Effect of Filler Distribution on Fracture Resistance of Modern Dental Composites

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Objective: To investigate how various filler-matrix systems (hybrid, microfill, and nanohybrid) with different microstructural morphologies (irregular versus spherical fillers), filler sizes and distributions (unimodal, bimodal, or multimodal), and matrix chemistries (methacrylate, ormocer, dimer acid-base, giomer, or DX-511 monomers) can influence the ability of a restorative material to resist fracture.

Background: Modern dental restorative composites exhibit features on more than one length scale, ranging from nano- and micro-fillers, to the polymerized macro-state of a high-molecular-weight polymeric matrix. This heterogeneous system can play a vital role in determining the bulk material properties and is likely to contain defects or flaws, ranging from millimeters down to nanometers or atomic scale, which can give rise to viscoelastic behavior, manifested as elastic bending or torsion. However, if stresses applied to this "composite solid" are too excessive, these structural flaws can become unstable and propagate catastrophically, culminating in bulk fracture. Thus, failure of dental composite restorations is closely associated with the fracture processes of the filler-matrix systems [1, 2]. It is the interest of this study to explore the relationship between structural defects and fracture resistance and to understand the effect of a heterogeneous system like dental composite, which contains at least two hierarchy levels, on fracture mechanics. The null hypothesis is: There exists no causal relationship between the dependent variable, fracture toughness, and the included independent variables such as matrix chemistries and filler morphologies, distributions, and size for various modern filler-matrix systems such as hybrid, microfill, and nanohybrid composites.

Materials and Methods

- Twenty rectangular, single-edge notch specimens (2.75 x 5 x 25 mm^3) per composite brand (n = 20 brands, see Table 1) were made from a stainless steel mold with a razor blade insert, producing a 2.5 mm notch depth.
- The samples were tested using a universal testing machine (Instron ElectroPuls E3000).
- Fracture toughness, K_{IC} [MPa $m^{0.5}$], values were calculated via measurements from the 3-point bending test (span = 20 mm, cross-head speed = 0.5 mm/min) applied on the single-edge notched-bend specimens.
- Filler and matrix microstructural features were analyzed by scanning electron microscopy.
- Filler sizes and distributions were assessed via a dynamic light scattering approach, which probes the Brownian motion of particles in a liquid suspension.
- First, a 0.3 ± 0.05 g sample per composite brand was weighed.
- Second, this sample was dispersed in 20 ml aliquot of acetone and ethanol and agitated to maximize particle de-agglomeration.
- Finally, filler sizes and distributions were measured by a laser particle analyzer (Zetasizer Nano ZS).
- Filler content was measured by thermogravimetric analysis.
- All specimens were stored in 37°C distilled water for 24 hours prior to testing.
- Statistics: Data were analyzed with ANOVA/Tukey (α=0.01) and regression.

References

[1] Randolph LD et al. Dent Mater 2016; 32(12):1586-1599.
[2] Ilie N et al. Clin Oral Investig 2012; 16(2):489-98.

Results: See Figures: 1 – 5. Fracture toughness values were found – rankings in descending order were: Nanohybrids > Hybrids > Microfills. Additionally, composites with multimodal distribution demonstrated significantly less fracture resistance than composites with either unimodal or bimodal distribution.

Table 1: Salient features of materials tested in this study

	Classification		Filler Weight [%] Mean	SD		Filler Size [nm] Min	Max	Range	K_{IC} [MPa $m^{0.5}$] Mean	SD	
Activia Bioactive	RMGI	Bimodal	55.17	0.48	l	91.28	1990	1899	1.48	0.22	b
Admira Fusion	Nanohybrid	Bimodal	84.90	1.43	b	43.82	2669	2625	0.85	0.07	jk
Beautifil II	Hybrid	Bimodal	79.00	0.10	c	164.20	5560	5396	1.14	0.08	fgh
Beautifil Bulk	Hybrid	Bimodal	74.71	0.08	de	50.75	5560	5509	1.07	0.07	gh
Beautifil Bulk Flow	Hybrid	Bimodal	68.40	0.49	i	342.00	3091	2749	1.34	0.10	c
Clearfil Majesty Posterior	Nanohybrid	Bimodal	88.90	0.23	a	32.67	4801	4768	1.29	0.09	cde
Durafill VS	Microfill	Multimodal	57.18	0.16	kl	68.06	1990	1922	0.72	0.03	kl
EPIC TMPT	Microfill	Multimodal	46.59	0.07	m	43.82	1106	1062	0.84	0.09	jk
Filtek One Bulk	Nanohybrid	Bimodal	71.43	0.28	gh	141.80	4801	4659	1.55	0.08	b
Filtek Supreme Ultra	Nanohybrid	Bimodal	72.80	0.21	efg	190.10	5560	5370	1.18	0.08	defgh
GC Kalore	Nanohybrid	Unimodal	69.65	0.14	hi	122.40	3091	2969	1.35	0.14	c
N 'Durance	Nanohybrid	Multimodal	75.00	0.71	d	37.84	3580	3542	0.92	0.06	ij
Point 4	Hybrid	Unimodal	73.84	0.00	def	105.70	1718	1612	1.06	0.13	hi
Renamel Microfill	Microfill	Multimodal	57.53	0.33	k	24.36	2305	2281	0.67	0.07	l
Renamel Nano Plus	Nanohybrid	Bimodal	77.96	0.38	c	50.75	4801	4750	1.10	0.14	fgh
Sonic Fill 2	Hybrid	Bimodal	72.26	0.35	fg	91.28	2669	2578	1.16	0.13	efgh
Tetric Evo Ceram Bulk	Hybrid	Bimodal	73.19	0.20	defg	50.75	3091	3040	1.19	0.12	efg
TPH Spectra ST (HV)	Nanohybrid	Bimodal	73.40	0.05	defg	141.80	4801	4659	1.27	0.09	cdef
Venus Diamond	Nanohybrid	Bimodal	78.17	0.78	c	105.70	3091	2985	1.70	0.12	a
Venus Diamond Flow	Nanohybrid	Bimodal	62.98	1.11	j	141.80	4145	4003	1.31	0.13	cd

Within the column, the same case letter is not significantly different than each other ($p > 0.01$)
RMGI = resin modified glass ionomer; SD = standard deviation; K_{IC} = fracture toughness

Figure 1: Effect of matrix chemistry and filler morphology, system, and distribution on fracture toughness. Columns with the same case letters are not statistically significant ($p > 0.01$).

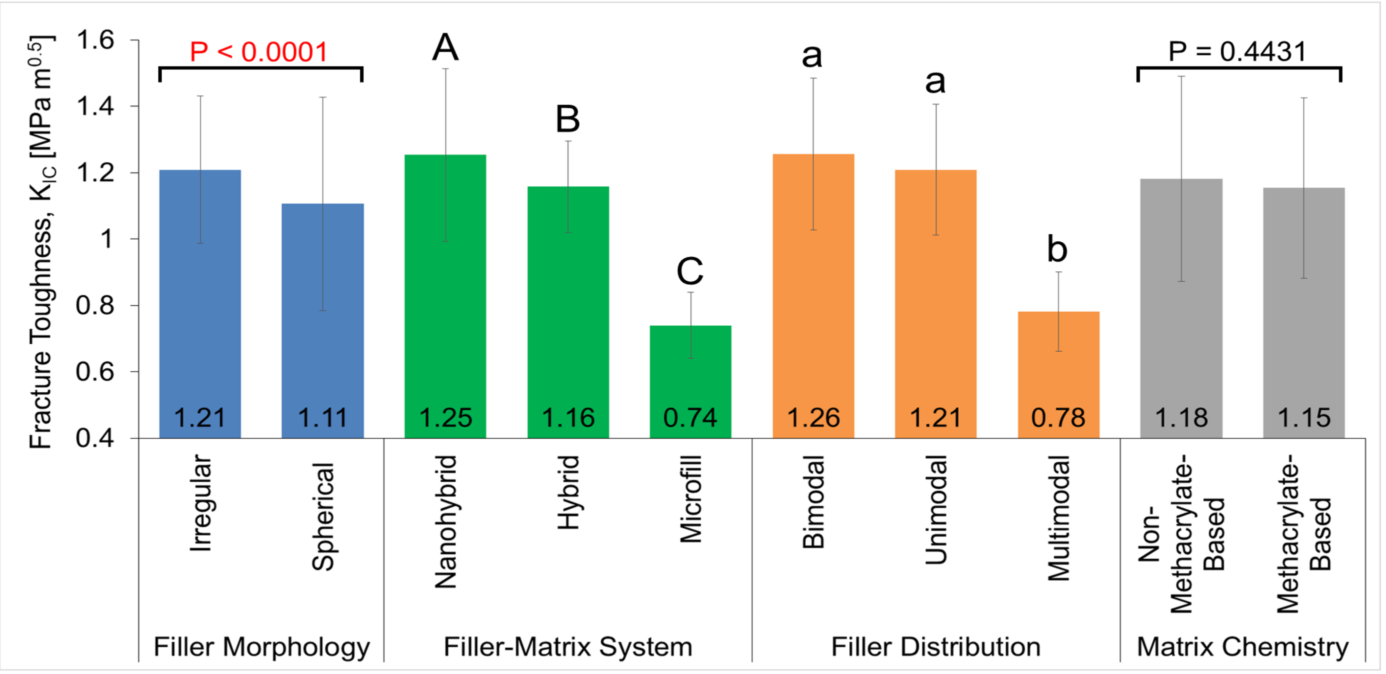


Figure 2: Fracture toughness as a function of filler weight.

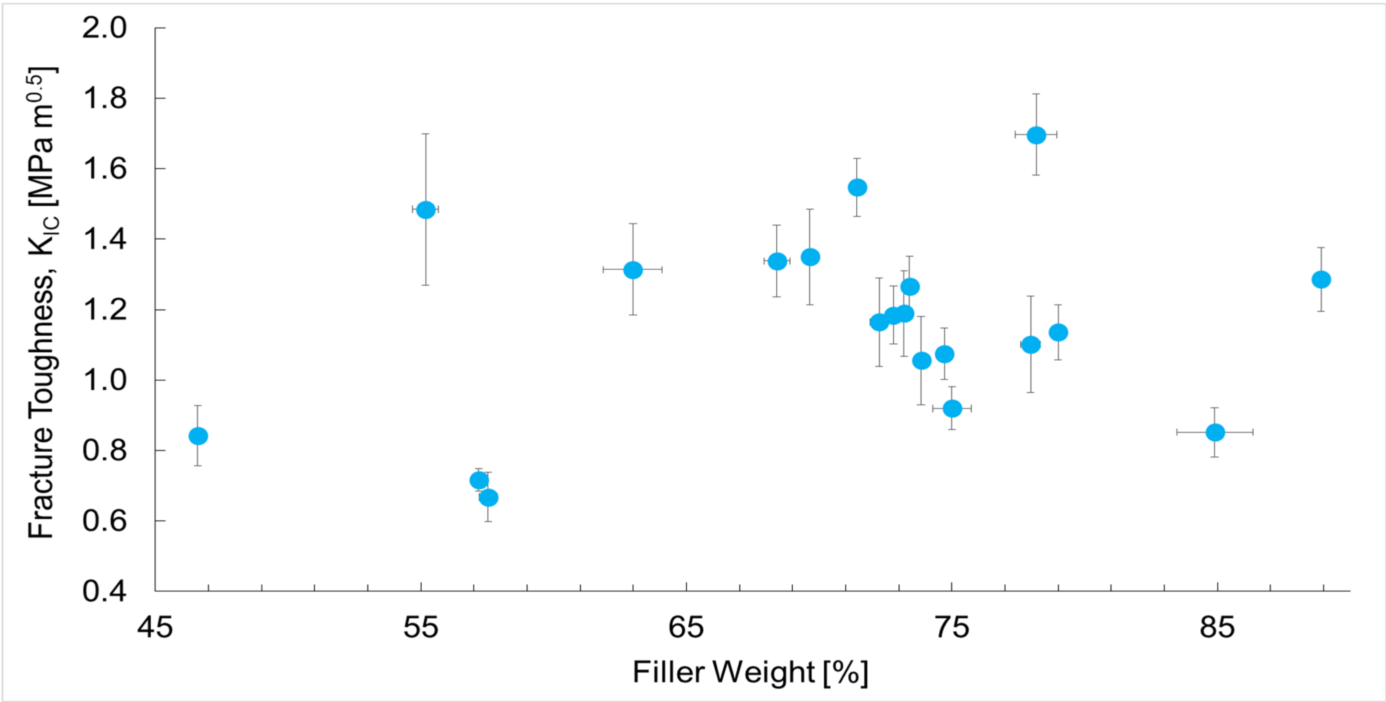


Figure 3: Fracture toughness as a function of filler median size [nm].

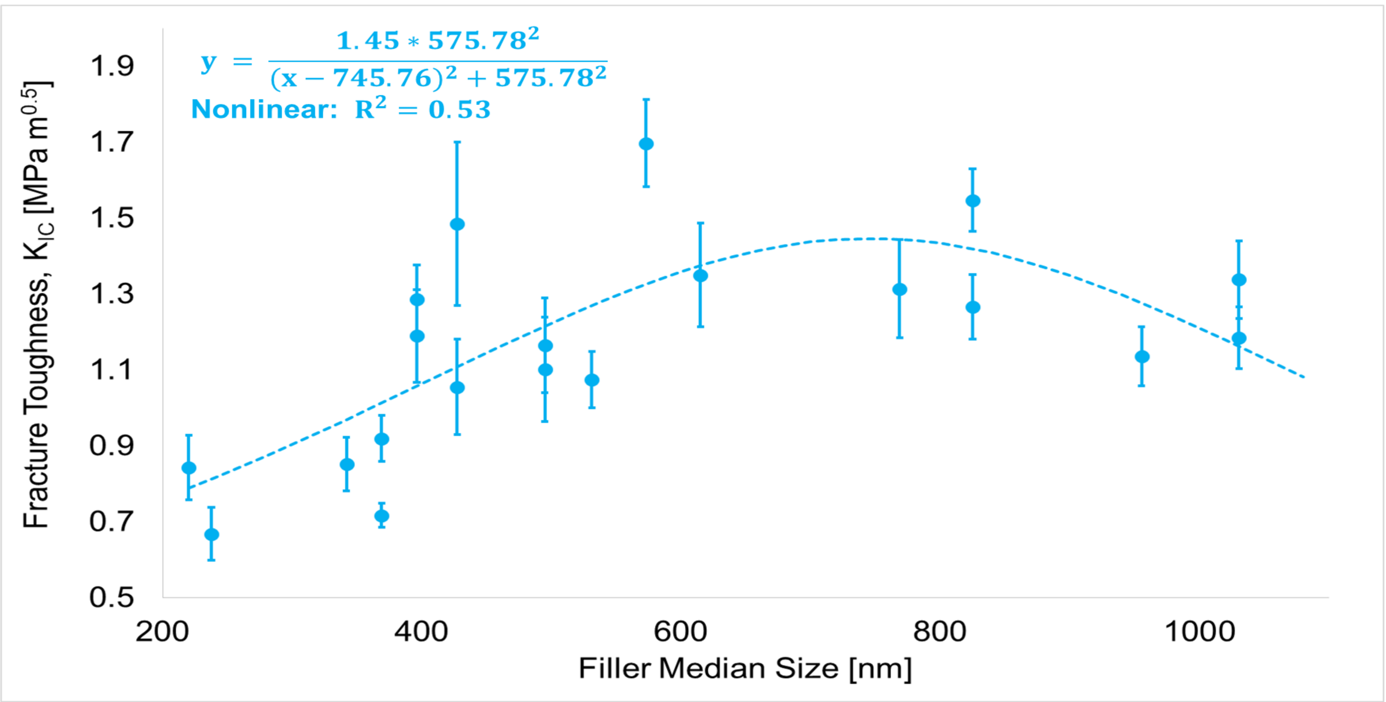
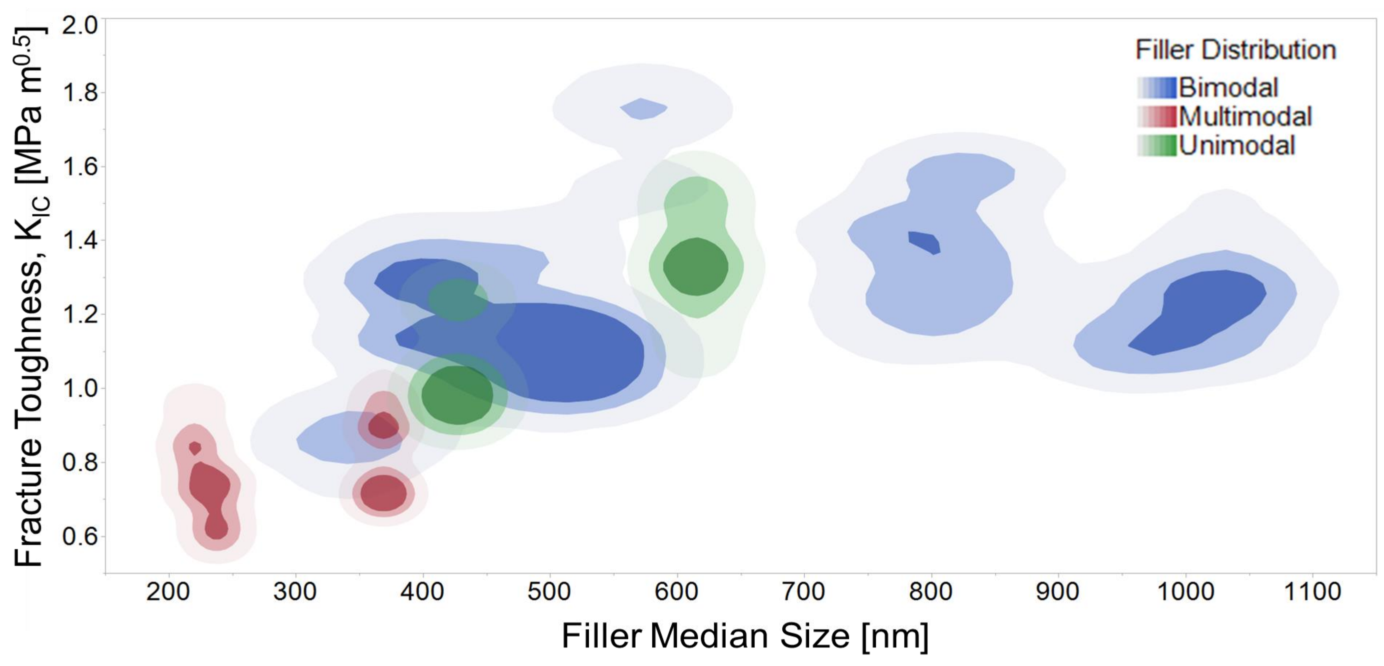


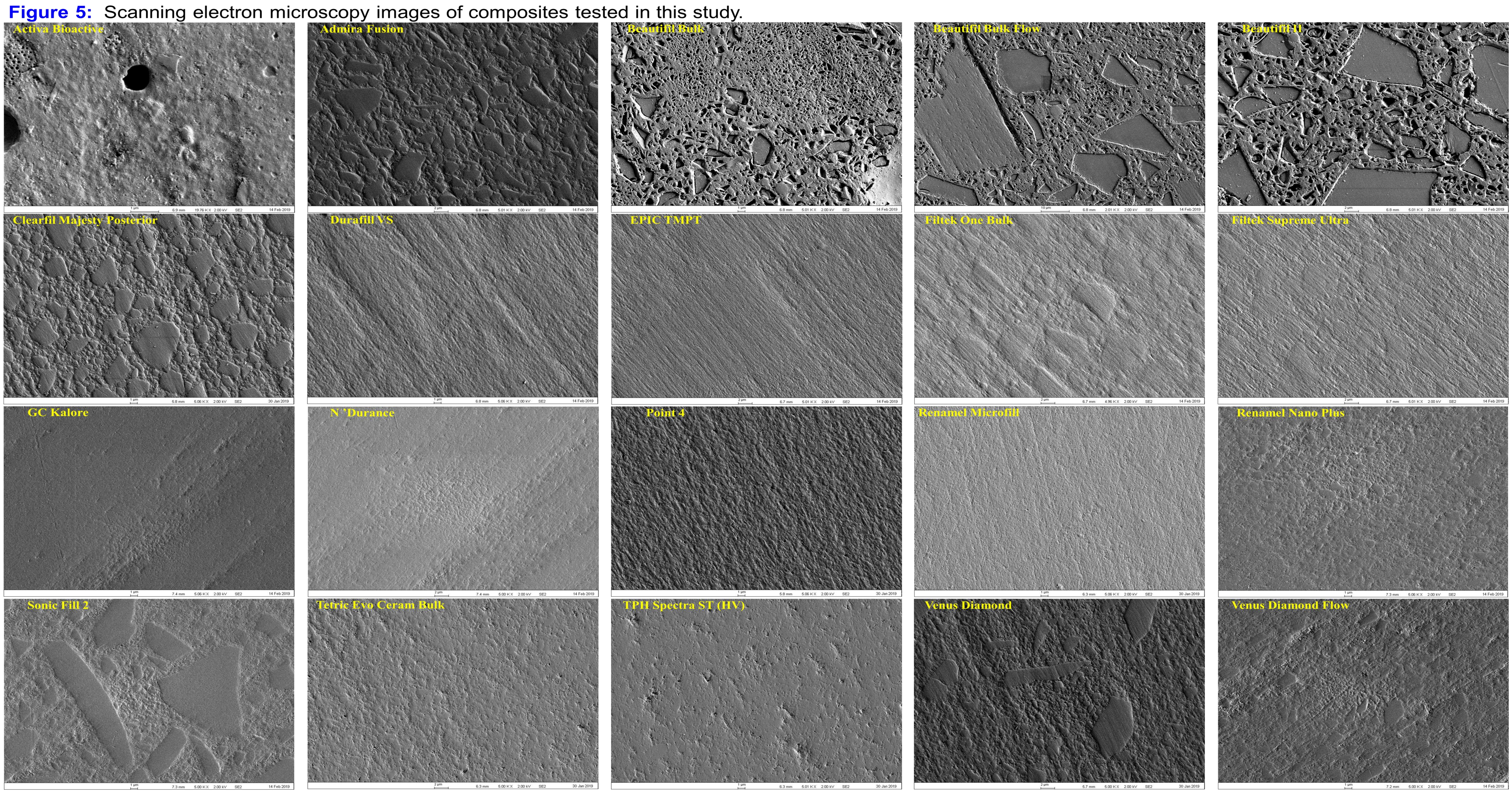
Figure 4: Fracture toughness as a function of filler median size [nm] with special emphasis on mapping clusters of Unimodal, Bimodal, and Multimodal distributions.



Conclusion

- Fracture toughness as a function of filler content increased with percent filler weight until a critical value of 75%, after which K_{IC} decreased with increasing percent filler weight.
- In a similar behavior, fracture toughness as a function of filler size range exhibited the highest fracture resistance at a critical value of 4µm.
- However, composites containing nano-fillers showed significantly higher fracture resistance than composites containing only micro-fillers.
- The fracture toughness of composite systems adopted with new polymeric matrix chemistry (i.e., ormocer, dimer acid-base, giomer, and DX-511 monomers) are not statistically different than methacrylate-based systems.

Figure 5: Scanning electron microscopy images of composites tested in this study.



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