

Laboratory scale study on rheological behavior, morphological and structural properties of crosslinked polyacrylamide composite hydrogels embedded with date seed powder

Ahmad. A. Adewunmi,^{1,2} Suzylawati Ismail,¹ Abdullah. S. Sultan²

¹School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Seberang Prai Selatan Pulau Penang, Malaysia

²Center for Petroleum and Minerals, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia

Correspondence to: A. A. Adewunmi (E-mail: ahmadadewunmi@yahoo.com) and S. Ismail (E-mail: chsuzy@usm.my)

ABSTRACT: Incorporation of biofillers in polymeric hydrogels has continued to receive great attention in recent times because of their excellent properties. In this study, polyacrylamide (PAM) and polyethyleneimine (PEI) were used to develop novel composite hydrogels filled with date seed powder (DSP) via chemical crosslinking technique. Pristine PAM/PEI hydrogel and PAM/PEI-DSP hydrogels at various DSP loadings were fabricated and subjected to gelation at 40°C for 24 h. The impact of various DSP loadings on the hydrogel samples developed was investigated using hybrid rheometer, SEM, XRD, and FTIR instruments, respectively. Rheological measurements confirmed the viscoelastic responses of the neat PAM/PEI hydrogel and the PAM/PEI-DSP hydrogels reinforced with various DSP contents (0.8, 2.4, and 4 wt %). The dynamic strain, dynamic frequency and time sweep tests demonstrated that PAM/PEI-DSP hydrogels were slightly more elastic than the virgin PAM/PEI hydrogel. The SEM characterization revealed the surface micrographs of the neat PAM/PEI hydrogel and the PAM/PEI-DSP hydrogels at different DSP loadings to be smooth, homogeneous, and dense. Besides, the SEM micrographs supported the incorporation of DSP in the PAM/PEI-DSP hydrogel samples. XRD analysis showed that the structures of neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels filled with various DSP contents were predominantly amorphous while FTIR results confirmed the functional groups and evidence of crosslinking in the neat PAM/PEI hydrogel and the PAM/PEI-DSP hydrogels embedded with different DSP contents. It is believed that these new hydrogels have huge development potentials and promising future in wastewater treatment and removal of heavy metal ions in aqueous solutions. © 2015 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 2015, 132, 42110.

KEYWORDS: composites; crosslinking; gels; morphology; rheology

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INTRODUCTION

The last decades have witnessed the swift development and investigation of polymeric composite hydrogels, because of their distinct mechanical, optical, and electronic properties at a low loading of nanoscale fillers, as compared to their counterparts.^{1–7} Inspired by the excellent properties of fillers in polymeric hydrogels, the incorporation of macro and nanoscale fillers into a polymer matrix is of great importance for researchers from industry and academia.¹

Hydrogels are known to be a class of polymers that can absorb and swell several times their dry volume. In recent times, composite hydrogels have continued to gain a great deal of attention because of characteristic properties such as swell ability in water, biocompatibility, and hydrophobicity. Among the known conventional hydrogels, polyacrylamide

(PAM) based hydrogels are quite important due to their utilization in many sorbent applications.⁸ However, because of the soft and brittle nature of PAM based hydrogels, studies have been focused on redesigning novel composite hydrogels embedded with different kinds of fillers in order to enhance their properties. Several types of fillers have been introduced in composite hydrogels.^{9–11} In fact, a comprehensive literature survey revealed that clay, carbon nanotubes and silicates are the leading reinforcing fillers in composite hydrogels. Kamoun and Menzel¹² reported in their study, the swelling, mechanical and thermal properties of composite hydrogels filled with organophilic sodium montmorillonite (Na-MMT) and Laponite-RD clays. Yavari-Gohar *et al.*¹³ studied the effect comonomer acrylamide and an organomodified clay on swelling stability of *N,N'*-methylenebisacrylamide (MBA)-cross-linked polymethylpropane sulfonic acid/acrylamide (AMPS-

AM). Moreover, many other macro/nano fillers incorporated in polymeric hydrogels have been reported in literatures.^{14–18}

Date seeds form part of a popular subsistence among the populace of the Arab countries. The fruit of date palm is well known as a staple food. It is made up of a fleshy pericarp and seed. Researchers have carried out some laboratory studies on the potential applications of date seeds (also called date pits). Besbes *et al.*¹⁹ investigated in their study, the chemical composition of date seed. According to the outcome of their results, carbohydrate constitutes the major compound in date seeds while protein, fat, ash and minerals are present in small quantities. Habib *et al.*²⁰ reported in their study, the amount of carotenoids, fat soluble vitamins and fatty acid present in different species of date seeds. Their findings showed significant variations in the amount of carotenoids, fat soluble vitamins and fatty acid obtained, among the varieties of date seeds used. Nehdi *et al.*²¹ also conducted related study on the chemical composition of fully ripened date palm seed. In addition, date seeds have been tested in the laboratory as pragmatic natural substances for the removal of toxic transition metal ions in aqueous solutions.²² Likewise, polyol was successfully synthesized through the modification of date pits.²³ Adewole and Sultan²⁴ also investigated the processing and chemical composition of date seed powder (DSP) for possible applications in enhanced oil recovery (EOR).

However, extensive literature survey showed that date seeds have not been fully utilized or modified to serve various applications. It is not uncommon to find date seeds as environmental waste materials, especially, in the Middle East regions and the Northern part of Africa. Hence, there is pragmatic need to process date seeds into value-added products. Date seed powder (DSP) derived from date seeds milling is a promising biofiller that could be introduced in crosslinked polymeric composite hydrogels with significant impact on the hydrogels rheological, morphological and structural properties. To the best of our knowledge, no previous work has reported the utilization of DSP as biofiller in polymeric composite hydrogels. In the current study, various hydrogel samples were developed using PAM crosslinked by polyethyleneimine (PEI) in the presence of DSP dispersed in distilled water. Neat PAM/PEI hydrogel and PAM/PEI-DSP composite hydrogels containing various DSP contents were formed via chemical crosslinking technique. The impact of DSP incorporation on the fabricated hydrogel samples was examined using rheological approach. Also, morphological and structural characterizations of the hydrogel samples developed were explained in detail.

EXPERIMENTAL

Materials

PAM and PEI solutions used in this study were supplied by the SNF Chemical, France. The properties of PAM and PEI solutions are shown in Table I. The date seeds used to process DSP were obtained from the Eastern region in the Kingdom of Saudi Arabia. The seeds were soaked in water for 1 day in order to remove their surface layers. Then, the seeds were washed several times with distilled water followed by drying and milling.

Table I. Properties of PAM and PEI Solutions

Variable	PAM	PEI
Molecular weight, (g/mol)	250,000 - 500,000	70,000
Density ^a , (g/cm ³)	1.09	1.07
pH ^a	4	12

^a Measured at room temperature (25°C).

Preparation of Neat PAM/PEI Hydrogel and PAM/PEI-DSP Hydrogels

The PAM/PEI-DSP composite hydrogels were prepared via the crosslinking of PAM with PEI as the crosslinker in distilled water dispersed with DSP. Table II lists hydrogels formulation containing various DSP quantities (i.e., 0.8 wt % (0.1 g), 2.4 wt % (0.3 g), and 4 wt % (0.5 g)), as well as the constant volumes of PAM, PEI and distilled water. The weight fraction of DSP added was calculated based on the water used (12 mL water = 12 g water). The PAM/PEI-DSP composite hydrogels were developed at room temperature by adding a measured quantity (in grams) of DSP into a 20-mL beaker containing 12 mL distilled water followed by mixing using magnetic stirrer for 1 min. Then, PAM and PEI were added in drops using separate syringe (PEI solution was added last as the crosslinker) and stirring was maintained for another 10 min. 6 mL PAM and 0.8 mL PEI were used throughout the synthesis of each hydrogel sample. Also, neat PAM/PEI hydrogel (without DSP) was developed. Afterwards, mixture of each composite hydrogel sample was transferred into separate glass tubes (outer diameter: 50 mm and height: 15 mm) and incubated in an oven at 40°C for 24 h. After the gelation was fully completed, hydrogels for the rheological tests were sliced to obtain samples of 30 mm in diameter and 10 mm in height. The remaining hydrogel samples were dried at 50°C in a vacuum oven to a constant weight for morphological and structural characterizations.

Characterizations

The particle size distributions of DSP were determined by Turbotrac S360 particle size analyzer (PSA). The oven-dried hydrogel samples were coated with gold to enhance conductivity and their morphologies were observed by JEOL scanning electron microscopy (SEM) using 20 kV voltage. Wide-angle XRD patterns of the dried hydrogels were measured using an X-ray equipment. (Ultima IV Model, manufactured by Rigaku, Japan). The tests were performed with scan speed of 2.5°/min within the range of 10–80°. The functional groups of hydrogel samples were characterized by Fourier transform infrared spectroscopy (FT-IR, Nicolet 6700) working in attenuated total reflectance mode.

Rheological Properties of Neat PAM/PEI Hydrogel and PAM/PEI-DSP Hydrogels

All rheological measurements were performed with a parallel plate and peltier plate geometry using an oscillatory testing mode at 30°C. To avoid evaporation during measurements, the gap between the two plates was covered with the solvent trap system. The dynamic strain sweep from 0.1 to 100% was

Table II. Composition of Neat PAM/PEI Hydrogel and PAM/PEI-DSP Hydrogels

Hydrogel sample	DSP (g)	PAM (mL)	Distilled water (mL)	PEI (mL)
PAM/PEI (without DSP)	–	6	12	0.8
PAM/PEI-0.8 wt % DSP	0.1	6	12	0.8
PAM/PEI-2.4 wt % DSP	0.3	6	12	0.8
PAM/PEI-4 wt % DSP	0.5	6	12	0.8

performed on the neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels filled with different DSP contents at 30°C and their respective storage modulus (G') was recorded in order to define linear viscoelastic region (LVR) in which the G' is independent of the strain amplitude. A strain of 1% was chosen in the subsequent oscillation tests so that the dynamic oscillatory deformation of each sample was within the LVR.

Also, the maturation gelation kinetics of the composite hydrogels developed were monitored through the evolution of storage and loss moduli with the reaction time at temperature of 30°C, strain of 1%, and frequency of 1 rad/s. The viscoelastic param-

eters, including storage modulus (G') and loss modulus G'' as a function of angular frequency (ω) were measured over the ω range of 0.1–100 rad/s at strain of 1% and temperature of 30°C. The complex modulus (G^*), complex viscosity (η^*), and loss tangent ($\tan \delta$) were estimated using eqs. (1–3), respectively:

$$G^* = \sqrt{G'^2 + G''^2} \quad (1)$$

$$\eta^* = \frac{G''}{\omega} \quad (2)$$

$$\tan \delta = \frac{G''}{G'} \quad (3)$$

RESULTS AND DISCUSSION

Reaction Mechanism Governing Hydrogel Formation

The DSP used in the current study is shown in Figure 1(a) and its particle size distributions are depicted in Figure 1(b). Majority of the DSP particle size fall within the range of 10–30 μm . The reaction mechanism governing the hydrogel (gel) formation between PAM and PEI embedded with DSP occurred through the substitution reaction where the nucleophilic amine nitrogen on PEI substitutes the acrylamide pendant groups on the PAM, resulting into a covalent bond between the nitrogen on PEI and the PAM amide group. Research studies have shown that variety of polymers containing acrylamide pendant groups react with nitrogen on PEI via a transamidation reaction pathway to produce gels.^{25,26} The schematic of preparation process of hydrogels developed is represented by Figure 2. Typical pristine PAM/PEI hydrogel and PAM/PEI-DSP hydrogel containing DSP contents are shown in Figure 3.

LVR of Neat PAM/PEI Hydrogel and PAM/PEI-DSP Hydrogels

Rheological property of polymeric composite hydrogel is very significant for fundamental and technology investigation, since it enables us understand the microstructure and dispersion of fillers in matrix.²⁷ Many factors can actually influence the rheological properties of composite hydrogels, for instance, the orientation, content, shape, dispersion of filler in matrix and the interfacial interaction between filler and composite hydrogel. Figure 4 shows the G' as a function of strain γ for the pristine PAM/PEI hydrogel and the PAM/PEI-DSP hydrogels filled with DSP ranging from 0.8 to 4 wt %. During the dynamic oscillation testing, the cyclic levels of γ (from 0.1 to 100%) were applied at $\omega = 1.0$ rad/s. In the region of LVR, the G' values of neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels containing various DSP quantities were completely independent of the applied strain, behaving like a viscoelastic solid. It can be observed that the pristine PAM/PEI hydrogel and PAM/PEI-DSP composite hydrogels containing various DSP loadings

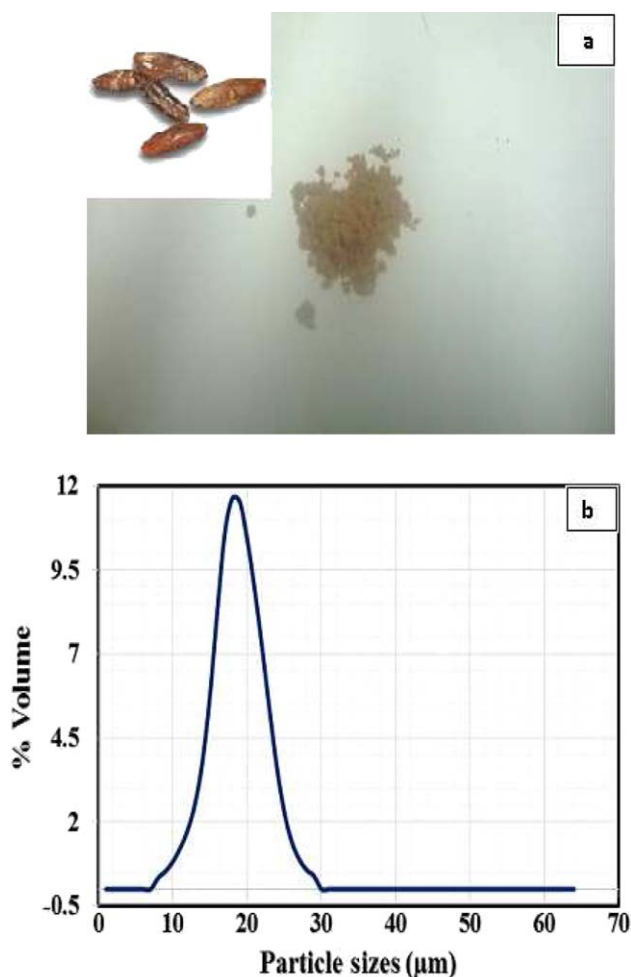


Figure 1. (a) DSP used in this study (b) The particle size distributions of DSP. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

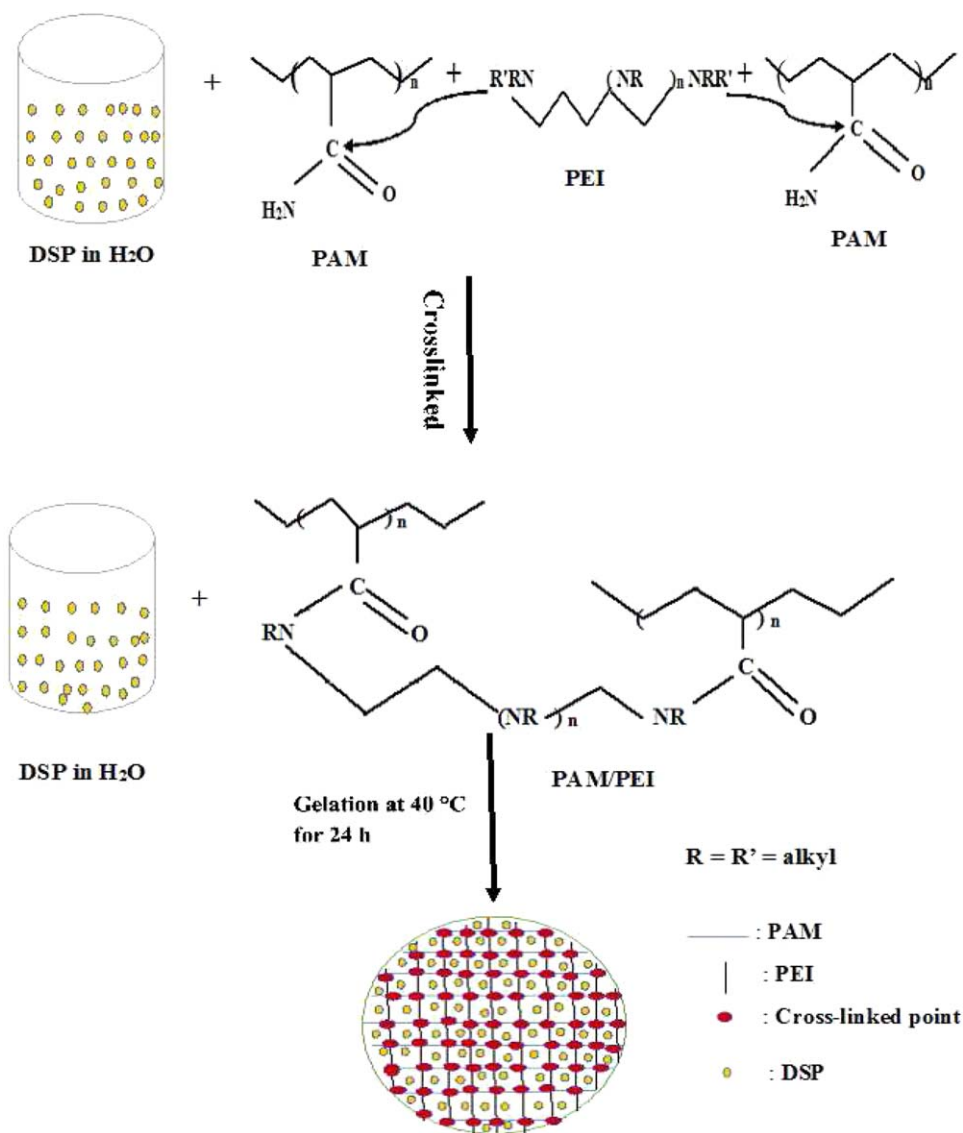


Figure 2. Schematic of PAM/PEI-DSP hydrogel preparation. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]



Figure 3. Neat PAM/PEI hydrogel (left); PAM/PEI-DSP hydrogel containing DSP (right). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

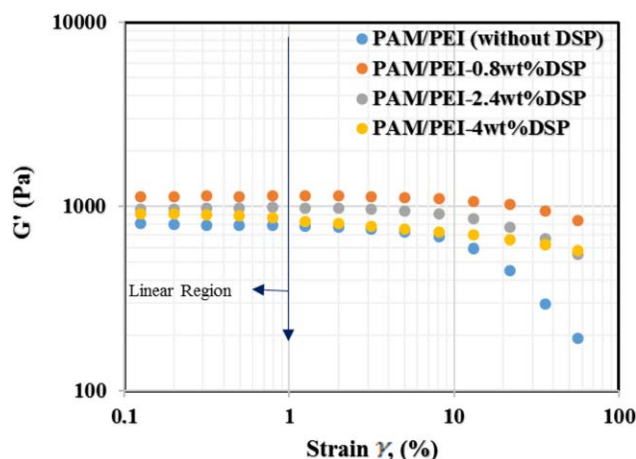


Figure 4. Storage modulus G' as a function of strain γ for the neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels at different DSP contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

displayed a linear viscoelastic behavior up to a strain amplitude of about 1%. Further observation from this plot (Figure 4) revealed the regions at which G' value of each hydrogel sample deviated by more than 5% from the plateau regions. These regions signified departure from the linear viscoelastic character and therefore were defined as the critical strains (γ_c) of each sample.^{28,29} Then, G' values of the neat PAM/PEI hydrogel and the PAM/PEI-DSP composite hydrogels containing various DSP loadings gradually decrease beyond the γ_c indicating a transition from the quasi-solid state to a quasi-liquid state. Consequently, reinforced PAM/PEI-DSP hydrogels embedded with different DSP contents demonstrated higher G' than the virgin PAM/PEI hydrogel within LVR, with PAM/PEI-0.8 wt % DSP hydrogel exhibiting the highest G' (1134 Pa), indicating that this hydrogel sample is the most rigid.

Dynamic Frequency Sweep of Neat PAM/PEI Hydrogel and PAM/PEI-DSP Hydrogels

To further understand the impact of various DSP loadings on the viscoelastic properties of the synthesized hydrogel samples, oscillatory measurements were performed at 30°C on the neat PAM/PEI hydrogel and the PAM/PEI-DSP composite hydrogels filled with various DSP amounts. Figure 5 depicts the storage modulus G' and loss modulus G'' as a function of frequency (ω) between 0.1 and 100 rad/s for all the hydrogel samples. As can be seen from this graph, the DSP-free hydrogel (i.e., neat PAM/PEI hydrogel) and the PAM/PEI-DSP composite hydrogels filled with different DSP contents exhibited very high G' than G'' throughout the whole frequency range, indicating solid-like behavior. This observation is in agreement with previous studies on composite hydrogels.^{4,30,31} Besides, the insensitivity of G' to frequency demonstrated by the neat PAM/PEI hydrogel and the PAM/PEI-DSP composite hydrogels embedded with various DSP amounts is also an indication of solid-like character. However, PAM/PEI-0.8 wt % DSP hydrogel showed marginal increment in G' above other hydrogel samples, signifying that it is the stiffest. Moreover, Figure 6 displays the $\tan \delta$ as a function ω which corroborates the

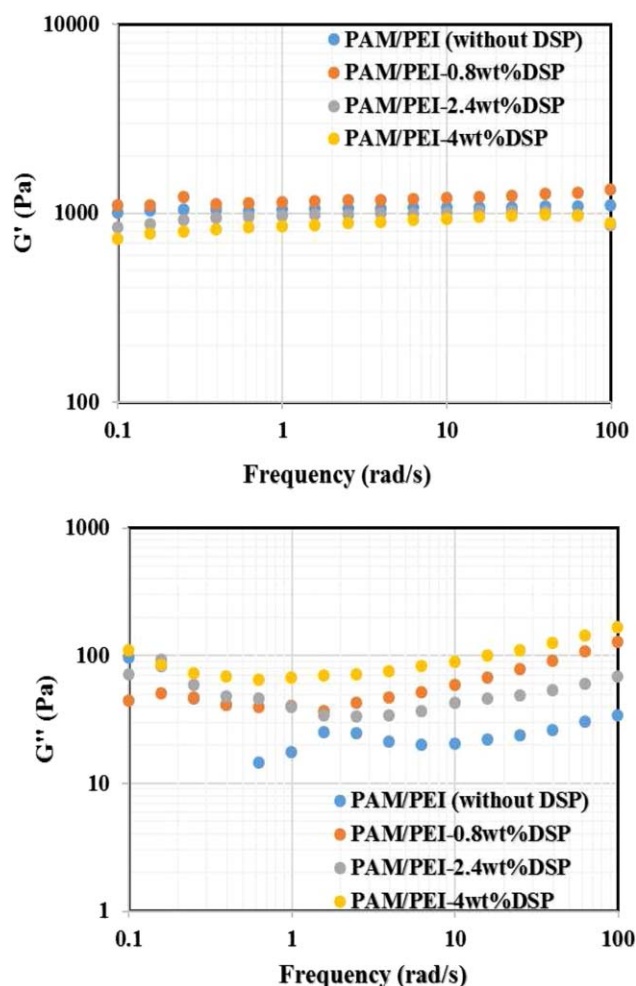


Figure 5. Storage and loss moduli (G' and G'') as a function of frequency for the neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels at different DSP contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

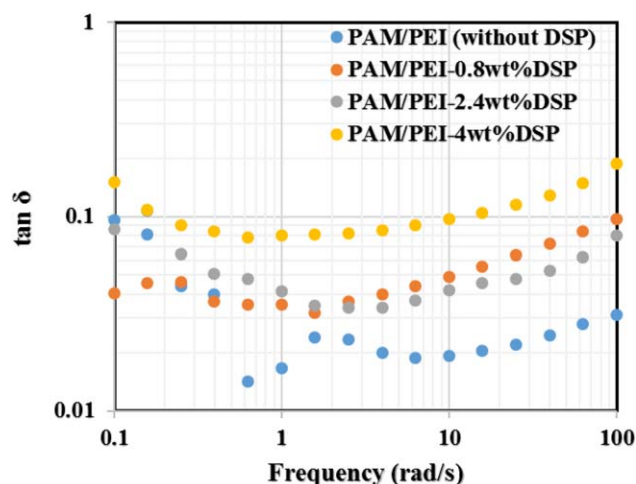


Figure 6. Frequency dependence of $\tan \delta$ for the neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels at different DSP contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

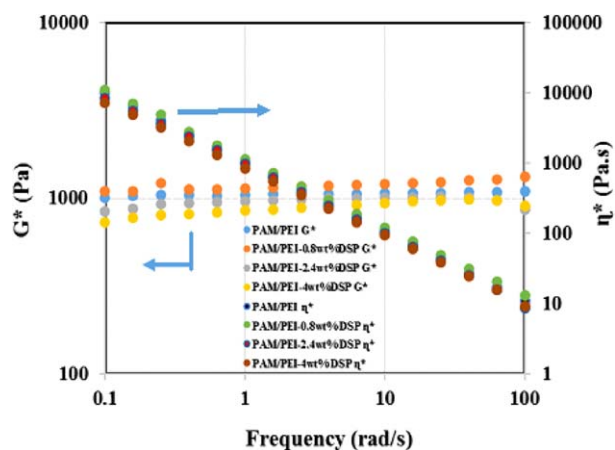


Figure 7. Frequency dependence of G^* and η^* for the neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels at different DSP contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

elastic nature of these hydrogel samples. As can be noticed in this graph (Figure 6), $\tan \delta < 1.0$ throughout the whole frequency range for the pristine PAM/PEI hydrogel and the PAM/PEI-DSP hydrogel samples containing various DSP loadings. This trend signifies that elastic character was predominant in these hydrogels, consequently, a typical solid-like behavior was common in all the hydrogel samples.

Further, the plots of complex modulus (G^*) and complex viscosity (η^*) as a function of ω shown in Figure 7 provided more explanation about these hydrogels. As can be seen in the graph (Figure 7), the complex modulus (G^*) of the composite hydrogels containing various DSP contents, as well as the virgin hydrogel exhibited almost similar trend throughout the whole frequency range with PAM/PEI-0.8 wt % DSP hydrogel showing marginal increment in the entire frequency range. Likewise, it can be observed in this graph (Figure 7) that the complex viscosity η^* values of neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogel samples filled with various DSP contents were almost similar throughout the whole frequency range. This observation suggests that the process-ability of these hydrogels on industrial scale will be easy. Besides, it can also be noticed that the η^* values of all the hydrogel samples decrease linearly as the applied frequency increases, indicating a typical rheological behavior for true hydrogel structures and also suggest a strong shear-thinning behavior.³²

Time Sweep of Neat PAM/PEI Hydrogel and PAM/PEI-DSP Hydrogels

The maturation kinetics of DSP-free hydrogel (i.e., neat PAM/PEI hydrogel) and PAM/PEI-DSP hydrogels filled with various DSP contents were studied by monitoring the evolution of the storage and loss moduli (G' and G'') with time at 30°C. In all cases, both moduli reached stability within 20 min of maturation. As can be noticed in Figure 8, the G' values of neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels incorporated with various DSP quantities were practically higher than the G'' throughout the reaction time, indicating that elastic behavior dominates in these hydrogels. However, the G' values of

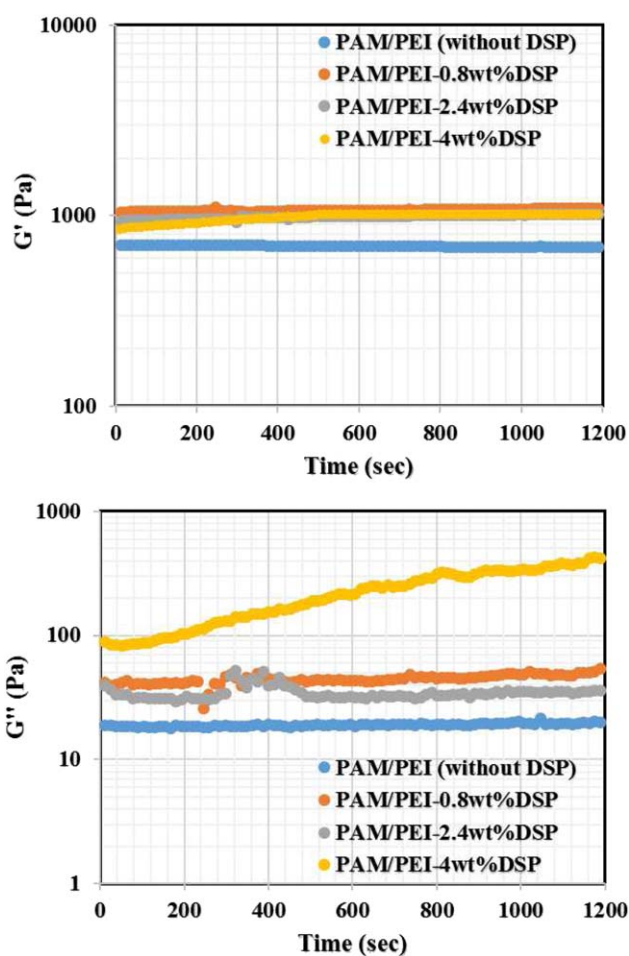


Figure 8. Time sweeps (for 1200 s at 30°C) for the neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels at different DSP contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

PAM/PEI-DSP composite hydrogels filled with various DSP contents were almost the same throughout the reaction time but their respective G' values were higher than that of the neat PAM/PEI hydrogel, signifying that PAM/PEI-DSP hydrogels containing DSP of different quantities are more elastic. It should be mentioned here that the high value of loss modulus G'' demonstrated by PAM/PEI-4 wt % DSP hydrogel is attributed to higher DSP quantities. This phenomenon suggests that the addition of DSP at higher amount (above 0.8 wt %) increases the viscous energy dissipating properties of the composite hydrogels. Similar observation has been reported in a previous study on PAM-based nanocomposite hydrogels.⁵ It is remarkable to point out that in all cases, a stable and fully developed hydrogel structures were attained within very short time of the experiment. This short time which is necessary to achieve stable hydrogel structure represents a significant advantage from an industrial point of view. Therefore, PAM/PEI-DSP composite hydrogels containing various DSP contents are believed to be economical hydrogel systems, which combine excellent compatibility (both PAM/PEI hydrogel matrix and DSP filler).

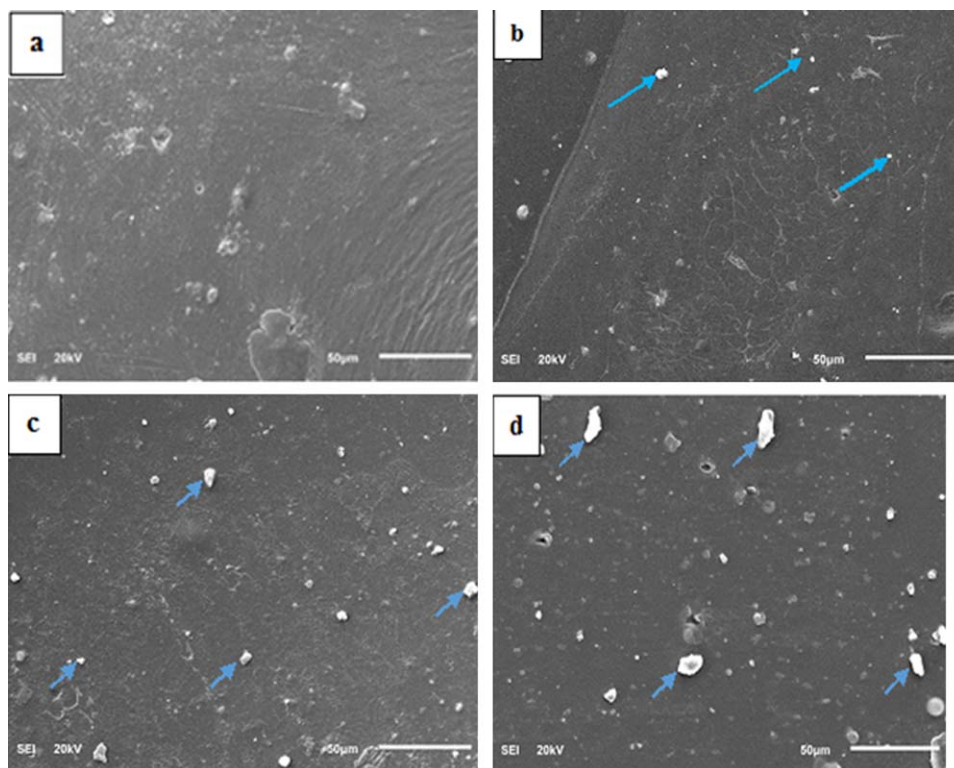


Figure 9. SEM surface micrographs: (a) neat PAM/PEI hydrogel (without DSP), (b) PAM/PEI-0.8 wt % DSP hydrogel, (c) PAM/PEI-2.4 wt % DSP hydrogel, and (d) PAM/PEI-4 wt % DSP hydrogel. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

SEM Studies of Neat PAM/PEI Hydrogel and PAM/PEI-DSP Hydrogels

Morphological characterization of the neat PAM/PEI hydrogel and the PAM/PEI-DSP hydrogels containing various DSP quantities confirmed the results obtained from the rheological measurements. The SEM micrographs revealing the surface morphology of each hydrogel sample are shown in Figure 9(a–d). The micrographs of neat PAM/PEI hydrogel and PAM/PEI-DSP composite hydrogels containing various DSP contents were observed to be smooth, homogeneous and dense. Further, the inclusion of DSP is evident in the micrographs of PAM/PEI-0.4 wt % DSP, PAM/PEI-2.4 wt % DSP, and PAM/PEI 4 wt % DSP composite hydrogels [Figure 9(b–d)]. The “blue-arrows” showing on the micrographs are used to identify the dispersed DSP particles appearing on the surface micrographs of these composite hydrogels.

XRD Studies of Neat PAM/PEI Hydrogel and PAM/PEI-DSP Hydrogels

The structural network of the hydrogels developed was analyzed using X-ray diffraction (XRD) technique. Figure 10 displays the XRD patterns of the virgin PAM/PEI hydrogel and the PAM/PEI-DSP hydrogels containing various DSP loadings. The XRD pattern of DSP-free hydrogel (neat PAM/PEI hydrogel) did not reveal any conspicuous peak as a result of the amorphous nature of PAM and PEI.^{33,34} Conversely, the XRD patterns of PAM/PEI-DSP composite hydrogels filled with DSP contents were quite distinct from that of the pure PAM/PEI hydrogel. The diffractograms of PAM/PEI-DSP hydrogel samples embedded with various DSP contents demonstrated similar character and noticeable broad peaks at $2\theta = 21.3\text{--}24.9^\circ$. These peaks

partly indicate the dispersion of DSP in the matrix of these composite hydrogels and also suggest semi-crystalline domain. The reduction of peaks at $2\theta = 21.3\text{--}24.9^\circ$ associated with the PAM/PEI-2.4 wt % DSP and PAM/PEI-4 wt % DSP hydrogels could be as a result of irregular chain packing, during hydrogel formation. Besides, the absence of prominent sharp peaks beyond $2\theta = 24.9^\circ$ in the XRD patterns of PAM/PEI-DSP

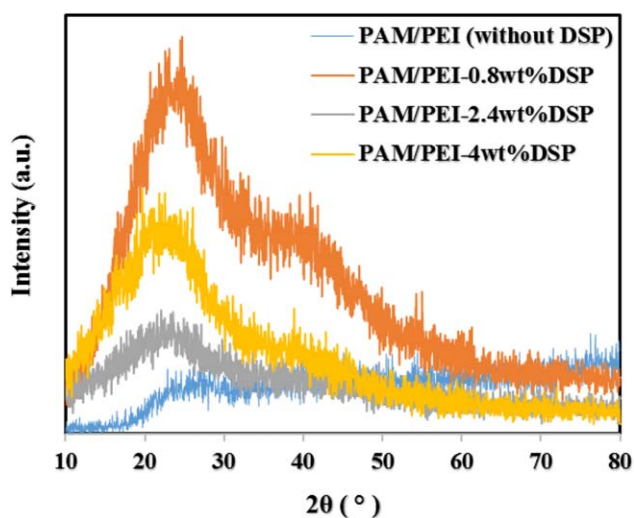


Figure 10. XRD patterns of the neat PAM/PEI hydrogel and the PAM/PEI-DSP hydrogels containing different DSP contents. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

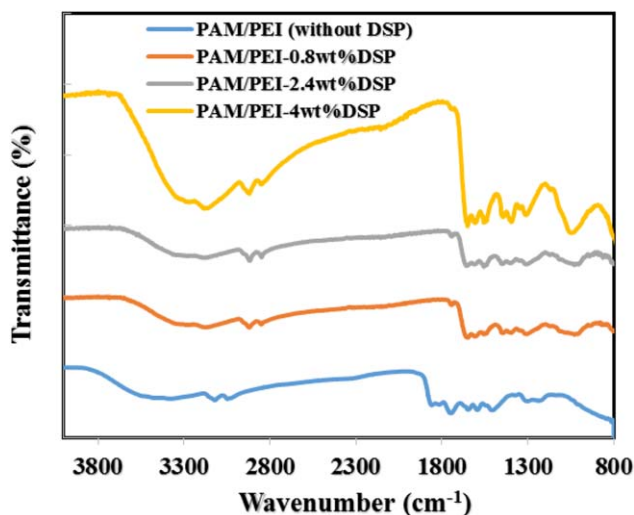


Figure 11. FTIR spectra of the neat PAM/PEI hydrogel and the PAM/PEI-DSP hydrogels containing different DSP contents. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://www.wileyonlinelibrary.com).]

hydrogel samples filled with various DSP loadings suggests that these hydrogel samples are predominantly amorphous.¹²

FTIR Analysis of Neat PAM/PEI Hydrogel and PAM/PEI-DSP Hydrogels

The presence of chemical functional groups in the neat PAM/PEI hydrogel and the PAM/PEI-DSP hydrogel samples incorporated with various DSP quantities were confirmed by FTIR spectra shown in Figure 11. The characteristic bands of the pristine PAM/PEI hydrogel at 1600 and 2935 cm^{-1} correspond to the carboxyl group and methyl group, respectively, providing evidence of crosslinking between PAM and PEI and also derivatization of the amide group. The FTIR spectra of PAM/PEI-DSP composite hydrogels filled with various DSP contents demonstrated peaks in the range of 1730–1735 cm^{-1} , which is attributed to the stretching vibration of C=O in hemicellulose²³ emanating from DSP. Besides, the neat PAM/PEI hydrogel as well as the PAM/PEI-DSP composite hydrogels loaded with various DSP quantities revealed peaks at 2843–2915 cm^{-1} , attributed to methyl stretching and this further corroborate successful crosslinking of the hydrogel samples during their preparation gelation processes.

Potential Applications of the PAM/PEI-DSP Composite Hydrogels

PAM/PEI-DSP composite hydrogels embedded with different quantities of DSP are new class of polymeric composite hydrogels that have promising potentials and opportunities in many fields, especially in the area of wastewater treatment and the removal of heavy metal ions from aqueous solutions. Besides, PAM/PEI-DSP composite hydrogels incorporated with various DSP amounts have the tendency of undergoing structural modifications that could ultimately lead into development of flat sheet polymeric membrane materials for separation process applications.

CONCLUSION

New class of polymeric composite hydrogels has been successfully developed through the crosslinking of polyacrylamide

(PAM) with polyethyleneimine (PEI) in distilled water dispersed with date seed powder (DSP) as biofiller. Neat PAM/PEI hydrogel and PAM/PEI-DSP composite hydrogels incorporated with DSP of various loadings were synthesized. Rheological measurements revealed the viscoelasticity behavior of neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels embedded with various DSP contents. The dynamic strain, dynamic frequency and time sweep tests demonstrated that the PAM/PEI-DSP hydrogel samples are marginally more rigid than the virgin PAM/PEI hydrogel. The SEM characterization showed that the surface micrographs of neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels filled with various DSP contents were smooth, homogeneous and dense. XRD results showed that the structures of neat PAM/PEI hydrogel and PAM/PEI-DSP hydrogels containing various DSP contents were predominantly amorphous while FTIR results confirmed the functional groups and evidence of crosslinking in the neat PAM/PEI hydrogel and the PAM/PEI-DSP hydrogels embedded with different DSP contents.

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