Effect of Talc and Synthetic Sodium Aluminum Silicate (SSAS) on the Properties of Poly (Methyl Methacrylate)

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ABSTRACT: The advent of plastics and the wide range of fillers that are available have made modification as precise as the tailored resins themselves. Compounds are prepared from poly (methyl methacrylate) (PMMA), and two fillers, talc and synthetic sodium aluminum silicate (SSAS), at varying concentrations (up to 40 wt%). The effects of both the fillers on the mechanical, thermal, and electrical properties, salt spray performance; rheological as well as optical properties of filled PMMA are measured. The impact strength of PMMA increased with increasing concentrations of talc and SSAS up to 20 and 15% loading, respectively, and thereafter declined continuously in both the cases. Increase in impact strength is more in the case of talc-filled PMMA than that of SSAS-filled PMMA. Tensile strength decreased with increasing concentration of both the fillers continuously. The elongation at break decreased drastically for PMMA with increasing concentrations of both the fillers. Electrical properties remained almost unchanged with respect to the concentration of the fillers. The zero shear viscosity increased with increasing concentration of both the fillers, but the rate of increase was more for SSAS than that for talc.

KEY WORDS: PMMA, talc, SSAS, filled PMMA, mechanical properties, thermal properties, electrical properties, salt spray performance, rheological properties, optical properties.

INTRODUCTION

WITH THE ADVENT of plastics and the wide range of fillers that have become available, modification can be as precise as the tailored resins

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themselves [1]. Fillers are widely used in polymer materials to reduce their cost, improve physical properties, particularly stiffness, hardness, dimensional stability, impact strength, electrical properties, flame retardancy, etc. [2,3]. It is an increasingly widespread practice to incorporate mineral fillers such as $CaCO_3$ into thermoplastics to extend them and to enhance certain properties, notably stiffness and heat distortion temperature (HDT). Often, these benefits are accompanied by a significant reduction in other properties, such as impact strength. Much can be done to offset these limits through the judicious choice of the filler origin, particle size distribution, and surface treatment to promote interfacial interaction between the polymer material and filler [4]. PMMA occupies an intermediate position between commodity and engineering polymers. It has excellent transparency and good tensile strength, and its resistance to weathering and light, makes it an obvious replacement for glass in many applications. PMMA has excellent resistance to inorganic acids and bases, aliphatic hydrocarbons, detergent solutions, cleaners, foodstuffs, and beverages. A unique combination of the electrical properties together with an excellent weather resistance has led to the extensive use of PMMA in outdoor electrical applications. Weather and moisture have little effect on the electrical properties. The surface resistivity is higher than that for most plastics [5].

Talc is the major constituent of rocks known as soapstone or steatite [6]. It is a hydrated magnesium silicate mineral widely used in polymers. The unique features of talc are softness, lubricity, oleophilicity with excellent wetting, and dispersion in plastics and other organics. The dispersability of talc in organic systems has created some commercial applications. Talc is considered as a reinforcing filler because of its platy nature. Its plate-like structure provides talc-filled materials with important properties, such as high resistivity and low gas permeability. Polymers filled with platy talc always exhibit a higher stiffness and creep resistance, at both ambient and elevated temperatures. In plastics, the addition of talc improves their heat distortion temperature, dimensional stability, scratch resistance, and impact resistance [7]. Other important properties include high brightness, blocking of infrared in agricultural film, antiblocking properties, and low abrasion of packaged components. Many other fillers like wollastonite [8] and glass fibers [9] also showed some improvement in the tensile and impact properties in PMMA. Mica [10]- and glass beads [11]-filled PMMA composites showed improvements in primary dispersion temperature, dynamic modulus, compressive strength, and bending modulus. Addition of fumed silica [12] to PMMA led to a decrease of flexural strength and an increase of Young's modulus. Effects of filler composition [13] on the thermal stability, optical properties, and morphology of neat copolymer and a series of hybrid materials, in the form of both coating and freestanding film, are also studied. It is also reported that the transmittance [12] of PMMA composites decreased with increasing filler content. Maspoch et al. [14] reported that surface treatment plays an important role in improving filler matrix interfacial adhesion in PMMA composites. Sodium aluminum silicate is a hydrated silicate containing two metal atoms (sodium and aluminum), in addition to hydroxyl groups. This type of silicate is highly polar in nature. Synthetic sodium aluminum silicate (SSAS) as a filler [15] is reported to enhance the flexural properties, and shear and extensional viscosities of the composites. Synthetic sodium aluminum silicate is being used in paints as a partial replacement for TiO_2 due to its high refractive index and specific particle size and shape, which occupy the interparticle voids in a pigmented film [16]. Synthetic sodium aluminum silicate is also reported to have some pigmentary properties and these are developed in rubber, ink, and [17,18] paper industries. Primary particles are spherical and amorphous while secondary aggregates have regular and fluffy shapes [19] in the structure of SSAS. The effect of any color can be described [20,21] in terms of a color plot, which describes L-, a-, b-values, where, L is the light or dark, a is the greener or redder; a^+ is the redder and a^- is the greener; b is the blue or yellow; b^+ is the yellow and b^- is the blue.

The effect of SSAS on viscous, mechanical, optical, and electrical properties of PMMA has not received any attention. The work reported here was therefore undertaken.

EXPERIMENTAL

Poly (methyl methacrylate) (PMMA, GSFC 876G GRADE with a MFI of 6 and density 1.2 g/cc) from Gujarat State Fertilizers Limited, India was used as the polymer matrix. Commercial talc (UF 520) obtained from Hindustan Mineral Products Co. Ltd., Mumbai and synthetic sodium aluminum silicate, grade Zeolex™ 323 obtained from JM Huber Corporation, USA, were used as fillers. The concentration of both the fillers varied from 0 to 40 wt%. Poly (methyl methacrylate) and both the fillers were predried at 85 ± 5°C for 4–6 h in an air-circulating oven. Poly (methyl methacrylate) with talc and SSAS were compounded with 5, 10, 15, 20, 25, 30, and 40 wt% concentrations of the above fillers using Haake Counter Rotating twin-screw extruder TW 100 (RC-9000, Haake, Germany). The temperature profile during extrusion was: Zone 1 – 200, Zone 2 – 220, Zone 3 – 240, and die temperature – 260°C and 100 revolutions per minute (rpm). The extruded strands were then pelletized and compression molded sheets of 1.55 mm thickness were prepared from the predried granules by using a compression molding machine (M/s Sterling

Hydraulic Co. Ltd., Mumbai) at 200°C and 153 kPa. The granules were allowed to soften and then pressed slowly. Breathing of the sample in the mold was carried out ≈5–6 times over a period of 2 min in order to remove the entrapped air from the mold. After breathing, the material was compressed to the full pressure of 153 kPa. The mold was then cooled slowly to 40 ± 5°C at 153 kPa by circulating water through the platen of the compression molding press. The specimens were cut from these sheets according to the ASTM methods. The tensile specimens were punched out from the compression molded sheets. The dimensional details are as per ASTM D638. The tensile strength and elongation at break of the specimens were evaluated according to ASTM D638 M-91, using a Universal Tensile Tester LR50K (Lloyd Instrument, UK) A crosshead speed of 50 mm/min was maintained for testing. Thin strips cut from the compression molded sheets were used for the determination of impact strength. The dimensional details of the specimens were as per ASTM D256. The notched impact strength was determined on an Avery Denison's pendulum-type, Impact Strength Tester, (model 6709) with a striking velocity of 3.46 m/s. The results reported are the average values of at least 5 specimens. The Vicat softening point was obtained according to ASTM D 1525 at a rate of 120°C/h using an instrument from Davenport (Lloyd Instrument, UK). The electrical properties of filled PMMA were measured as per ASTM D149 using Zaran Instrument, India. The voltage was increased slowly and the voltage at which the current penetrates through the sample was noted. Also, the effect of filler concentration on volume resistivity, and surface resistivity of PMMA was measured using a Zaran Instrument, India.

RESULTS AND DISCUSSION

Mechanical Properties

Tables 1 and 2 summarize the mechanical properties of PMMA filled with talc and SSAS. The results reported are average values from at least

Table 1. Mechanical properties of PMMA filled with talc (The results reported are average values from at least 5 specimens.).

	Composition (wt%, talc)							
Properties	**0**	**5**	**10**	**15**	**20**	**25**	**30**	**40**
Tensile strength (MPa)	51.8	47.0	44.1	43.8	37.4	33.7	20.1	18.6
Elongation at break (%)	7.8	5.9	5.8	5.4	4.4	4.1	3.8	2.4
Impact strength (kg cm/cm)	1.9	3.2	3.3	3.3	3.1	2.3	2.1	1.6

Table 2. Mechanical properties of PMMA filled with SSAS (The results reported are average values from at least 5 specimens.).

	Composition (wt%, SSAS)							
Properties	**0**	**5**	**10**	**15**	**20**	**25**	**30**	**40**
Tensile strength (MPa)	51.8	23.3	15.7	15.4	24.2	26.8	25.1	14.7
Elongation at break (%)	7.8	3.9	1.5	1.5	2.4	3.0	2.3	1.3
Impact strength (kg cm/cm)	1.9	1.8	2.3	2.9	2.2	1.9	1.1	1.1

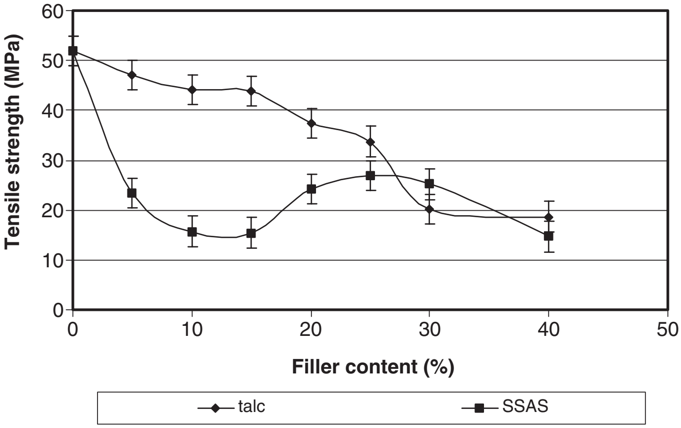

Figure 1. *Variation in tensile strength of PMMA filled with talc and SSAS.*

5 specimens. The tensile strength of PMMA decreased steadily with increase in loading of both fillers. Figure 1 shows the tensile strength of PMMA filled with talc and SSAS. It is clear from the figure that the tensile strength of PMMA due to incorporation of SSAS was seriously affected. The elongation at break decreased drastically for PMMA with increasing concentrations of both the fillers. The maximum load at break for PMMA filled with talc and SSAS decreased continuously with increase in loading. Figure 2 shows the variation in elongation at break of PMMA filled with talc and SSAS. Figure 3 shows the impact strength of PMMA filled with talc and SSAS. The impact strength of PMMA filled with talc is higher than that for PMMA filled with SSAS. It is interesting to note that the impact strength of PMMA filled with both the fillers increased steadily up to 20% loading and then decreased to a significant extent as the percent loading is increased.

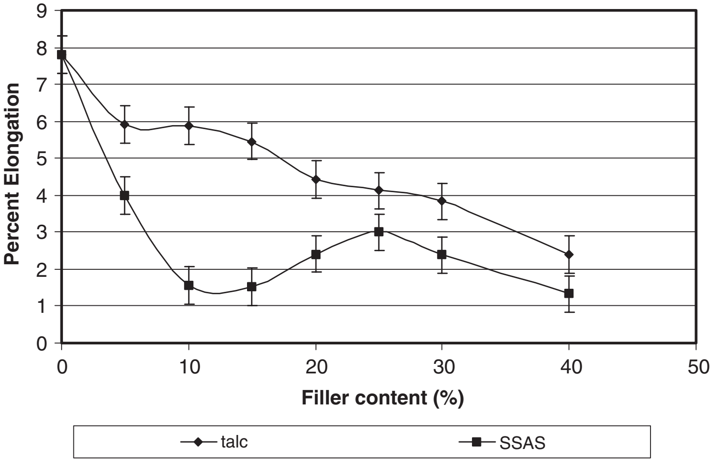

Figure 2. *Variation in % elongation of PMMA filled with talc and SSAS.*

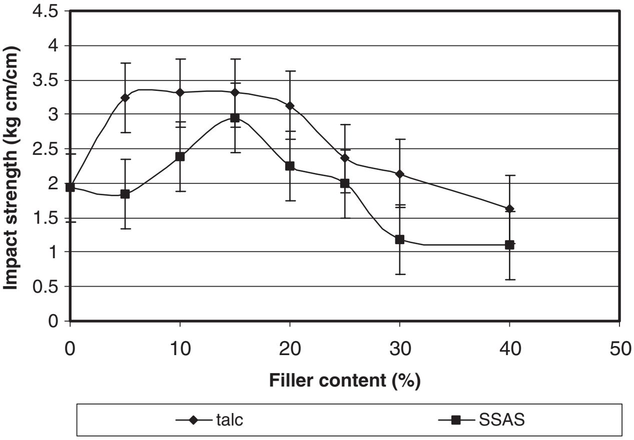

Figure 3. *Variation in impact strength of PMMA filled with talc and SSAS.*

Electrical Properties

Tables 3 and 4 list the electrical properties of PMMA filled with talc and SSAS. The dielectric strength increased up to 25% loading and at a higher loading, decreased significantly. Similarly, surface resistivity reduced marginally till ≈20% loading, while at a higher loading it decreased considerably. Figure 4 shows the dielectric strength of PMMA filled with talc and SSAS.

Table 3. Electrical properties of PMMA filled with talc (The results reported are average values from at least 5 specimens.).

	Composition (wt%, talc)							
Properties	**0**	**5**	**10**	**15**	**20**	**25**	**30**	**40**
Dielectric strength (kV/mm)	15.7	24.3	23.1	21.8	21.2	25	22.5	20.1
Surface resistivity × 10^{-14} (Ω)	1.5	0.8	0.2	1.2	2.5	0.02	0.13	0.1
Volume resistivity × 10^{-14} (Ω)	5	2.5	1	0.7	0.6	0.7	0.7	0.5

Table 4. Electrical properties of PMMA filled with SSAS (The results reported are average values from at least 5 specimens.).

	Composition (wt%, SSAS)							
Properties	**0**	**5**	**10**	**15**	**20**	**25**	**30**	**40**
Dielectric strength (kV/mm)	15.7	19.3	20	18.0	15.4	14.8	18.7	19.3
Surface resistivity × 10^{-14} (Ω)	1.5	0.8	0.6	0.25	0.02	0.02	0.19	0.5
Volume resistivity × 10^{-14} (Ω)	5	1.3	0.6	0.4	0.5	0.25	0.2	0.15

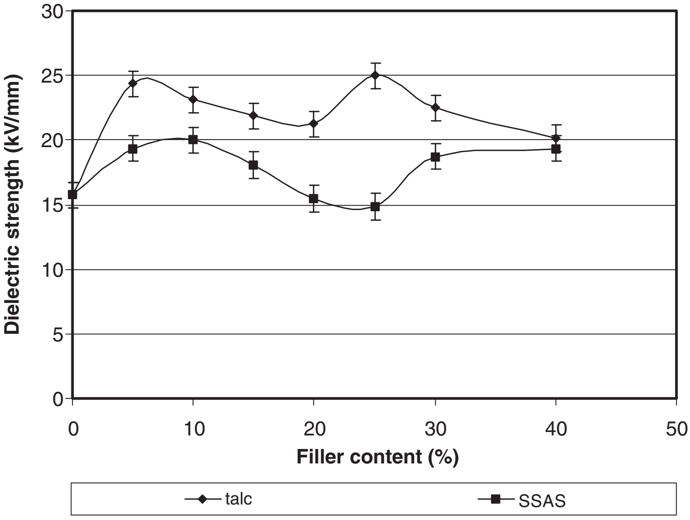

Figure 4. *Variation in dielectric strength of PMMA filled with talc and SSAS.*

Thermal Properties

The Vicat softening temperature (VST) may be taken as the material ultimate use temperature for a short period of time. Table 5 lists the

***Table 5.** Thermal properties (VST) of PMMA filled with talc/SSAS.*

	Composition (wt%, talc/SSAS)							
Properties	**0**	**5**	**10**	**15**	**20**	**25**	**30**	**40**
Vicat softening point °C for PMMA/talc	104	106	107	108	110	111	113	117
Vicat softening point °C for PMMA/SSAS	104	105	106	107	109	110	111	113

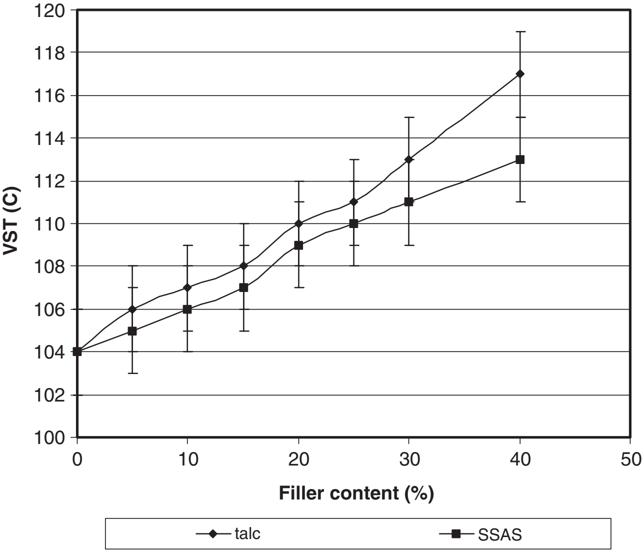

Figure 5. *Variation in VST of PMMA filled with talc and SSAS.*

softening point for PMMA filled with talc and SSAS. Figure 5 reports the dependence of VST with filler content.

Optical Properties

Tables 6 and 7 present the optical properties of PMMA filled with talc and SSAS. The whiteness index decreased and yellowness index (YI) increased at a higher filler loading. These two indices, however improved when the concentration of both fillers was 15%. The L-, a-, and b-values calculated from the spectral characteristics using the Optiview ProPalette 2.0 e® software for all the compounds are listed in Tables 6 and 7. $L = 100$, indicates the whitest appearance, and $L = 0$ indicates the darkest appearance. The positive and negative values of a indicate red and green shades, respectively, while the positive and negative values of b indicate yellow

Table 6. Optical properties of PMMA filled with talc.

	Composition (wt%, talc)							
Properties	**0**	**5**	**10**	**15**	**20**	**25**	**30**	**40**
Yellowness index	−0.60	−4.47	2.89	2.27	7.58	7.63	9.54	12.93
Whiteness index	11.11	28.48	16.87	24.17	11.68	14.13	1.83	5.07
L	31.24	41.28	51.27	56.48	59.39	61.45	63.57	67.12
a	0.024	−0.67	−0.80	−0.85	−1.01	−0.83	−0.99	−0.84
b	−0.098	−1.11	1.09	0.99	3.13	3.24	4.17	5.89

Table 7. Optical properties of PMMA filled with SSAS.

	Composition (wt%, SSAS)							
Properties	**0**	**5**	**10**	**15**	**20**	**25**	**30**	**40**
Yellowness index	−0.60	0.01	−4.64	−10.5	−5.10	−8.46	−3.36	9.95
Whiteness index	11.11	15.64	24.55	44.88	26.92	43.82	23.68	−15.9
L	31.24	40.44	35.22	41.61	37.01	46.62	39.52	38.32
a	0.024	−0.27	−0.37	−0.88	−0.56	−1.12	−0.93	−0.56
b	−0.098	0.065	−1.02	−2.74	−1.14	−2.44	−0.74	2.60

and blue shades, respectively. The *L*-values increased with increase in filler concentration in both the fillers. The negative values of *a* shows that the composite has turned toward greener shades.

Rheological Properties

Figures 6 and 7 show the variation in shear viscosity (η) with shear rate (γ) of virgin PMMA and PMMA filled with talc and SSAS. The figures show the gradual increase in zero shear viscosity (η_0) with increasing concentration of talc as well as SSAS in PMMA. The ratio of $G^{//}$ (loss modulus)/$G^{/}$ (elastic modulus) i.e., $\tan\delta$ is a measure of viscoelasticity. As the filler content increased, the $\tan\delta$ value decreased for both the cases as shown in Figures 8 and 9.

Resistance to Salt Spray and Accelerated Weathering Properties

Tables 8 lists the changes in optical properties after exposure to the accelerated weathering for 250 and 500 h. The yellowness index increases with exposure for unfilled as well as filled PMMA but the presence of the filler reduces the rate of yellowing in both the cases.

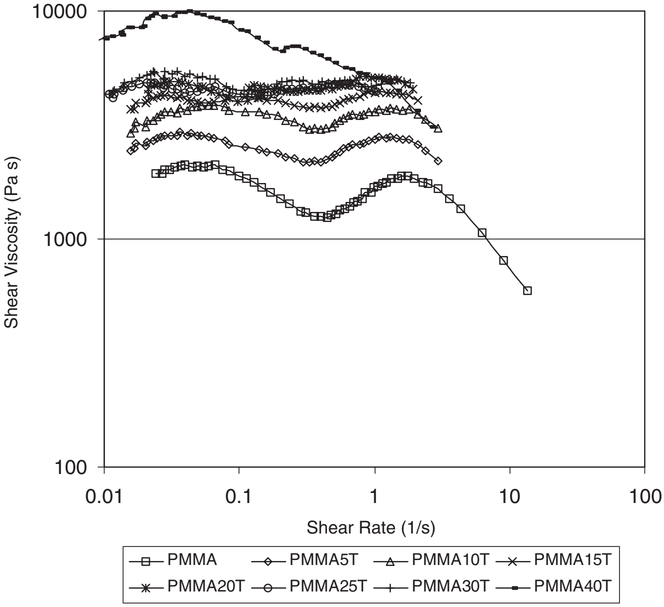

Figure 6. *Flow behavior of PMMA filled with talc at 250°C.*

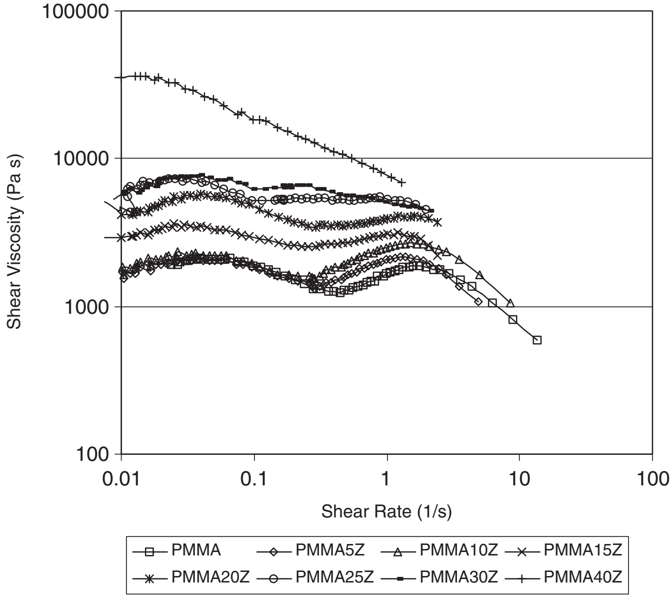

Figure 7. *Flow behavior of PMMA filled with SSAS at 250°C.*

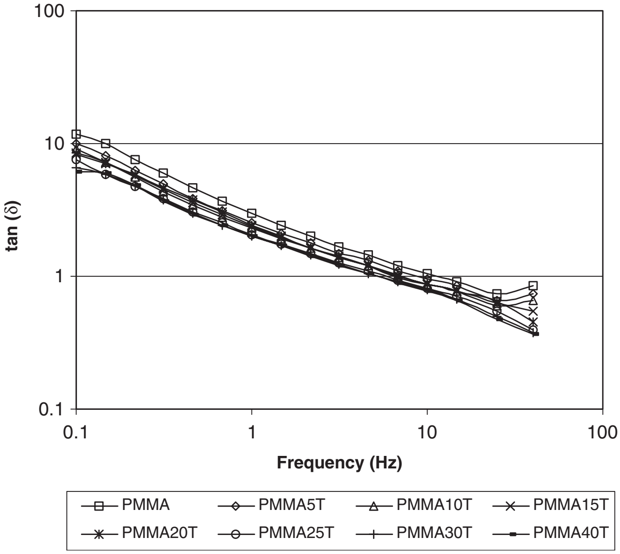

Figure 8. *Variation in tan δ for PMMA filled with talc at 250°C.*

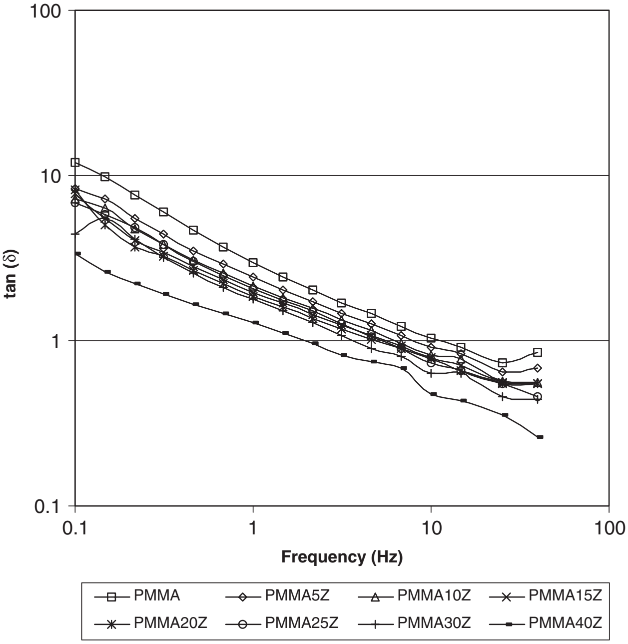

Figure 9. *Variation in tan δ for PMMA filled with SSAS at 250°C.*

Table 8. Optical properties of PMMA filled with talc/SSAS after 250 and 500 h exposure to accelerated weathering.

Filler concentration (%)	Talc				SSAS			
	Yellowness		Whiteness index		Yellowness index		Whiteness index	
	250 h	500 h	250 h	500 h	250 h	500 h	250 h	500 h
0	2.58	−0.68	4.36	11.70	2.58	−0.68	4.36	11.70
5	2.91	−4.34	26.77	25.5	−3.10	−1.53	29.27	24.2
10	1.81	2.50	19.76	18.5	−5.57	−5.67	24.07	24.7
15	2.17	0.98	25.21	31.3	−4.89	−4.10	27.04	25.1
20	6.64	6.60	16.27	17.2	−3.46	−3.25	19.66	19.2
25	6.85	7.32	19.19	17.11	−6.97	−7.30	29.13	33.7
30	10.32	10.11	12.11	12.23	−5.30	−5.49	23.16	23.3
40	15.06	13.14	5.55	10.53	4.77	8.30	2.99	10.1

CONCLUSIONS

Impact strength of PMMA is increased with increasing concentrations of talc and SSAS up to 20% loading and thereafter declined continuously. Increase in impact strength is more in the case of talc filled PMMA than that of SSAS filled PMMA. Tensile strength was decreased with increasing concentration of both the fillers. The % elongation at break decreased drastically for PMMA with increasing concentrations of both the fillers. Electrical properties remained almost unchanged with respect to concentration of fillers. At lower concentrations, the fillers induced dull appearance, which disappeared as the concentration increased.

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