

Contents

Nomenclature.....	V
1 Introduction.....	1
2 State of the art	3
2.1 Precipitation process	3
2.1.1 Theory of silica particle synthesis	3
2.1.1.1 Polymerization and primary particle formation.....	5
2.1.1.2 Aggregation and gelation.....	7
2.1.1.3 Fragmentation and reorganization	11
2.1.1.4 Stabilization and functionalisation	12
2.1.2 Relevance of precipitated silica aggregates	13
2.2 Dispersion process.....	14
2.2.1 Wetting behavior	17
2.2.2 Stabilization.....	17
2.2.3 Stress mechanism	20
2.2.3.1 Dissolver	23
2.2.3.2 Stirred media mill.....	24
2.2.4 Dispersion kinetics	25
2.3 Micromechanical aggregate properties	27
2.3.1 Deformation behavior.....	27
2.3.2 Conditions for aggregate breakage.....	30
2.3.3 Simulation of aggregate deformation and breakage behavior	32
2.3.4 Aggregate strength models.....	36
2.3.5 Nanoindentation.....	37
2.3.5.1 Fundamentals of Nanoindentation.....	38
2.3.5.2 Nanoindentation of particles and colloidal systems	43
3 Experimental procedures	45
3.1 Analytical aggregate characterization techniques	45
3.1.1 Dynamic light scattering	45
3.1.2 Laser diffraction.....	46
3.1.3 X-ray-diffraction	47
3.1.4 BET method (specific surface measurement)	47
3.1.5 Mercury porosimetry	48
3.1.6 SEM and SEM-FIB imaging	49
3.2 Experimental set-up – procedure	49
3.2.1 Precipitation process	50

3.2.2	Dispersion process	52
3.2.2.1	Dissolver	53
3.2.2.2	Stirred media mill - precipitated silica	54
3.2.2.3	Stirred media mill - silica model aggregate system	56
4	Material – synthesis and characterization	59
4.1	Precipitated nanostructured silica aggregates.....	59
4.2	Model aggregate system	61
5	Micromechanical aggregate properties	65
5.1	Establishment of the measurement method	65
5.1.1	Nanoindentation measurement system	65
5.1.2	Sample preparation.....	66
5.1.3	Measurement procedure	68
5.1.4	Analysis of the nanoindentation results	69
5.2	Characterization of aggregate deformation.....	71
5.2.1	Interpretation of measurement data	71
5.2.2	Local and global sample stressing	72
5.2.3	Stressing model silica aggregates.....	76
5.2.3.1	Effect of primary particle size.....	76
5.2.3.2	Effect of the strength and stiffness of solid bridges.....	78
5.2.3.3	Determination of solid bridges.....	81
5.2.3.4	Analytical modeling of the micromechanical properties.....	82
5.2.4	Stressing precipitated silica aggregates	85
5.2.4.1	Effect of precipitation temperature	85
5.2.4.2	Influence of mechanical stressing during synthesis	89
5.2.4.3	Effect of precipitation additives	95
5.3	Characterization of aggregate breakage.....	101
5.3.1	Interpretation of measurement data	101
5.3.2	Determination of the breakage behavior	102
5.4	Effect of measurement condition during nanoindentation	104
5.4.1	Effect of loading rate and multiple aggregate stressing	104
5.4.2	Effect of aggregate diameter.....	104
5.4.3	Influence of the drying temperature	106
6	Modeling the micromechanical properties	107
6.1	DEM-contact model.....	107
6.1.1	Particle-Particle contact model	108
6.1.2	Particle-geometry contact model	110
6.1.3	Dimensioning.....	110

6.1.4	Aggregate build-up.....	111
6.2	Modeling the nanoindentation process.....	111
6.2.1	Sensitivity analysis of contact model parameters.....	111
6.2.2	Crack and deformation pattern.....	114
6.2.3	Aggregate deformation and breakage characteristics.....	116
6.2.4	Effect of aggregate structure and location of primary aggregate cracks on tangential stress tensors	118
6.2.5	Effect of the aggregate structure on indentation forces and correlation to aggregate strength models	121
7	Correlation with dispersion processes.....	123
7.1	Kinetic model for the dispersion process.....	123
7.2	Correlation of process and formulation parameters during synthesis with dispersion results.....	126
7.3	Correlation of aggregate structure and micromechanical properties with dispersion results.....	128
7.3.1	Determination of the stress energy	129
7.3.2	Representation of the fracture behavior.....	130
7.3.3	Correlation of the stress energy- and breakage energy distribution.....	131
7.3.4	Comparison of the effective dispersion fraction with dispersion results.....	132
8	Conclusion and future prospects	135
9	Bibliography.....	139
10	Appendix.....	157