (a) SEM image of the powder resulting from experiment 01; (b) SEM image of the powder resulting from experiment 09.

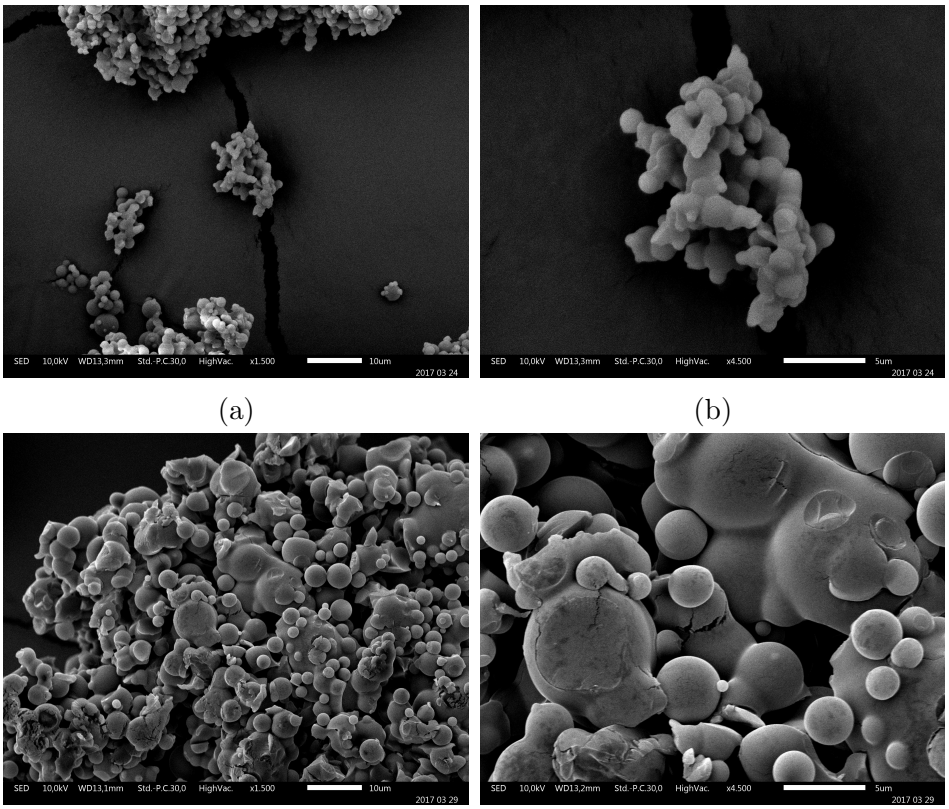


Figure 11: SEM images of strongly associated agglomerates present in selected powders from the SD process parameter design (Table 4). (a) & (b) SEM images of the powder resulting from experiment 11 at x1500 and x4500 magnification, respectively; (c) & (d) SEM images of the powder resulting from experiment 28 at x1500 and x4500 magnification, respectively.

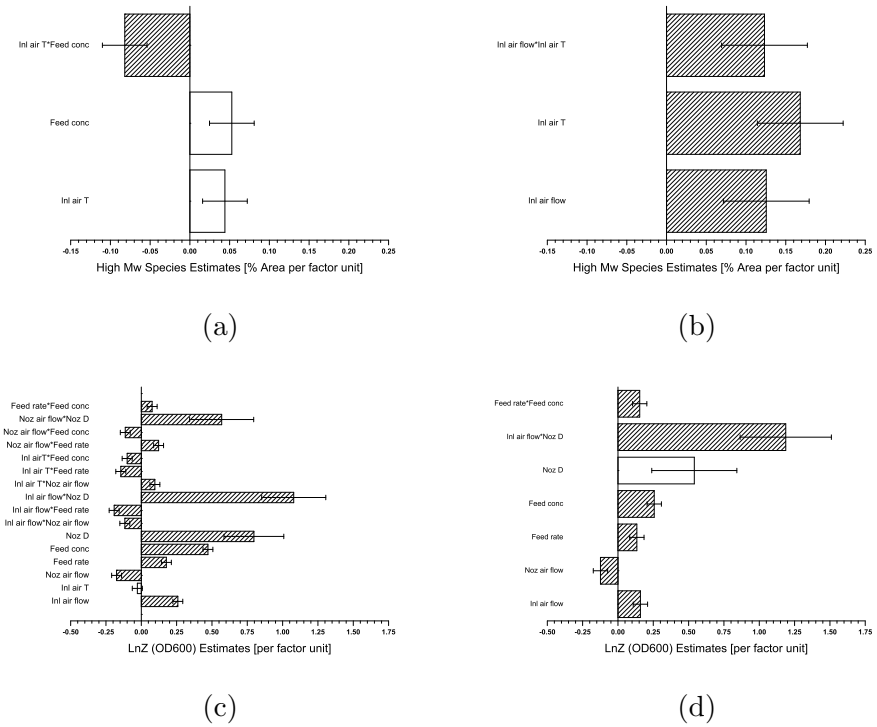
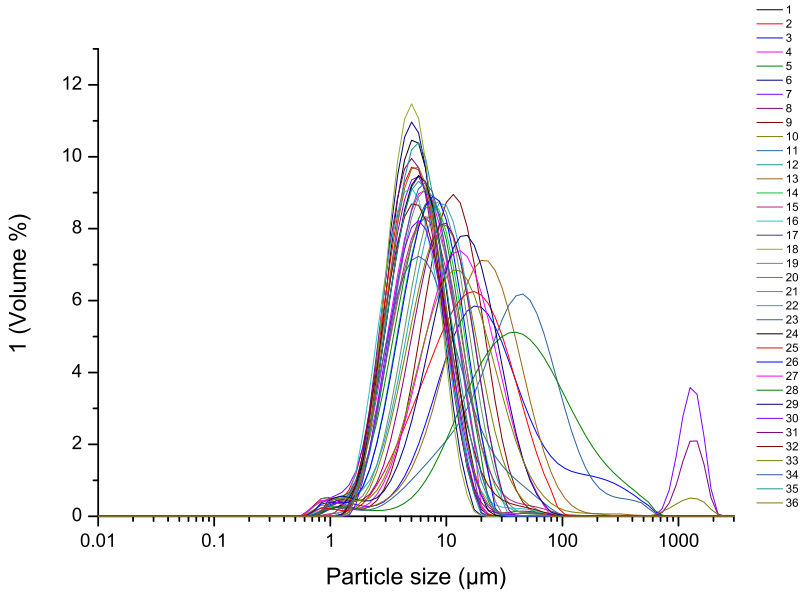
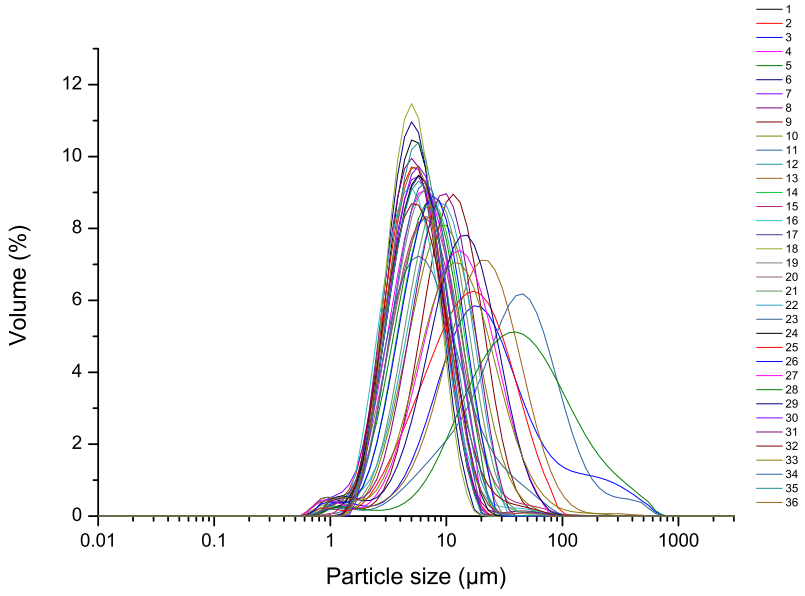


Figure 12: Parameter estimates plot of the models fitted for the high Mw species (n=1) and OD600 (n=3) responses of the SD process parameter design (Table 4). Open bars identify non-significant parameters, while parameters that are statistically significant at the 0.05 level are identified by shaded bars. Error bars depict the estimated standard error for each of the estimated parameters. (a) Parameter estimates plot of the high Mw species response of samples stored at 5 °C. Intercept estimate = 6.68 ± 0.03 (SE) % Area; (b) Parameter estimates plot of the high Mw species response of samples stored at 40 °C during 4 weeks. Intercept estimate = 8.61 ± 0.05 (SE) % Area; (c) Parameter estimates plot of the LnZ (OD600) response of samples stored at 5 °C. Intercept estimate = -3.90 ± 0.13 (SE); (d) Parameter estimates plot of the LnZ (OD600) response of samples stored at 40 °C during 4 weeks. Intercept estimate = -3.70 ± 0.19 (SE).

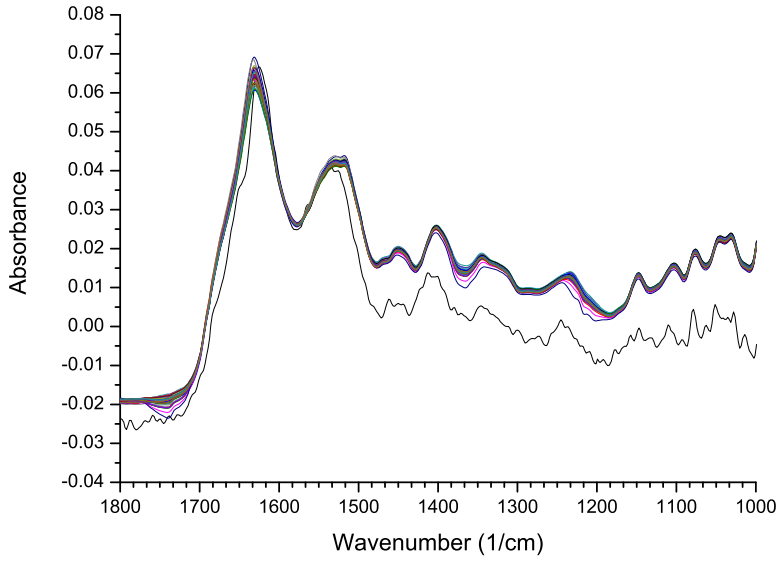


(a)

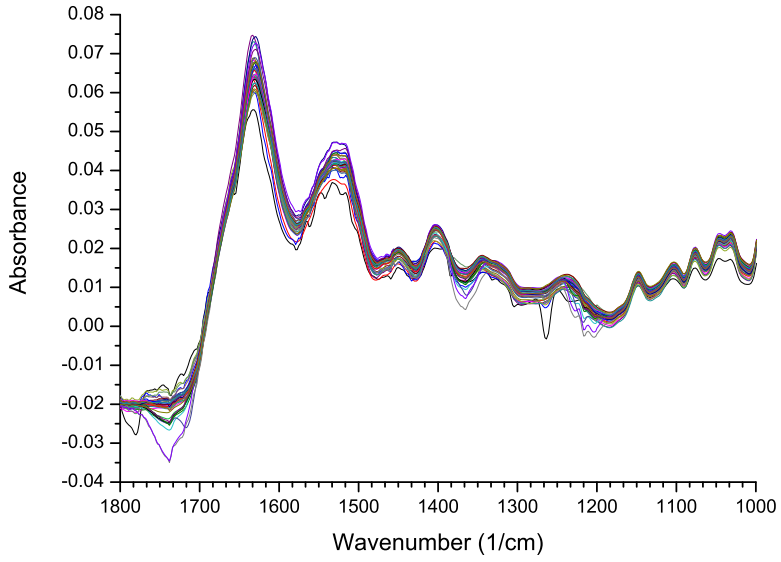


(b)

Figure 13: Overlay of the particle size distribution curves of all powders collected from the SD process parameter design (Table 4) and stored at 5 °C, with numbers in the legend indicating the respective experiment numbers. (a) The particle size distribution curves with the default 2000 μm cut-off applied for all samples and (b) the particle size distribution curves with the 512 μm cut-off applied for powders resulting from experiments 30, 31 and 33.



(a)



(b)

Figure 14: Overlay of the FTIR spectra of all powders collected from the SD process parameter design (Table 4) (a) after storage at 5 °C and (b) after storage at 40 °C during 4 weeks.