# Gelation of Methylcellulose Hydrogels Under Isothermal Conditions

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**ABSTRACT:** The current work deals with the gelation of methylcellulose (MC) in aqueous solutions under isothermal conditions. The isothermal gelation was monitored during two consecutive heating and cooling cycles. The gelation was observed to occur early during the second cycle and progress at a higher rate. Micelle-like structures were found to form in MC solutions during the first gelation cycle. These were largely responsible for the accelerated gelation during the second heating cycle. The influence from the state of water was examined by studying the gelation phenomenon for MC using either cold or hot DI water as solvent. The possible mechanism involved is discussed. A gel indexing method was established to provide a quantitative measure for the state of gelation achievable using different MC concentrations with either cold or hot water solvent. Stabilization kinetics for the gel under isothermal conditions was described using the Malkin and Kulichikhin model. The kinetics parameters were determined. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 107: 2101–2108, 2008

**Key words:** methylcellulose; isothermal gelation; hydrogel; gel indices; state of water

## INTRODUCTION

Cellulose is insoluble in water due to the formation of strong intermolecular hydrogen bonds between cellulose molecules.[1] Hydrophobic substitution with either methyl or hydroxypropylmethyl groups allows methylcellulose (MC) and its derivatives to dissolve in water showing formation of gel upon heating and reverting back to the solution state upon cooling. The sol–gel transformation of MC with different degrees of substitution has been widely studied in a temperature range of 20–90°C. The driving force to such physical gelation at elevated temperature is believed to be resulting from the disruptive effect on the structure of water.[2] At low temperatures, ice-like structures of water form around the hydrophobic groups (i.e., methyl groups) of MC. Heating of the solutions results in the destruction of these water structures and subsequent exposure of the hydrophobic regions of MC to water, leading to the formation of the hydrophobic aggregates.[3] These aggregates join together to form a three-dimensional gel network involving enthalpy changes.[4,5] When all such interactions of the polymer groups are completed, the network takes in additional water, causing the network to expand. The continued expansion or swelling is due to osmotic forces, which are in opposition with the elastic retraction forces of the network. Eventually an equilibrium and a stabilized state is reached.[6] Thus, the entire gelation process for MC may be described by two processes—(1) transition from solution state to the formation of gel network and (2) swelling and stabilization of the gel. The state of solvent and the magnitude of temperature play key roles in the gelation process.

Gelation of different hydrogels under constant temperatures has been reported by some researchers. Nisbet et al.[7] conducted isothermal (at 37°C) rheological studies on Xyloglucan hydrogels. The aqueous solutions of Xyloglucan at 37°C exhibited a steady rise in the elastic modulus ($G'$) which was frequency-dependent up to the gel point at which drastic increase in the elastic modulus was observed. At the gelation point, $G'$ of Xyloglucan solutions nearly reached a steady value and became frequency-independent. For the isothermal gelation of high-methoxyl pectin, de Silva et al.[8] observed that $G'$ increased rapidly first and then gently before reaching the pseudoplateau region. The time period for the rise in $G'$ was observed to be pectin concentration dependent and the gelation rate close to the gel point could be described as a second-order rate process. Malik and Nandi[9] studied the gelation kinetics of Poly(3-alkyl thiophenes) in xylene using the test-tube tilting method. It was observed that the gelation rate increased in a nonlinear way with concentration, but at higher concentration it leveled up. At a fixed polymer concentration, a gradually decreasing gela-
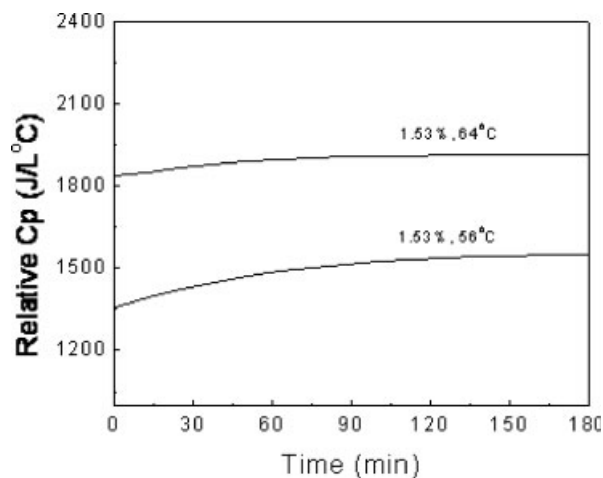
**Figure 1** Relative heat capacity of MC solutions as a function of time under isothermal conditions.[5]

tion rate was noticed with the increasing temperature. The researchers concluded that the gelation of the system proceeded based on three dimensional percolation model. The macroscopic mechanism of gelation did not change with pendent alkyl chain length of Poly(3-alkyl thiophenes). The microscopic mechanism reportedly has followed a two-step process of coil-to-rod transformation and fibrillar crystallization. The present author and his team[5] have reported some preliminary results for MC hydrogels obtained by differential scanning calorimeter (DSC) at constant temperatures. The upward-looking trend for the rate of energy influx under isothermal conditions as shown in Figure 1, reproduced from our earlier publication,[5] is an indication of the energy intake in the hermitically sealed environment provided during DSC tests. Because the temperatures used for these measurements are very close to but a little higher than the onset temperature for the sol–gel transitions in MC solutions, it is possible that gelation occurred at these temperatures although the degree of gelation was low.

All these studies indicate that the sol–gel transformations do take place under isothermal conditions and depend on many parameters. In this article, gelation of MC hydrogels of various concentrations is studied under isothermal conditions. The MC solutions were allowed to undergo two cycles of heating and cooling. The physical appearance of the MC samples was captured at various stages of gelation at constant temperature and subsequently analyzed to build a scale of gel indices to describe the state of gelation. Rheological measurements were carried out to detect the change in the viscoelastic properties of the MC solutions. In some cases, hot deionized (DI) water was used as solvent for preparing MC solutions to examine its effect on the gelation. The possible underlying mechanisms are discussed. The

kinetics of gel stabilization under isothermal conditions for MC hydrogels was examined using the Malkin and Kulichikhin model[10] based on the storage modulus measurements.

## EXPERIMENTAL

### Materials

The methylcellulose (MC) as available in white powder form was purchased from Sigma-Aldrich. Its average molecular weight is 40,000 g/mol. The degree of substitution of methoxy groups is in the range of 1.6–1.9 and the viscosity for a 2% MC solution is 400 cps at 20°C. The MC powder was dried at 60°C overnight and stored in a desiccator before use. Cold DI water was obtained from Millipore α-Q water purifying system.

### Preparation of MC solutions

MC samples with nine different concentrations (0.5, 0.75, 1.0, 1.25, 1.5, 1.75, 2.0, 2.25, and 2.5%) were prepared, using both cold and hot DI water as solvent. All % MC concentrations considered in this study are based on weight percentage. The desired % of MC was dispersed into deionized water at ambient temperature. The mixture was stirred using magnetic stirrer for 24 h. For preparing MC solutions in hot DI water, cold DI water was first heated to 95°C before adding the dried MC powder. All MC solutions were kept in a refrigerator before carrying out any measurements.

### Monitoring of sol–gel transitions

MC solutions in cold DI water with different concentrations of MC were transferred to an oven with the bottle lids removed and their temperature was monitored. The temperature was allowed to stabilize at a desired value. Digital images highlighting the state of each MC solution were captured at every 10-min interval using a digital camera, after the set temperatures as desired, i.e., 40, 50, and 60°C, were reached. The gelation process for each sample was visualized for two heating–cooling cycles. The MC gels after the first heating process were allowed to cool down naturally and kept in the refrigerator for 24 h. The next heating–cooling cycle was performed after this under the same conditions as used for the first cycle. The images of the sol–gels obtained at different time intervals were analyzed using Adobe Photoshop for examining the state of gelation under the chosen isothermal conditions. A detailed discussion of this protocol is included in the Appendix. In summary, the index between 1 and 3 was considered to be indicating the solution state. Index 3–6 represented the

sol–gel state, and Index 6–10 was allocated to the gel state. Index 6 indicated weaker gel network as compared with the stronger gel network indexed as 10.

The microstructural changes during the gel formation process were examined using a confocal microscope (Axiotron 2, Zeiss) coupled with a Linkam hot plate (CSS 450, Linkam Scientific Instruments). The temperature of the hot plate was stabilized at 60°C before any MC sample was loaded on to the plate. The sol–gel transition was continuously monitored until no further change in the microstructure could be observed.

### Rheological measurements

A strain-controlled rheometer (ARES, Rheometric Scientific) was used to measure the viscoelastic properties of the MC solutions under isothermal conditions. The rheometer works in oscillatory mode and is equipped with two sensitive force transducers for torque measurements ranging from 0.004 to 100 g cm. A set of parallel plates of 50 mm in diameter was used for the test. A small amount of silicone oil was applied at the periphery of the gap between the plates holding the MC solution to prevent dehydration. The temperature of the sample was increased from 25 to 50°C and maintained at 50°C by circulating water from a circulator through the jacket of the lower parallel plate. A strain sweep at a low frequency (1 rad/s) was carried out to determine the linear regime of the viscoelasticity. The subsequent rheological measurements were conducted at the same frequency and at the strain amplitude of 0.5%, which ensured the linear viscoelastic behavior of the MC hydrogels.

### RESULTS AND DISCUSSIONS

### Temperature selection for isothermal gelation

Based on a previous study, the onset temperature for the gelation of aqueous MC solutions was below 60°C.[4] Therefore, the constant temperatures chosen for the current study were 40, 50, and 60°C, respectively. It was found that the gelation rate for different MC samples was insignificant at 40°C, while a relatively high rate of gelation was observed before the MC solutions reached the set temperature of 60°C. The thermal gelation of MC under the isothermal conditions at 50°C was found to be occurring at the right pace required for experimental observations. Thus, the temperature of 50°C was chosen for the subsequent measurements and imaging for defining the gel indices. It may be noted that the gel formation temperature can be brought down with the addition of certain human-body-compatible salts.[4] However; this was avoided in the current studies so as to help in clear understanding of the solvent effects.

### Early gelation of MC during the second heating–cooling cycle

Each sample prepared in cold DI water was allowed to go through two consecutive heating–cooling cycles. Monitoring of the gelation process began after the sample temperature reached 50°C. The minimum MC concentration necessary for the obvious gelation when the sample reached the set temperature of 50°C was 1.75% for the first cycle. It decreased to 1.25% when the studies were based on two cycles. As an example, the images obtained for the first and the second gelation cycles for MC with a concentration of 1.5% are shown in Figure 2. The state of gelation for each MC sample was obtained by analyzing the images captured at different time and the results of the analysis are presented in Figure 3 in terms of gelation index. With reference to Figure 3(a), for the first gelation cycle, the MC samples with low concentrations (<1.5%) remained in the solution state throughout the observation period of 85 min in the oven. The transition from solution to the gel state was observed for the high concentration samples
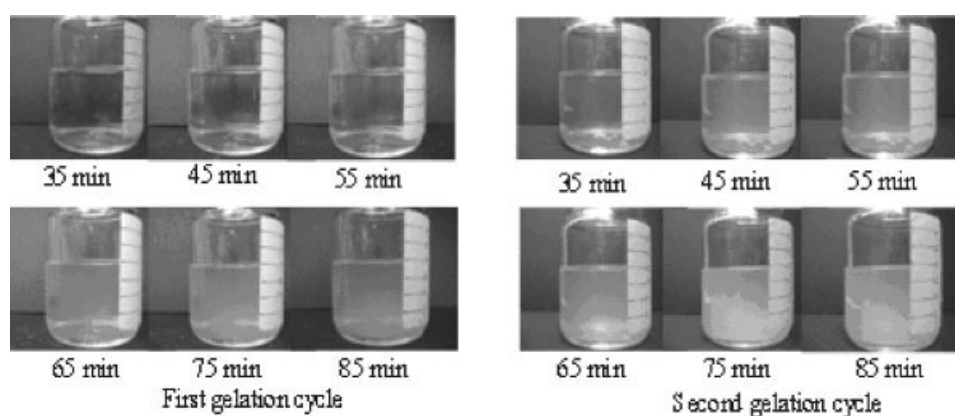


**Figure 2** State of gelation observed for 1.5% MC in cold DI water at 50°C.
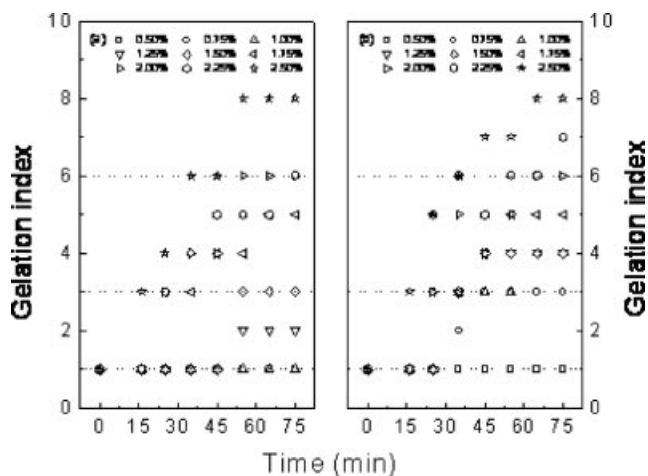
**Figure 3** Gel index variations showing distribution of gel density for MC gel in cold DI water at 50°C isothermal hold: (a) for the 1st gelation cycle; (b) for the 2nd gelation cycle.



**Figure 4** Micelle-like structures formed in MC solutions after one gelation cycle.

(i.e., % of MC ≥ 1.5%). By maintaining the MC solutions at 50°C, the sol–gel transition of the MC samples with concentrations between 1.75 and 2.25% commenced after 25 min and did not complete even by 75th minute. The sol–gel transition of 2.5% MC solution commenced after 16 min and completed by 35th minute. During the second gelation cycle, no gelation was observed for the MC solutions of 0.5 and 0.75% within the first 75 min at 50°C; refer Figure 3(b). MC solution of 1.5% exhibited initiation of the sol–gel transition within the first 35–45 min range as compared with 55–65 min range observed for the first heating cycle. For the same time frame, the turbidity during the second cycle was apparently higher than that during the first cycle. The MC solution of 2.0% became gel after 65 min whereas 2.5% solution turned into a gel within 35 min at 50°C. These observations clearly indicate that the sol–gel transition occurred earlier during the second heating cycle at an enhanced gelation rate.

To find out the possible reasons for the increased gelation rate and the early occurrence of MC gelation during the second cycle, the confocal microscope was used to monitor the changes in the microstructure at a steady temperature. Interestingly, micelle-like structures (Fig. 4) which could not be found in fresh MC solution were observed in the sample that underwent one heating cycle. These micelle-like structures might be formed through intramolecular micellization, which was concomitant with the gelation due to intermolecular association based on formation of hydrophobic junctions.[11,12] Hydrophobic association of the methoxy groups on the same side of a MC chain may result in the formation of isolated loops as shown in Figure 5(a). It is supposed that a substantial amount of energy is

required to facilitate the movement of such isolated loops in the solution. A certain number of such isolated loops, which are held in place by surrounding water molecules, integrate together and turn into micelle-like structures; see Figure 5(b). It should be noted that the micelle-like structures are quite stable because they could be observed even after the refrigeration of the samples. The existence of such micelle-like structures accelerate the MC gelation during the second heating cycle because of the early exposure of the hydrophobic groups associated with the isolated loops when the water cages surrounding them break. Additionally, water molecules in MC solution gain internal energy due to heating and become more dispersed. The water cages thus formed around the hydrophobic methoxyl groups might not be as compact as they originally were before any gelation took place. As a result, the sample during the
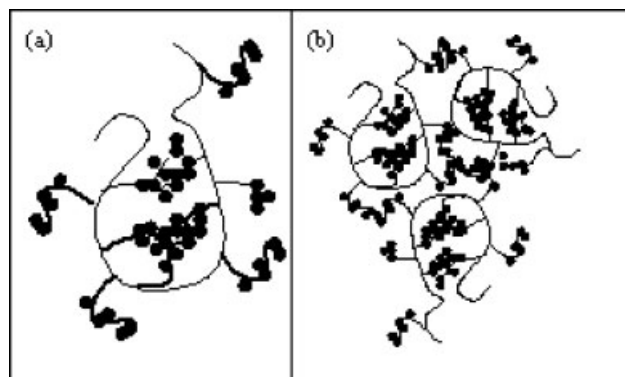


**Figure 5** Schematic illustration of MC micelle formation: (a) MC isolated loop formed by intramolecular association; (b) MC micelle formed when a small number of isolated loops of MC associate together. (Black dots represent the water molecules, light lines are the MC chains, and dark lines are the methoxy side groups).
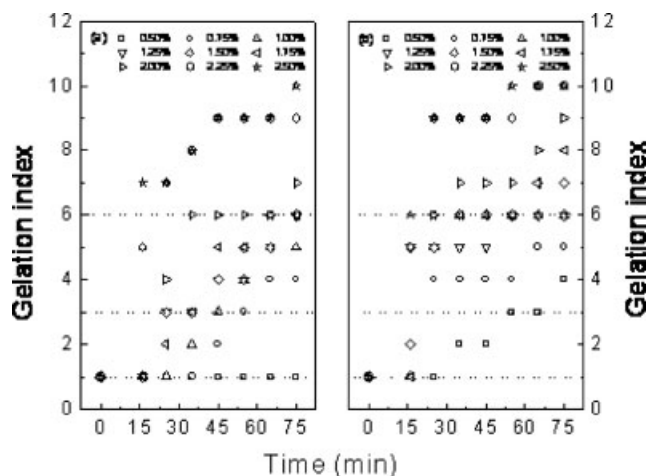
**Figure 6** Gel index variations showing distribution of gel density for MC gel in hot DI water at 50°C isothermal hold: (a) for the 1st gelation cycle; (b) for the 2nd gelation cycle.

second gelation cycle required shorter time to absorb enough energy to break the water cages when held at 50°C. This, in turn, resulted in early exposure of the hydrophobic groups and early occurrence of the hydrophobic association which accelerated the sol–gel transition.

### Influence of the state of water

To investigate the possible influence of the state of the water solvent, similar experimental studies were carried out on the MC hydrogels prepared in hot DI water. Figure 6 shows the state of gelation of these samples during the first heating cycle at 50°C. The minimum MC concentration for an obvious change to a sol–gel state at 50°C was 1.25% as compared with 1.75% sample prepared in cold DI water. As compared with Figure 3(a), 0.75% MC concentration in hot DI water no longer remained a transparent solution, but changed to a sol–gel state after 58 min at 50°C. The sol–gel transition of the MC samples with

0.75–1.75% concentrations commenced at an earlier time as compared with the corresponding samples prepared in cold DI water although the transition did not complete within 75 min. The sol–gel transition of the MC samples with concentrations beyond 1.75% commenced earlier as compared with the corresponding samples prepared in cold DI water. The sol–gel transition of 2.0, 2.25, and 2.5% MC samples completed after 72, 23, and 13.5 min, respectively. Under the same conditions, the MC solutions prepared in hot DI water exhibited higher gelation rate than those prepared in cold DI water. It is supposed that the dispersion of MC is very fast and the hydration of MC occurs almost immediately after the MC powder is added to DI water at 95°C. Since the strength of the hydrogen bonds between the water molecules decreases with the increasing temperature, some hydrogen bonds in water structure break and intermolecular hydrogen bonds between the water and the MC molecules come into being.[13,14] At the same time, the probability of intramolecular association of MC chains increases and the early commencement of sol–gel transition becomes possible. This process, although not very obvious as compared with the actual heating cycles mentioned in the previous section, may be considered as a type of elementary gelation process. Therefore, some micelle-like structures may have formed during such mixing process at 95°C. This might have also added to the early occurrence of gelation and the enhanced gelation rate for the MC hydrogels prepared in hot DI water.

The time data for the onset, and completion of the gel formation process for the 2nd cycle, deduced from Figures 3(b) and 6(b), is presented in Table I. For the 2nd heating cycle using cold DI water as solvent, the onset time of the MC gelation shows a clear dependency on MC concentration. With the increase in MC concentration from 0.75 to 2.5%, the initiation of gelation decreased from 45th minute to 16th minute. The completion time for gelation reduced from 65 to 35 min when the MC concentration was increased from 2.0 to 2.5%. Thus, the respective gela-

## TABLE I
### MC Gel Initiation and Formation Time for the 2nd Cycle

| MC Concentration (%) | 0.50 | 0.75 | 1.00 | 1.25 | 1.50 | 1.75 | 2.00 | 2.25 | 2.50 |
|---|---|---|---|---|---|---|---|---|---|
| Cold DI water as solvent | | | | | | | | | |
| Onset time (min) | a | 45 | 35 | 35 | 35 | 25 | 25 | b | 16 |
| Completion time (min) | a | a | a | a | a | a | 65 | 35 | 35 |
| Hot DI water as solvent | | | | | | | | | |
| Onset time (min) | 55 | b | b | b | b | b | b | b | b |
| Completion time (min) | a | a | 35 | 55 | 35 | 25 | 25 | b | 16 |

[a] Complete gel formation was not observed within the first 85 min.
[b] Gel initiation point was not captured. As the gelation rate was higher, sol–gel transition completed before any image of taken.
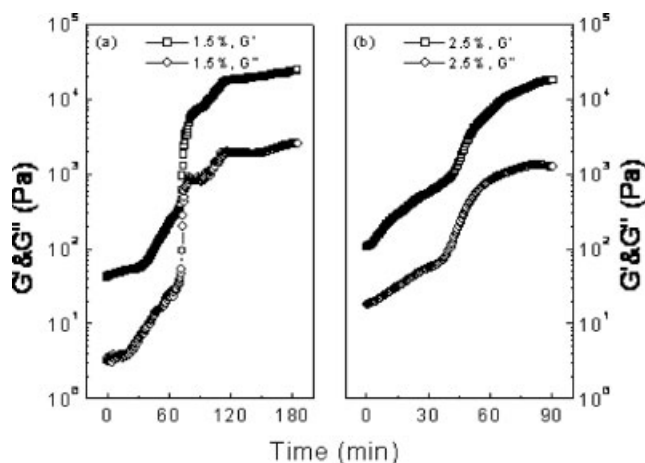
**Figure 7** Time dependence of $G'$ and $G''$ for MC (1.5 and 2.5%) prepared in cold DI water (a strain amplitude of 0.5% and an angular frequency of 1 rad/s were used. The measurements were carried out during the 1st gelation cycle at 50°C).

tion time for the MC samples of 2.0 and 2.5% was 40 and 19 min. For the 2nd heating cycle for hot DI water MC solutions, the onset time for gelation was not captured for most MC samples because of the higher gelation rate and the less amount of time required for completion of the sol–gel transition. However, the dependency of gel completion time on the MC concentration can be observed clearly; the completion time decreased from 55 to 16 min with the increase in the MC concentration from 1.25 to 2.5%. Hot DI water as solvent resulted in reduced gel formation time; for example, for 2.0% MC sample, it reduced from 65 to 25 min whereas it dropped from 35 to 16 min for 2.5%, as compared with cold DI water as solvent.

Based on the observations, the sol–gel transition of 2.5% MC hydrogel occurred at an earlier time as compared with the other samples and the time needed for the completion of the gelation was also the shortest one. Therefore, 2.5% appears to be the best MC concentration among the set of gel solutions studied. The 2.5% MC produced a thicker gel due to the higher number of hydrophobic groups and proved optimum.

## Viscoelastic behavior upon heating

Figure 7 shows $G'$ and $G''$ measurements for MC samples of 1.5 and 2.5% in cold DI water as a function of time under isothermal conditions at 50°C during the 1st gelation cycle. As seen from Figure 7(a), both $G'$ and $G''$ for the MC sample of 1.5% concentration increased steadily upon heating. A sudden increase in $G'$ and $G''$ was observed after 40 min. This may be due to the completion of inter-molecular association of MC chains, indicating the

formation of three-dimensional gel network.[15] Thereafter, both $G'$ and $G''$ increased gradually with time until they leveled off and became time independent. This is an indication of the swelling and stabilization of the gel where the gel gained strength with time. During the entire heating process, the value of $G'$ was higher than that of $G''$. At the MC concentration of 2.5%, the largest change in $G'$ and $G''$ occurred at an early stage, refer Figure 7(b). Thus, the initial rise in $G'$ and $G''$ was faster and less time was needed to reach a plateau. The thermal behavior of MC hydro-gels revealed by the rheological measurements is consistent with that observed from the gelation indexing in Figure 3. Figure 8 shows $G'$ and $G''$ for 1.5% MC hydrogels prepared in hot and cold DI waters, respectively. Upon heating to 50°C, the sample prepared in hot DI water showed an early and faster rise in $G'$ and $G''$ as compared with the sample prepared in cold DI water. This is another proof to the fact that the hot solvent favors gelation.
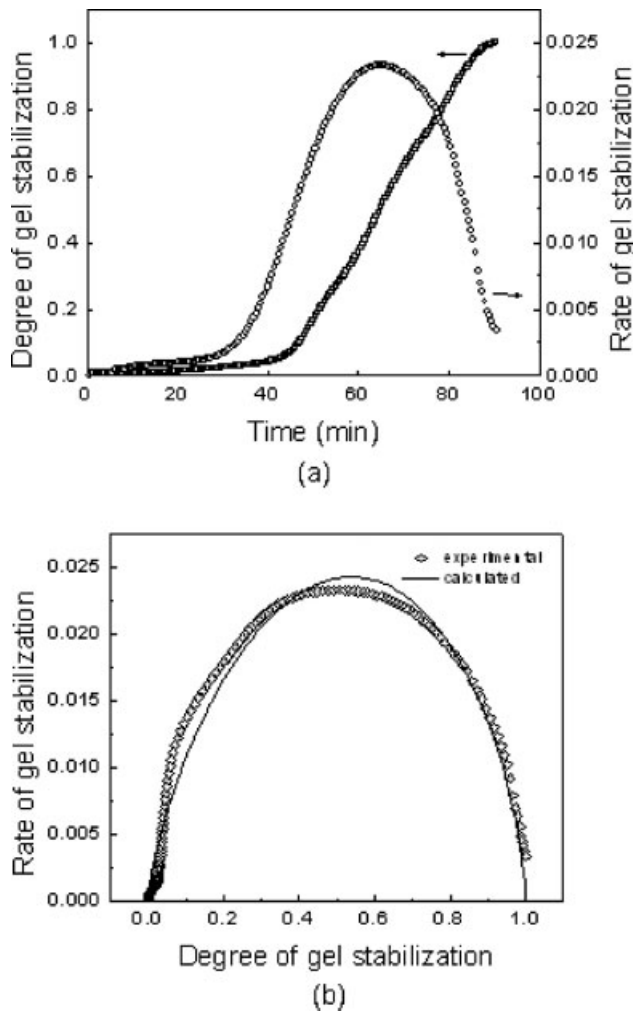
## Isothermal gel stabilization kinetics

The gel stabilization kinetics for 2.5% MC gel prepared in cold water was evaluated using the rheological data obtained for the 1st gelation cycle [Fig. 7(b)]. The degree of gel stabilization, α, was used to indicate the state of dynamic equilibrium attained by the gel and was evaluated at 50°C from the time dependence of $G'$ [Fig. 7(b)] using the relation:

$$\alpha_i = \frac{G'_i - G'_0}{G'_f - G'_0} \tag{1}$$

where, $G'_i$ is the value of the storage modulus at time $t_i$, and $G'_0$ and $G'_f$ are the values of the storage
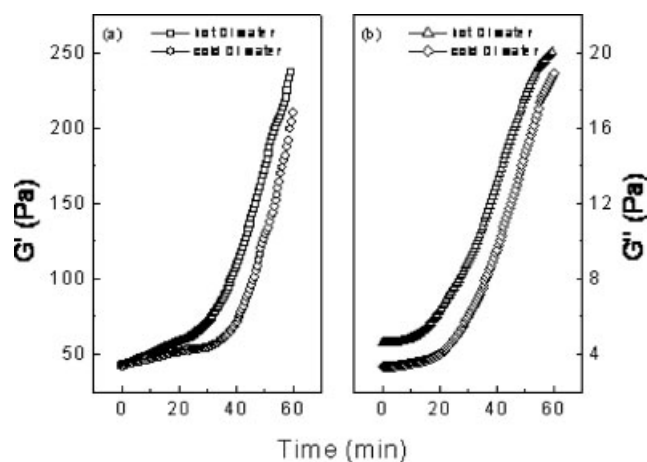


**Figure 8** Comparison of $G'$ and $G''$ for MC (1.5%) prepared in cold and hot DI water (A strain amplitude of 0.5% and an angular frequency of 1 rad/s were used. The measurements were carried out during the 1st gelation cycle at 50°C).

Figure 9 Kinetics of isothermal gel stabilization during the 1st cycle at 50°C for MC (2.5%) prepared in cold DI water: (a) degree of gel stabilization as well as rate of gel stabilization as a function of time; (b) rate of gel stabilization as a function of degree of gel stabilization. The symbols are experimental data and the line is calculated from Eq. (3).

modulus at the beginning of the experiment and at the maximum gelation time, respectively. Figure 9(a) shows the degree of stabilization [determined using Eq. (1) and the data presented in Fig. 7(b)] as a function of time. At a constant temperature of 50°C, $\alpha$ was extremely low until 35 min, which signified the onset of water uptake and gel stabilization process. Thereafter $\alpha$ increased significantly with time.

Figure 9(a) also shows the rate of gel stabilization, $(d\alpha/dt)_i$, calculated numerically as:

$$\left(\frac{d\alpha}{dt}\right)_i = \frac{G'_i - G'_{i-1}}{t_i - t_{i-1}} \tag{2}$$

where $G'_i$ and $G'_{i-1}$ are the values of storage modulus at time $t_i$ and $t_{i-1}$ during 1st isothermal gelation cycle at 50°C, and $(d\alpha/dt)_i$ is the rate of gel stabilization at

time $t_i$. For a MC solution with 2.5% concentration, the sol–gel transition occurred at 16th minute [Fig. 3(a)] during the 1st cycle but the rate of gel formation was very low. Around 35th minute from the beginning of experiment, the sol–gel transition phase completed [Fig. 3(a)]. The gel observed at this stage was very weak and far from its stabilized state. Therefore, the state of gel continued to vary with further heating at 50°C and increased gel modulus was observed during the rheological measurements. Such process that started after the completion of sol–gel transition is an indicative of gel stabilization process, which represents the progressive development of gel strength due to water uptake leading to swelling and orderliness of the gel network structure. As shown in Figure 9(a), a rapid increase in the rate of gel stabilization was observed after 35 min until a maximum value was reached at around 65th minute from the beginning of the measurement. Subsequently, the rate of gel stabilization decreased to a lower value within the next 20 min, indicating that the gel had approached its necessary equilibrium state.

Further, a popular model from Malkin and Kulichikhin[10] as in Eq. (3), was used to describe the gel stabilization kinetics for the same 2.5% MC gel under isothermal conditions

$$\left(\frac{d\alpha}{dt}\right)_i = (k_1 + k_2\alpha_i^m)(1 - \alpha_i)^n \tag{3}$$

where $k_1$ and $k_2$ are temperature-dependent rate constants for the early and late stages of the gel stabilization. $m$ and $n$ are empirical constants, the sum of which gives the order of the stabilized gel. In Figure 9(b), the symbols are the experimental data representing the rate of gel stabilization $(d\alpha/dt)_i$ as a function of $\alpha_i$. The value of $k_1$ in Eq. (3) was calculated graphically as the initial rate of gel stabilization at $t = 0$ given by the intercept to the curve $(d\alpha/dt)_i$ versus $t_i$. The other kinetic parameters $k_2$, $m$, and $n$ were determined by fitting the experimental data presented in Figure 9(b) to Eq. (3) using nonlinear regression analysis. The values for all the four parameters are shown in Table II. The curve in Figure 9(b) was computed using Eq. (3) with $k_1$ $k_2$, $m$, and $n$ as best-fit parameters. Clearly, the thermally-induced stabilization kinetics of MC gels can be well

**TABLE II**
**Kinetic Parameters for Isothermal Gel Stabilization During the 1st Cycle at 50°C for MC (2.5%) Prepared in Cold DI Water**

| Temperature (°C) | $k_1$ (min$^{-1}$) | $k_2$ (min$^{-1}$) | $m$ | $n$ |
|---|---|---|---|---|
| 50 | 0.0001 | 0.0626 | 0.7441 | 0.6344 |

described by the Malkin and Kulichikhin model. Since this model is generic in nature, it can be employed to describe isothermal gelation of aqueous MC systems with any desired concentration.

## CONCLUSIONS

MC displays the ability to gel under isothermal conditions. Conditioning of aqueous solutions of MC during sol–gel transformations for at least one cycle facilitates the formation of micelles, which further favors accelerated gel formation. The initial state of the solvent is seen to affect the gel formation process. Hot DI water has favorable impact on the rate and the state of gel formation as compared with the cold DI water. This can be attributed to the decreased intensity of hydrogen bond and the early exposure of hydrophobic side groups to the MC chains in the solution. Gelation index based on digital images of the solutions could be used as a quantitative measure for understanding the state of gelation achievable using different MC concentration with either cold or hot solvents. The generic model given by Malkin and Kulichikhin worked very well in describing the stabilization kinetics for MC hydrogels based on rheological measurements.

## APPENDIX: INDEXING FOR STATE OF GELATION

An attempt was made to index the different states of MC samples using a scale of 1–10. The Adobe Photoshop software was used as the tool for analyzing the color of the digital images of the MC solutions. The primary color of the MC solutions was analyzed by breaking it into three basic color components—red, blue, and green. Each color component had a value between 0 and 255. The value for each of the basic components is 0 for pure black while pure white yields a value of 255. A grid of 1-mm size squares was created on the image of the gel container so that the same spot on image could be selected in analysis for consistency. The spot chosen was in the middle at coordinates (8 mm, −3 mm). To establish the scale

for the gel index from 1 to 10, two samples of MC were chosen as reference. An image of the sample with 0.5% of MC prepared using cold DI water observed at starting point of the first gelation cycle was used for describing Index 1. The image of the sample with 2.5% of MC prepared using hot DI water observed at time equal to 75 min of the second gelation cycle was used to describe Index 10. The solution with Index 1 had 96 red, 82 blue, and 97 green components whereas the gel with Index 10 was much thicker and had 197 red, 207 blue, and 214 green components. The values of the three basic color components were subsequently interpolated for arriving at the remaining gel indices. The index between 1 and 3 was considered to be indicating the solution state. Index 3–6 represented the sol–gel state of an increasing % of gel, and Index 6–10 was allocated to the gel state. No. 6 indicated weaker gel as compared with the stronger gel indexed as 10.

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