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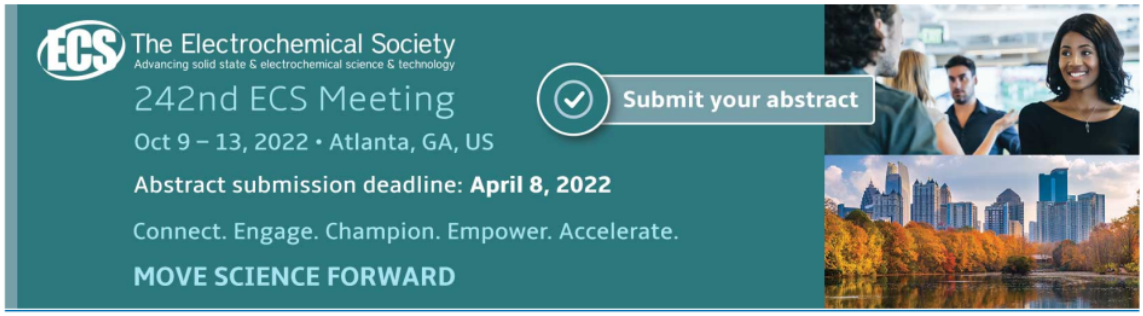

# Thermomechanical properties of PEG-based composites with micro-quartz fillers

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**Abstract**. Micro-quartz-reinforced - polyethylene glycol (PEG) composites were fabricated by a simple liquid method. This study reveals the thermomechanical properties of the PEG/quartz composites around its melting transition temperature. The quartz microparticles were prepared from natural silica sand collected from Tanah Laut Pelaihari, South Kalimantan, by magnetic separation, HCl immersion, and water cleaning. Then, quartz powders were heated in various temperatures, i.e., at 500, 1000, and 1200 °C where we found quartz particle sizes of 168, 217, and 249 nm, respectively. PEG/quartz composites with 10 wt% of these sizes were synthesized and a pure PEG sample was also prepared for comparison. The Fourier-transform infra-red spectroscopy (FTIR) and x-ray diffraction (XRD) data showed that there was no additional peak in composite patterns which verified the successful synthesis of the PEG/micro-quartz composites. Meanwhile, the dynamic mechanical analysis (DMA) data and analysis revealed that the maximum storage moduli ($G'$) and melting transition temperature ($T_m$) exhibited by the composite with quartz heated at 500 °C, i.e., 610.78 MPa and 53.4 °C, respectively. This value is almost six times of that of pure PEG. In general, quartz particle size shifted the G'and $T_m$ to the higher values.

## 1. Introduction

Nowadays, we often find many polymer-based materials because of their outstanding properties such as ease of manufacture, lightweight, low cost, and facility to control [1]. Due to these attractive properties, polymers have widely replaced several metal-based materials. For example, in recent decades, several industries have used polymer-based materials in some of their products, including in packaging, automotive, household goods, medicine, and aircraft industries [2]. One of such functional polymers is polyethylene glycol (PEG). Its outstanding properties, e.g., non-toxic and ease of fabrication [3], however, come with low resistances of temperature and external forces. One way to eliminate this weakness is by combining it with stronger ceramic powders to form filler-dispersed composites [4].

Ceramic powders can be developed as a filler for polymers due to its strength and resistance to high temperatures. Literature shows that silica [5,6], zircon [7,8], and zirconia [9] have been used as fillers of various polymers. According to Ramirez et al. [10], in an isotropic filler, the physical properties of composites can be affected by three of the most important characteristics of fillers, i.e., the particle size, the polymer-filler bonding capability, and the particle shape. In general, the mechanism is the smaller filler, the larger surface area of the filler-matrix interactions. Thus, the interaction between filler and matrix is very crucial.

In our previous report, PEG/quartz silica composites had been studied [5] but not the effect of the filler size. It was found that quartz filler size of 162 nm gave DMA shear storage modulus (*G'*) of about twelve times of the pure PEG. Here, we describe the influence of the quartz size by enlarging that from previous work [5] through heating prior to the synthesis of the composites.

## 2. Methods

### *2.1. Micro-quartz powders preparation*

Natural silica sand from Tanah Laut, South Kalimantan, Indonesia was used as the source of the quartz powders. The purification of the sand had been described previously [5]. It included four main steps, i.e., magnetic separation, immersion in HCl (2 M for an hour), washing with distilled water to neutralize the pH, and drying at 100 °C to acquire the initial quartz powder. The powder was pure quartz, with a size of ~160 nm. Then, the quartz powder was heated at 500, 1000, and 1200 °C for 3 hours to obtain the micro-quartz powders. They were denoted as Q500, Q1000, and Q1200, respectively.

### *2.2. Composites preparation and characterization*

The procedure for the composites preparation was also reported in the previous work [5], i.e., by a liquid mixing method. We used polyethylene glycol (PEG 4000) from Merck, Germany. As much as 10 wt.% of micro-quartz powder from each size was added into a melted PEG matrix. Then, the samples were prepared in uniform geometry, i.e., 5 mm length × 5 mm width and 1 mm thickness. The composite with a certain filler is denoted similarly to the filler. We ensured the success of composites formation using XRD (Philips X'Pert MPD diffractometer) and FTIR (Bruker Vertex7.0v). Then, DMA (SDTA861e Mettler Toledo) measurement in shear mode was carried out to determine the thermomechanical properties of the composites. The DMA characteristics of the composites were evaluated at a temperature range of 25 ºC up to 80 ºC, a heating rate of 5 °C $min^{-1}$, and a frequency of 1 Hz.

## 3. Results and Discussion

The XRD patterns and the analysis results of the micro-quartz powders are presented in Figure 1 and Table 1, respectively. Q500 powder exhibits pure quartz (PDF No. 16-2490) at Q500, but Q1000 and Q1200 powders show a secondary phase of cristobalite (PDF No. 39-1425) as indicated by a peak at approximately 21° 2θ. A higher cristobalite peak is found in the Q1200 data than in the Q1000 one. It appears, the higher the calcination temperature, the more the cristobalite. To quantify the phase composition, we further analyze the XRD data using Rietica software and the results are presented in Table 1. This is a transformation from quartz to cristobalite in the samples heated at 1000 °C and 1200 °C by an amount of 0.53 and 10.47 wt.%, respectively. These results are in line with other studies which state that the temperature of cristobalite formation is about 950 °C [5,11,12]. Furthermore, Table 1 reveals the increase in the estimated quartz crystallite size from 162 nm (for unheated quartz [5]) to 168, 217, and 249 nm for Q500, Q1000, and Q1200, respectively. The size values indicate sub-micron range according to XRD data but could be much higher when a direct size measurement is applied. We state here that the synthesis of micro-quartz powders which will be used as fillers in the PEG-based composites is successful.

Figures 1 and 2 show the XRD patterns and FTIR spectra of PEG, micro-quartz powders, and PEG/micro-quartz composites. In both data, the micro-quartz and PEG peaks are separated from each other indicating that there is no reaction between the materials, hence confirming that the composites have been successfully synthesized. A detailed analysis of the FTIR spectra (Figure 2) shows a broad band around 3750-2500 $cm^{-1}$ (denoted as A) which can be assigned to a *O-H stretching* [5] in PEG molecules. Meanwhile, the broad transmission band around 1130-1000 $cm^{-1}$ (B) is likely due to the *Si-O-Si asymstreching* [13], the broad band around 1500-800 $cm^{-1}$, reveals *C-O-C stretching* [5].

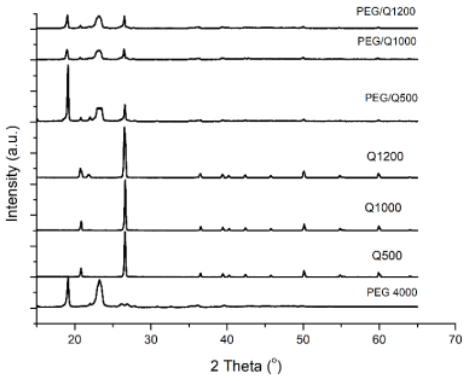


**Figure 1.** XRD patterns (Cu-Kα radiation) of pure PEG, quartz powders and PEG/micro-quartz composites 10 wt.%.

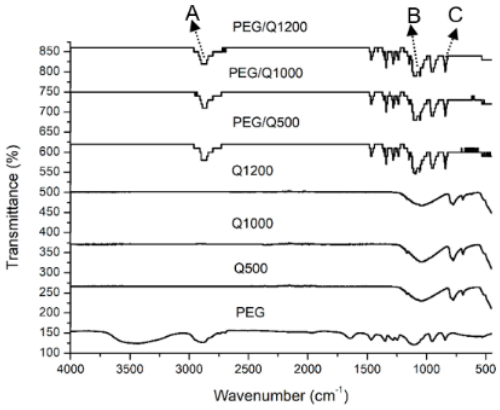


**Figure 2.** FTIR spectra of pure PEG, quartz powders and PEG/micro-quartz composites 10 wt.%.

**Table 1.** Output of the quantitative analysis of the XRD data of quartz micropowders using the Rietveld-based *Rietica* and *MAUD* softwares.

| Sample | Phase composition (wt.%) | | Size (nm) |
|---|---|---|---|
| | Quartz | Cristobalite | |
| **Q500** | 100 | - | 168(8) |
| **Q1000** | 99.47(1) | 0.53(2) | 217(4) |
| **Q1200** | 89.53(2) | 10.47(3) | 249(7) |

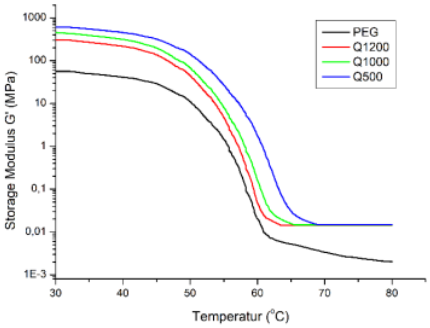


**Figure 3.** DMA shear storage moduli of PEG/micro-quartz composites (10 wt.% filler).

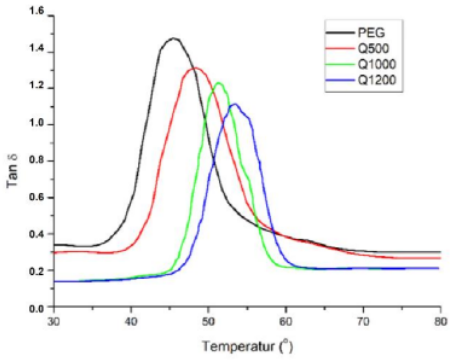


**Figure 4.** DMA tan δ (shear mode moduli) of PEG/micro-quartz composites (10 wt.% filler).

**Table 2.** The shear storage moduli and melting temperature ($T_m$) of PEG/micro-quartz composites.

| Quartz concentration (wt.%) | G' (MPa) | | | $T_m$ (°C) | | |
|---|---|---|---|---|---|---|
| | Q500 | Q1000 | Q1200 | Q500 | Q1000 | Q1200 |
| **0** | 93.00 | 93.00 | 93.00 | 42.0 | 42.0 | 42.0 |
| **10** | 610.78 | 454.13 | 308.28 | 53.4 | 51.3 | 48.3 |

Figures 3 and 4 present the shear storage moduli (*G'*) and tan δ plots as a function of temperature from the DMA measurement for the pure PEG and the composites. The pure PEG exhibits the lowest shear storage moduli (*G'*) among all samples. Their room temperature *G'* values are presented in Table 2. It can be seen that the Q500 composite shows the optimum modulus, i.e., six times of the pure PEG, and followed by the Q1000 and Q1200 composites. It appears pure quartz powder and smaller quartz size simultaneously gives higher *G'*. As discussed previously, Q1000 and Q1200 powders contain cristobalite as a secondary phase. Lower *G'* values for the Q1000 and Q1200 composites can be associated with the lower theoretical shear modulus of cristobalite than quartz (i.e., 39.1 as compared to 41.1 GPa [14]). However, this difference is much smaller than the associated *G'* values. We argue here that the effect of quartz size is more pronounced. A smaller filler causes a greater area of contact between the matrix and the filler, and hence stronger bonding between them. This finding is in line with the previous report for PMMA/zircon composites [7]. Furthermore, in terms of cristobalite as a filler, it tends to form a coral shape morphology which causes more heterogeneous distribution of external forces [12].

The existence of hard and rigid quartz will inhibit the free movement of the polymer chains in the transition temperature of PEG (here the melting temperature, $T_m$). This temperature changes to the higher values as larger quartz was added, i.e., from 42 (pure PEG) to 48.3, 51.3, and 53.4 °C for Q1200, Q1000, and Q500 composites, respectively. The presence of quartz in the composite limits PEG chain movement [15]. Furthermore, the transition temperature of a material depends on its latent heat (the ability to store energy during temperature transitions). Literature states that quartz silica has a higher latent heat than

pure PEG (238 $Jg^{-1}$ [16] vs. 209 $Jg^{-1}$ [17]). As a result, the presence of quartz filler increases the absorbed thermal energy when heating is applied.

## 4. Conclusion

This work revealed how quartz filler size could affect the storage moduli *G'* and transition temperature $T_m$ of PEG/micro-quartz composites. The smaller the filler size, the higher the *G'* and $T_m$ values. In addition, the presence of a secondary phase (cristobalite phase) can reduce those values due to its macroscopic modulus which is lower than that of quartz and its coral shape morphology which causes a relatively heterogeneous force distribution.

## Acknowledgement

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