Enzymatic Coupled Mechanical Defibrillation Process for the Production of Corn (Zea Mays) Cob Microfibrillated Cellulose: Preparation, Characterization and Evaluation as Pickering Emulsifier for Oil-In-Water Emulsion

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Research Article

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Enzymatic coupled mechanical defibrillation process for the production of corn (*Zea mays*) cob microfibrillated cellulose: preparation, characterization and evaluation as Pickering emulsifier for oil-in-water emulsion

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Abstract

Microfibrillated cellulose (MFC) is a type of nanocellulose having multiple functionalities. Typically, MFC was produced from mechanical high pressure homogenization process. However, this process is energy intensive and the fibrous nature of MFC often causes instrument blockage. The present study aims to utilize endoglucanse enzyme as environmentally friendly approach to pretreat fiber structure prior to undergoing mechanical defibrillation for the production of MFC from corn cob. Alkaline and bleached pretreated corn cob was treated with endoglucanase Fibercare R from 0% to 2.5% before passing through high pressure homogenizer. It was found that incorporation of 0.02% of endoglucanase was sufficient to soften the corn cob cellulose and further prevent the blockage of homogenizer. Subsequently, the 0.02% endoglucanse treated corn cob was passed through different cycles of homogenization from 0 cycle to 10 cycle for MFC production. It was observed that the water retention, zeta potential and shear viscosity of the MFC increases with homogenization cycle. MFC produced had a gel like consistency. Next, emulsifying stabilizing properties of MFC produced from cycle 0 to cycle 10 as well as their amount from 0 % to 1% were also assessed. Increase in homogenization cycle and the amount of MFC promote emulsion stability as observed from the low creaming index which is mainly attributed to the high shear viscosity and G'G'' crossover of the emulsion. In all, the MFC derived from corn cob *via* enzymatic coupled with high pressure homogenization process has the potential to be used as gel like stabilizer in oil-in-water food emulsion system.

Keywords: nanocellulose, emulsion, rheology, stability, corn cob

1 Introduction

Microfibrillated cellulose (MFC) is a purified form of cellulose that is isolated from cellulose fiber having a high aspect ratio with few nanometer in diameter and several microns in length. When suspended in water, MFC creates a strong three-dimensional network structure that is able to give rise to viscous-gel like properties. Attributed to its sustainable and multifunctional properties, MFC received a lot of attention for used in various applications in electronic, biomedical and food industries. Traditionally, MFC was produced from softwood pulp. However, today, a lot of attention focuses on the use of herbaceous plant or agricultural waste materials for MFC production due to sustainability issue. Herbaceous plant offer more advantages over softwood as it is

readily available, sustainable, and possesses simple structural make up than wood. Thus, herbaceous plan requires a lesser and simpler pretreatment process to disintegrate and defibrillate the fiber during the production of MFC (Sedjo and Lyon 2015; Trache et al. 2017).

Often, MFC were prepared using mechanical shearing approach such as high pressure homogenization to defibrillate cellulose bundle into filament form. Winuprasith & Suphantharika (2013) employed high pressure homogenization process to produce MFC from mangosteen skin. However, high emulsification process has its setbacks. It is often regarded as an energy intensive process and frequently causes blockage or breakdown of high pressure homogenizer especially when the sample treated is hard in texture or has uneven particle size (Henriksson et al. 2007). As a result, upscale and commercialize of MFC produced *via* high pressure homogenization approach is considered expensive and a challenging task.

An emulsion is a mixture of two or more liquids that are immiscible. Conventionally, emulsion was stabilized by stabilizers that is mainly derived from surfactants. Nevertheless, emulsion can also be stabilized by solid particles and the emulsion formed is known as 'Pickering emulsions' (Pickering 1907). Example of solid particles that can be utilized as Pickering emulsifier include: hydrophobized fumed silica (Frelichowska et al. 2010), food-grade particles like protein, fat crystal, and polysaccharide complexes (Tavernier et al. 2016), bacterial cellulose nanocrystals (Kalashnikova et al. 2011). Particle stabilized emulsion offers several advantages than those prepared from surfactants. Pickering emulsion offers to be a good source in replacing or reducing the usage of emulsifier that may often cause adverse health effects such as irritancy. Furthermore, emulsion stabilized by solid particle is extremely stable against coalescence. Apart from the above mentioned particles, nanocellulose such as MFC or nanocrystal cellulose was the recent particle used for stabilizing emulsion. For instance, study performed by Winuprasith et al (2013) utilized 0.7% of nanocellulose from mangosteen rind to stabilize a 30% and 10% w/w soybean oil emulsion system. The study revealed that MFC demonstrated the ability to restrict the movement of emulsion droplets and prevent the dispersed phase from coalescing in oil-in-water emulsion system (Winuprasith and Suphantharika 2013). Unlike other solid stabilizers, cellulose based stabilizer possesses extra attributes because of it zero calorie characteristic (Anderson and Eastwood 1989; Slavin 2005). This property further extended the utilization of MFC

in food industries as low calorie stabilizer in developing healthy low or reduced calorie food emulsion products.

Corn is one of the few important cereal crops after wheat and rice that is utilized widely as staple food worldwide. Out of total corn production, there is around 15% of waste being generated from corn processing in the form of corn cob (Gradinaru et al. 2018). Today, majority of the corn cob produced is widely used as heat generator, animal bedding, oil sorbents, polishing agents, biofuel and activated carbons. Nevertheless, corn cob is a rich reservoir of carbohydrates (Maha et al., 2010). Study found that corn cob consists mainly of carbohydrate in the form of 38.8% cellulose, 44.4% hemicellulose and 11.9% lignin (Pointner et al. 2014). Furthermore, it has relatively low impurities since it is covered with husk and corn kernel.

Therefore, the present study aims to explore the transformation of underutilized corn cob waste into value added MFC that can be used as rheological modifier to stabilize oil-in-water emulsion system using an improved and milder high pressure homogenization approach *via* the aid of enzyme. The current work investigated the facilitation of endoglucanse Fibercare R to defibrillate fiber bundle for the production of MFC using high pressure homogenizer to resolve the blockage issue of high pressure homogenize. In the present study, the effect of different amount of endoglucanse enzyme (0% to 2.5%) on the MFC properties produced as well as the tendency of blockage of homogenizer was evaluated. Subsequently, the current work also investigate the influence of the cycles of homogenization (cycle 0 to cycle 10) and amount of MFC (o% to 1%) in stabilizing oil-in-water emulsion.

2 Materials and Methods

2.1 Materials

Corn cob was obtained from local corn supplier Nelson's Franchise (M) Sdn Bhd (Shah Alam, Selangor, Malaysia). The raw corn cob consisted of 43.8% cellulose and 47.7% hemicellulose, respectively. It was ground using coconut grater machine and subsequently pressed using coconut milk separator to remove the juice from the corn cob. Subsequently, the corn cob was dried in an oven at 55^{o}C and sieved through 1mm mesh size sieve. Pulpzyme HC (xylanase) and Fibercare R were purchased from Novozyme (Bagsvaerd, Denmark). Palm olein (Buruh, Malaysia) with IV 56 was purchased from local hypermarket. All the reagents and chemicals inclusive of sulphuric acid (Merck,

USA) potassium permanganate (Merck, USA), sodium hydroxide (Merck, USA) and sodium hypochlorite (Merck, USA) used were of analytical grade.

2.2 Production of MFC

Alkaline pretreatment was performed to remove the hemicellulose from corn cob. A 6% (w/w) dried corn cob was treated with 0.5M sodium hydroxide for 30 min at 80 °C. After alkaline treatment, the corn cob suspension was filtered and washed thoroughly with ultrapure water. Alkaline treated corn cob was then treated with Pulpzyme HC. Pulpzyme HC of 0.03476% (v/v) was added to the 3% alkaline treated corn cob that was suspended in buffer solution containing 0.11M KH_2PO_4 and 0.09M Na_2HPO_4. After 45 min, the corn cob fiber was treated with endoglucanse Fibercare R of varies concentration from 0%, 0.02%, 0.10%, 0.5% to 2.5% (denoted as G0, G0.02, G0.10, G0.50 and G2.50) for 2 hours. Endoglucanse Fibercare R was used for softening the corn cob to ease the fiber passage through the subsequent high pressure homogenization process used for the production of MFC. The enzymatic reaction was terminated by heating the suspension at 80 °C for 30 min. Then, bleaching was performed to remove lignin from the treated corn cob. The pretreated corn cob were bleached with 0.6% sodium hypochlorite at 80 °C for 2 hours and the filtrate were washed thoroughly with ultrapure water. Next, a 0.5% of the 0.02% endoglucanase treated corn cob were homogenized using high pressure homogenizer (HPH) (Panda 2K, Niro Soavi, Deutschland, Lubeck, Germany) at 1000 bar at different passes consisting of 0, 2, 4, 6, 8 to 10, respectively to produce MFC. In order to evaluate the addition of Fibercare R in easing the homogenization process, any blockage in the high pressure homogenizer were recorded based on observation. Blockage was considered when fiber suspension were not able to pass through high pressure homogenizer. Alkaline and bleaching pretreatment managed to successfully removed hemicellulose and lignin giving rise to MFC with 78.9% of cellulose and 14.9% of hemicellulose content.

2.3 Characterization of MFC

2.3.1 Degree of polymerization

The degree of polymerization of MFC and raw corn cob were determined according to ISO 5351:2012 method and calculated from the intrinsic viscosities at 25 °C using the equation (1):

$$[\eta] = 0.891DP^{0.936}\text{--------}(1)$$

where η represents intrinsic viscosity and DP represents degree of polymerization

2.3.2 Stability

Stability index and water holding capacity of MFC were evaluated using optical centrifuge analyzer (Lumifuge 114 LUM GmbH, Berlin, Germany) with a 2 mm path length rectangular sample tubes. MFC was centrifuged at 2000 rpm for 127.5 min at 25 °C. Stability index was calculated based on equation 2:

$$\frac{\text{length of low trasmision area (MFC area)}}{\text{Total sample length}} x\ 100\text{------}(2)$$

Water holding capacity of MFC produced were calculated using equation 3:

$$\frac{\text{Stability index}}{\text{MFC dried weight x 100}} \text{x volume------}(3)$$

Zeta potential of MFC were measured using Zetasizer Nano ZS (Malvern Instruments, Malvern, UK) via a disposable folded capillary cells (DTS 1060) at 25 °C.

Water evaporation test was carried out by incubating the MFC at 60 °C for a duration of one week. The percentage of water lost was calculated using equation 4:

$$\frac{\text{initial weight} - \text{final weight}}{\text{inital weight}} \text{x100------}(4)$$

2.3.3 Morphology

Surface morphology of the MFC were examined with a JEOL 6400V scanning electron microscope (JEOL USA, Inc., Peabody, MA, USA). MFC were freeze dried and subsequently affixed to an aluminum stub using carbon filled tape. The assembly was then coated with gold before examine under electron microscope.

2.3.4 Rheology

Rheological properties of the freshly prepared MFC suspensions were measured using a rheometer (Thermo Scientific HAAKE Rheostress 6000 Universal Rheometer, Buckinghamshire, Germany) equipped with a 60 mm diameter titanium plate-and-plate with universal temperature controller. A 2.9ml of sample was rest for 5 minutes on peltier plate to erase additional stress as well as to equilibrate the temperature. Shear viscosity was monitored by increasing the shear rate from 0.1 to 300 s^{-1} at 25 °C. A linear

viscoelastic range (LVR) was determined with a strain sweep at frequency of 1 Hz from 0.01 Pa until 100 Pa for dynamic viscoelastic measurements. The determined LVR was subsequently used for the frequency sweep. A dynamic frequency sweep was conducted by applying a constant strain of 0.1 Pa (determined by LVR) which was within the linear region over a frequency range between 0.1 Hz and 10 Hz.

2.4 MFC stabilized oil-in-water emulsion

MFC obtained under HPH cycles of 0, 2, 4, 6, 8 and 10 cycles (denoted as C0, C2, C4, C6, C8 and C10) were utilized to stabilize oil-in-water emulsions. The emulsions were prepared by pre-mixing 9 parts of 0.5% MFC with 1 part palm olein (w/w) using rotor-stator (Silverson L4R, Buckinghamshire, UK) set at 7000 rpm for 5 minutes The coarse dispersions were then passed through high pressure homogenizer (Panda 2 K, Niro Soavi, Deutschland, Lubeck, Germany) at 500 bar for 3 passes. The MFC produced at cycle 10 were also used to stabilize oil-in-water emulsions at varying concentration ranging from 0.00% to 1.00% (w/w) in order to assess the influence of MFC amount on the emulsion stability.

2.5 Characterization of the MFC stabilized emulsion

2.5.1 Particle size

The particle sizes of the emulsions were analyzed using Mastersizer 2000 instrument (Malvern Instruments Ltd, Worcestershire, UK). Emulsions were diluted to around 0.05% (w/w) with distilled water to prevent multiple scattering effects. Subsequently, samples were dispersed in distilled water at 1200 rpm until obscuration rate of 15% was obtained (Laca et al. 2010). The reflective index of dispersant (water) and disperser (emulsion) were set at 1.33 and 1.46, respectively. The particle size parameters were recorded as follow: volume-weighted mean diameter $d_{4,3}$, surface weighted mean diameter $D_{3,2}$ span index-quantification distribution width: (D90 – D10) / D50, D10, D50, and D90 (cumulative volume of particle sizes that make up of emulsion volume of 10%, 50%, and 90%).

2.5.2 Stability

The stability of emulsions under accelerated conditions were tested using optical centrifuge analyzer (Lumifuge 114 LUM GmnH, Berlin, Germany) with 2 mm path

length rectangular sample tubes. Emulsions were centrifuged at 1000 rpm for 127.5 mins (representing 255 cycles) at 3 different temperatures of 5 °C, 25 °C and 45 °C, respectively to evaluate its stability when stored under different temperature conditions. Creaming index were calculated as equation (6):

$$(CI)\% = \frac{final\ position\ of\ bewteen\ clear\ phase\ \&\ creaming\ layer}{Total\ sample\ height}\ x\ 100\text{------}(6)$$

2.5.3 Rheology

The rheological properties of freshly prepared emulsions which include shear viscosity and dynamic frequency sweep were evaluated using rheometer (Rheostress 6000 Haake, Buckinghamshire, Germany) coupled with 60 mm plate to plate probe. The shear viscosity and dynamic frequency sweep test were set in accordance to Section 2.3.6. Additional thixotropy measurements on the emulsion were also conducted. The degree of thixotropy was assessed based on the area difference of the hysteresis loops. Hysteresis loop was generated from 0 s^{-1} to 300 s^{-1} for 5 mins then immediately returned to 0 s^{-1}.

2.5.4 Morphology

The morphology of the emulsion samples prepared from C2 and C10 were visualized using the JEM-2100F field emission analytical electron microscope (JOEL, Tokyo, Japan). A drop of the emulsion was placed onto a 400-mesh formvar carbon film-coated copper grid and the excess solution was blotted with filter paper. Then, the grid was negatively stained by uranyl acetate (Meena et al. 2012). The excess solution was blot with filter paper. The grid were then dried for 15 mins prior to visualization.

2.6 Statistical analysis

All measurements were performed in triplicates unless stated. Significant differences ($P<0.05$) of the samples were analyzed using one way analysis of variance (ANOVA) by LSD test. Results are expressed as mean values ± standard deviations.

3 Results and discussions

3.1 Corn cob pretreated with endoglucanse

Corn cob fiber was treated with 0 %, 0.02%, 0.1%, 0.5% to 2.5% of Fibercare R. As an endoglucanse enzyme, the Fibercare R enables softening of corn cob fiber that ease its passage through high pressure homogenizer thus preventing the blockage or breakdown

of the equipment. Table 1 shows that endoglucanse treatment from 0.02% to 2.5% on blockage of high pressure homogenizer. Incorporation of the endoglucanase enables the fiber suspension to pass through the high pressure homogenizer smoothly without causing any blockage whilst those without endoglucanase treatment show to block the high pressure homogenizer. Also, endoglucanse pretreatment reduces the degree of polymerization of the fiber. Study shows that the reduction in the degree of polymerization was the smallest when corn cob fiber was treated with 0.02% of endoglucanase whilst a significant (P<0.05) reduction in the degree of polymerization was observed when corn cob was treated with 2.5% of endoglucanse enzyme. It should be highlighted that fiber with low degree of polymerization is undesirable for MFC production as it lead to the formation of microcrystalline cellulose instead.

Table 1: Degree of Polymerization (DP) and frequency of high pressure homogenizer blocked by fiber

Fiber	Total number of HPH blocked within 10 tests	DP
G0.0 (Control)	2	991±4[a]
G0.02	0	934±4[b]
G0.1	0	735±3[c]
G0.5	0	717±5[d]
G2.5	0	688±7[e]

Data were performed in triplicate. Mean±standard deviation values in the same column followed by different letters are significantly different (p<0.05).

3.2 Characteristic of MFC

Since 0.02% of endoglucanse enzyme was sufficient to prevent blockage of high pressure homogenizer while still retain a high degree of polymerization, it was selected to treat the corn con fiber to soften the texture before defibrillation process performed by high pressure homogenizer. However, corn cob fiber suspension treated with 0.02 % of endoglucanse tend to sediment to the bottom instead of dispersing well in the suspension. MFC that is well dispersed is important to act as stabiliser for oil-in-water emulsions. Therefore, high pressure homogenization was used to improve the stability of the MFC suspension. Endgolucanse treated corn cob was defibrillate using high pressure homogenizer from cycle 0 to cycle 10.

3.2.1 Stability

It was found that homogenization successfully transform the MFC from liquid suspension into a gel like suspension. MFC tends to absorb water after undergoing defibrillation process using high pressure homogenizer which leads to the formation of gel like consistency. Stability of MFC gel increased in tandem with the number of cycles of high pressure homogenization and reached a plateau state after homogenizing for 6 cycles. The MFC produced at C10 was the most stable indicated by the highest water holding capacity of 12763% that represents the absorption of 127.63 gram of water per gram of sample.

The water holding capacity properties of MFC was further confirmed by the % of water evaporation or water loss due to evaporation. It was found that the water evaporation from MFC reduced with high homogenization cycles. Without homogenization, water evaporation of MFC was 52.42%. Homogenization for 10 cycle reduced the water evaporation of MFC to 34.90%. A reduction in water evaporation after homogenization could be attributed to the increase in the defibrillation process that breaks the fiber along the lateral direction thus making it become thinner. Defibrillation successfully exposed the hydroxyl side groups of MFC further attract the binding of water molecules to MFC *via* hydrogen bonding. Also, the amount of MFC released from fiber bundle increases after homogenization which restricted the movement of water molecule making it to have gel like texture. The gel like properties of MFC makes it an excellent water stabilizing or wetting and control release agent.

Stability of the MFC was also verified by evaluating its zeta potential. Typically, samples with zeta potential less -30mV or higher than +30mV are considered to be stable as the value was sufficient for the molecules to repel each other preventing them from coalescene. Table 2 shows the zeta potential of the MFC. Zeta potential of MFC increased with HPH cycles. Control without passing through high pressure homogenizer possess zeta potential of ~23.20mV while MFC produced at cycle 10 attained zeta potential of ~30mV. All the MFC have zeta potential value were lower than ~30mV regardless of the number of cycles of homogenization demonstrating that the MFC produced are relatively stable.

Table 2: **Degree of polymerization (DP), water holding capacity (WHC), stability, evaporation and zeta potential of microfibrillated cellulose treated with 0.02% of endoglucanase after undergoing different cycles of homogenization**

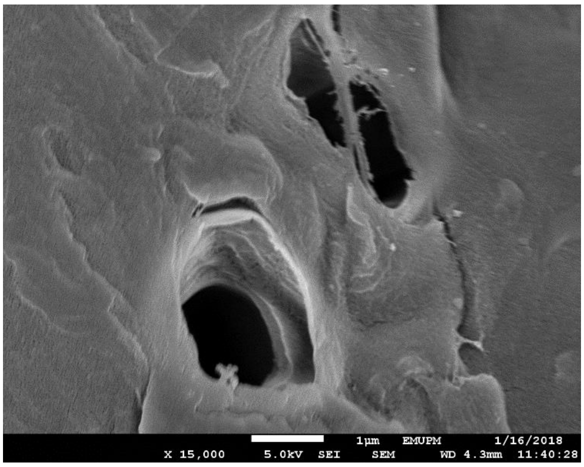
	Analysis performed				
Cycle	DP (unit)	WHC (g/g)	Stability (%)	Evaporation (%)	Zeta potential (mV)
C0	934±4^{a}	36.51±0.21^{a}	18.18±0.01^{a}	52.42±2.66^{a}	-23.20±0.26^{a}
C2	894±5^{b}	80.56±0.11^{b}	40.00±0.20^{a}	46.86±2.88^{ab}	-33.63±0.90^{b}
C4	864±8^{c}	97.56±1.54^{c}	47.95±0.46^{c}	47.09±3.25^{ab}	-36.53±1.11^{b}
C6	838±1^{d}	121.39±3.78^{d}	60.45±0.40^{d}	42.90±1.09^{b}	-36.07±4.50^{b}
C8	738±4^{e}	124.56±1.29^{d}	61.59±0.41^{d}	31.35±2.33^{c}	-30.67±0.32^{b}
C10	729±2^{e}	127.63±0.55^{d}	62.73±0.46^{d}	34.90±5.89^{c}	-36.00±3.14^{b}

Data were performed in triplicate. Mean±standard deviation values in the same column followed by different letters are significantly different ($p<0.05$).

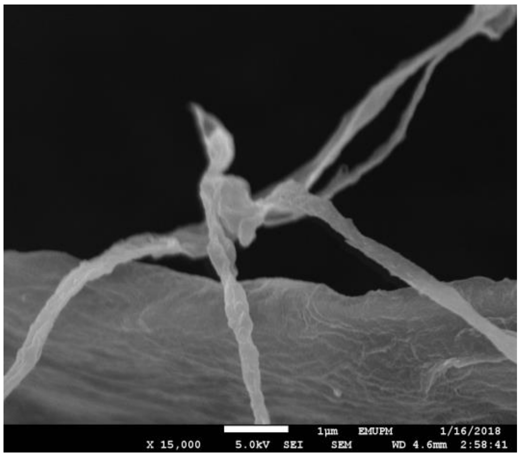
3.2.2 Microstructure

Figure 1 A-F shows the microstructure of MFC observed under SEM. Prior to undergoing homogenization the corn cob fiber retain much of their intact structure (Figure 1A). After high pressure homogenization, corn cob fiber bundle disintegrated into filaments. The filaments entangled into a three dimensional network-like after undergoing homogenization for 6-10 cycles (Figure 1 B -F). Homogenization also reduced the diameter of the MFC. It was estimated that the diameter of MFC observed under 15000x resolution was around 50nm as shown in Figure 1 F.

A



B



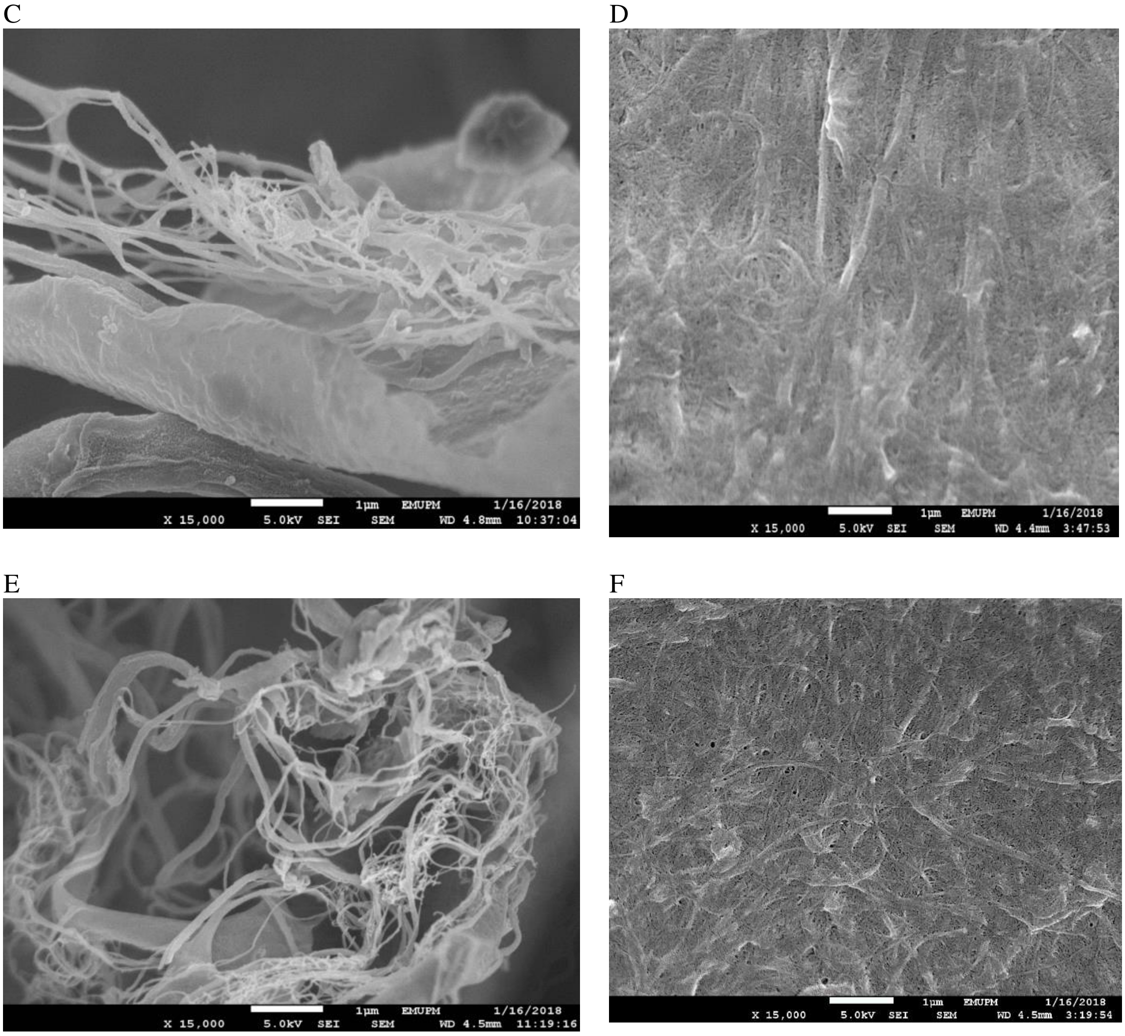


Figure 1: SEM photograph of MFC produced from different cycles of HPH. (A) Pretreated corn cob pulp-cycle 0, (B) cycle 2, (C) cycle 4, (D) cycle 6, (E) cycle 8 and (F) cycle 10

3.2.3 Rheology

Figure 2A shows the shear viscosity of MFC produced from cycle 0 to cycle 10 of homogenization. All the MFC samples showed shear thinning/pseudoplastic behavior. Shear viscosity decreased from around 50 Pa.s to 0.01 Pa.s when the shear rate increased from 0.1 to 300 1/s. C2 had the lowest shear viscosity. Viscosity of MFC increased with high pressure homogenization cycle. Increase in homogenization cycle resulted in disintegration of the fiber and thus releasing more MFC from the fiber bundle. Attributed to the high aspect ratio, MFC as such creates a network-like structure which can resist shear force.

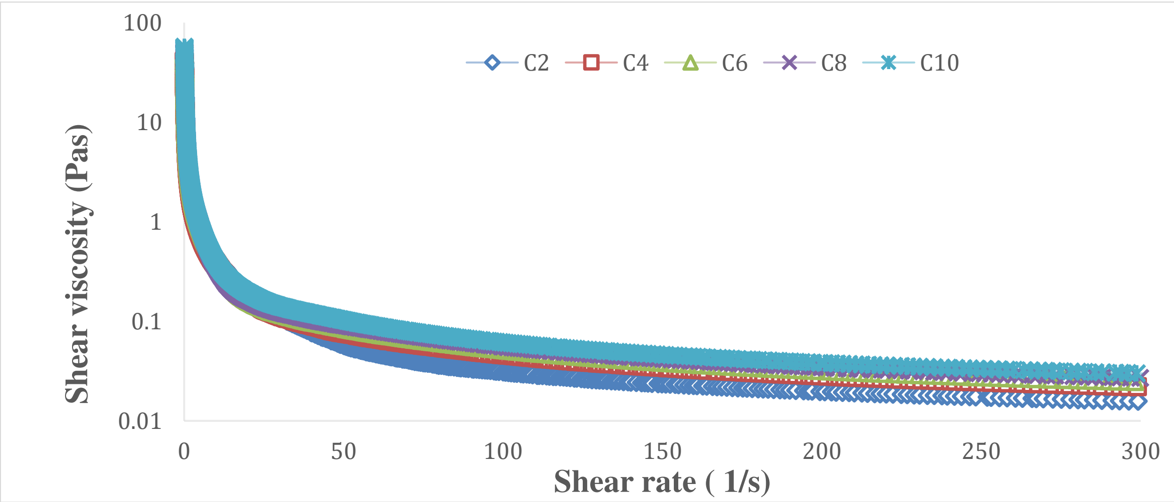
Figure 2B shows the shear viscosity at log scale to assess the flat plateau region at low shear rate used for the estimation of zero shear viscosity marked as X. Zero shear viscosity gave the indication of molecular weight distribution. A substance with a broad molecular weight distribution (MWD) will have shorter zero shear plateau region and vice versa. Figure 2 B illustrates that C4 and C6 had a border molecular weight distribution. It can be inferred that C4 and C6 contain a mixture of different molecular structure of the cellulose either in the form of bundles or cellulose thread which might be due to the partial released of fiber from the fiber bundles (Figure 1 C-D). Meanwhile, C2 and C10 samples had a narrow molecular weight distribution. C2 mainly contained similar large sized fiber bundle (Figure 1 B) whilst C10 has almost all its cellulose converted to MFC (Figure 1 F). Thus, the molecular structure of cellulose in this two situations is homogenous.

Yield stress behavior is an important feature for gel-like or semisolid-like substance. A yield stress is the minimum stress required for a material to initiate to flow. Prior to this, sample usually exhibit elastic deformation or simply solid-like property. A high stress value indicated that more stress needs to be applied for the sample to flow. As shown in Figure 2C, cycle 10 has the highest yield stress while cycle 2 has the lowest yield stress indicating that MFC gel produced at cycle 10 is more stable and stiffer than MFC gel produced at cycle 2.

Figure 3 A-B shows amplitude sweep tests of MFC produced under cycle 0 to cycle 10 of high pressure homogenization: G’ storage modulus (elastic Component) and G’’ loss modulus (viscous component) vs oscillation stress (frequency fixed at 1Hz). Viscoelastic material usually exhibit linear viscous region where G’>G’’. It is independent to stress until the structure breaks down and the material switches to G’<G’’ and losses its linearity. As illustrated from Figure 3A, the linear viscoelastic region progressively increased from C2 to C10. A large linear viscoelastic region in C10 revealed that MFC gel was stiff and have a better stability. A larger linear viscoelastic region is preferred when MFC is used as a stabilizer for oil-in-water emulsion since it will provide better stability and greater coating/thickness to the emulsion.

As for the frequency sweep test, it can be seen that the frequency has only slight influence on the G’ G’’ for all the samples. An unstable sample will tend to sediment when G’’> G’. All the samples exhibited G’>G’’ at low and high frequency regions indicating that the MFC was resistant towards long term instability.

A



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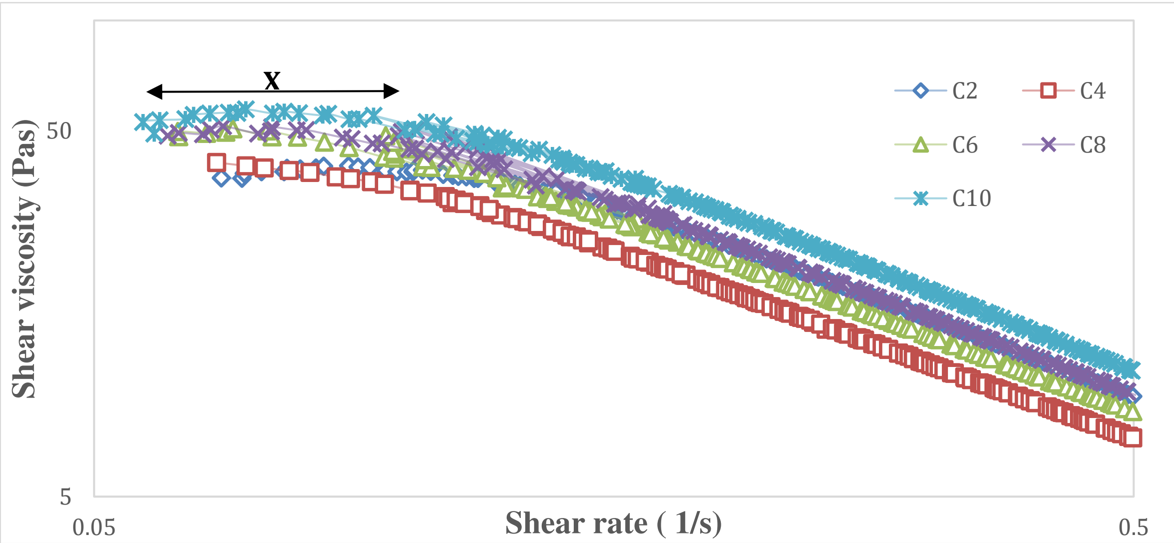


C

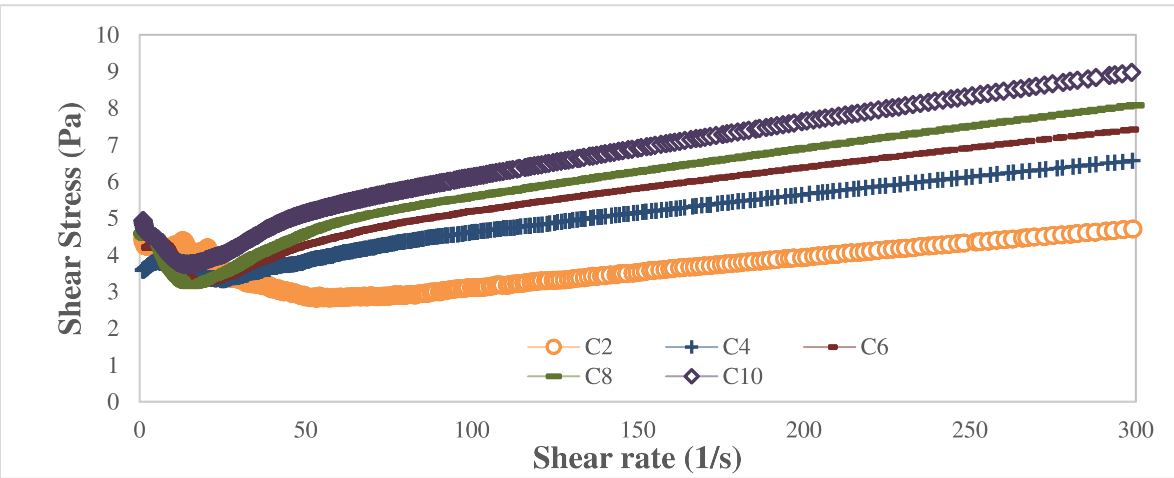
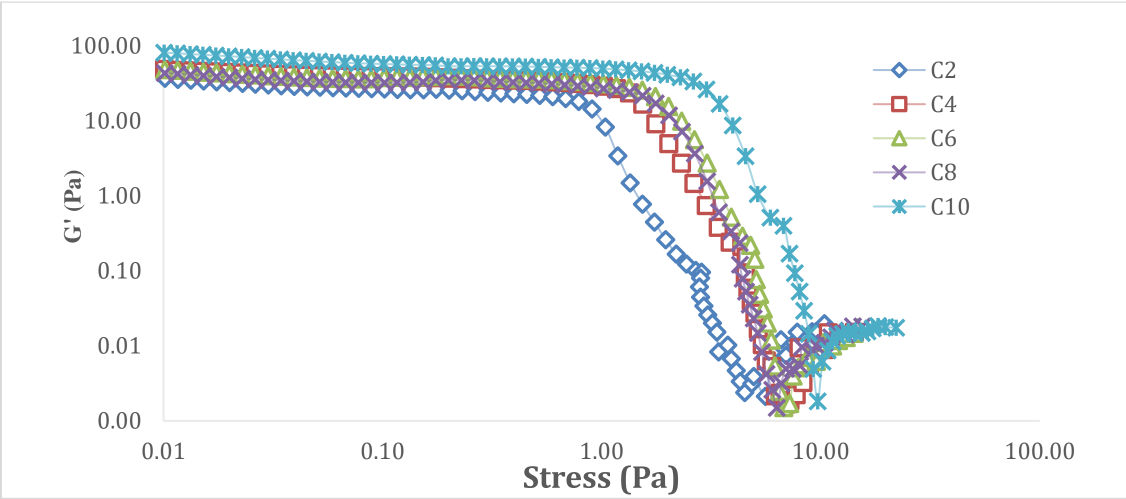


Figure 2: Flow properties of MFC from different cycles of HPH. (A) shear stress as a function of shear rate, (B) shear stress as a function of shear rate log scale (C) Shear stress as function of shear rate. C0 represented cycle 0, C2 represented cycle 2, C4 represented cycle 4, C6 represented cycle 6, C8 represented cycle 8, C10 represented cycle 10

A



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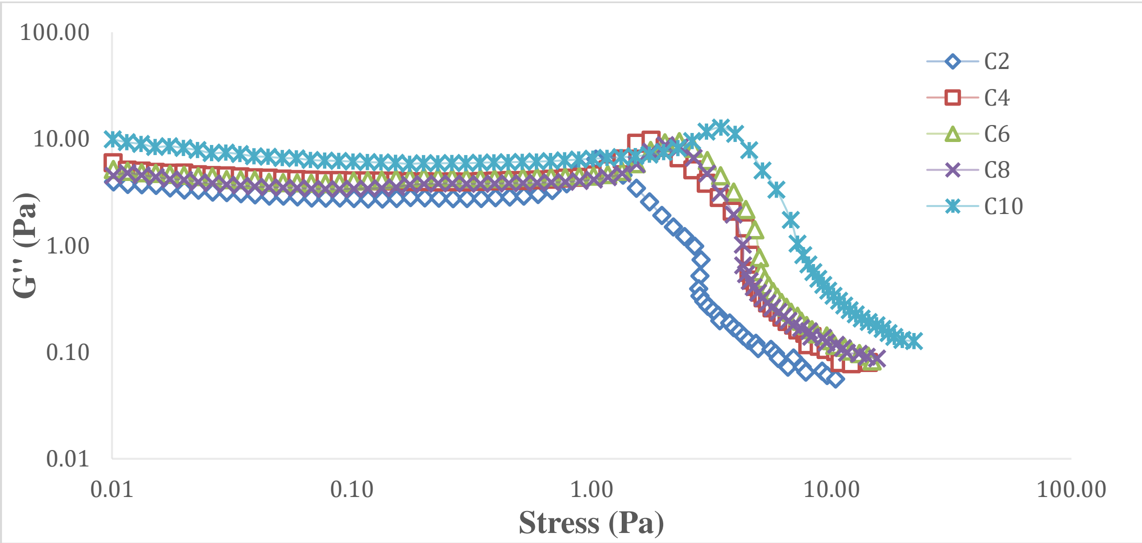

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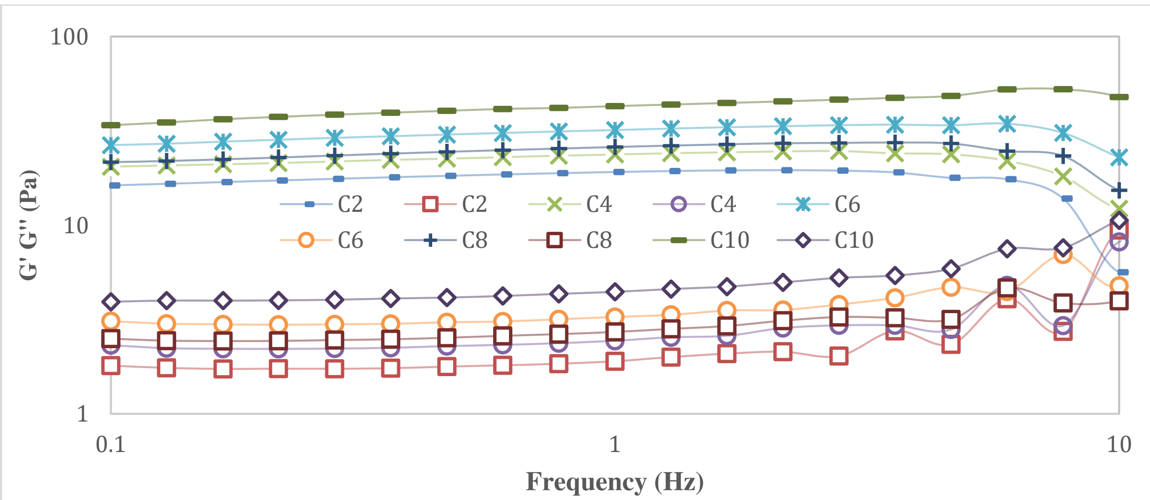


Figure 3: Viscoelastic properties of MFC produced from different cycles of HPH (A) storage modulus G' as a function of stress, (B) loss modulus as a function of stress log scale (C) G' (closed symbol) G''

(open symbol) as function of frequency. C0 represented cycle 0, C2 represented cycle 2, C4 represented cycle 4, C6 represented cycle 6, C8 represented cycle 8, C10 represented cycle 10.

3.3 Emulsion stabilized by MFC prepared from homogenization cycle 0 to cycle 10

MFC produced from cycle 0 to cycle 10 was further utilized to stabilize oil-in-water emulsion and the stability was assessed.

3.3.1 Particle size

Table 3 shows the particle size distribution of emulsion stabilized by MFC produced from cycle 0 to cycle 10. The MFC-stabilized emulsion showed a nearly monomodal particle size distribution (Figure 4). Emulsion stabilized by MFC produced from high pressure homogenizer had mean volume weighted diameter ($d_{4,3}$), surface weighted diameter ($D_{3,2}$), D10, D50 and D90 value that were significantly ($P<0.05$) lower than emulsion prepared from MFC without undergoing homogenization. Homogenization resulted in more MFC to be released from the fiber bundle. Furthermore, homogenization also reduced the size of the MFC. Similar result was also disclosed by Winuprasith et al. (2015) who reported that particle size of MFC-stabilized emulsion reduced when MFC of mangosteen rind from higher cycle of homogenization was used to stabilize the emulsion.

Table 3: Particle size profiles of emulsions produced using MFC from different high pressure homogenization cycles.

	Droplet mean diameter					
HPH cycle	Span index	$d_{4,3}$ (µm)	$d_{3,2}$ (µm)	D (0.1)	D (0.5)	D (0.9)
C0	1.86±0.03^{a}	93.20±10.12^{a}	17.61±1.68^{a}	24.34±3.09^{a}	81.78±7.85^{a}	176.66±20.42^{a}
C2	1.88±0.01^{a}	77.09±7.93^{b}	14.59±1.31^{be}	17.27±2.78^{bc}	68.64±6.38^{b}	146.35±15.66^{b}
C4	1.80v0.03^{b}	76.89±8.50^{b}	14.74±1.51^{be}	19.39±2.99^{c}	69.11±6.64^{b}	143.84±16.94^{b}
C6	1.93±0.02^{c}	73.65±7.96^{b}	13.50±1.37^{bc}	14.00±2.19^{bd}	66.01±6.34^{b}	141.70±15.92^{b}
C8	2.02±0.01^{d}	68.68±5.10^{b}	11.81±0.77^{c}	10.30±1.41^{d}	61.34±3.98^{b}	134.31±10.29^{b}
C10	1.95±0.03^{c}	70.17±4.88^{b}	12.54±0.72^{ce}	12.70±1.67^{d}	62.63±3.54^{b}	135.04±10.27^{b}

The C0 represented emulsion produced use MFC from homogenization cycle 0, so on and so forth. The MFC amount in the emulsion were 0.45%. Test were performed in triplicates. Mean value±standard deviation followed by same letter in each column are not significantly different ($P>0.05$)(span index; $d_{4,3}$:volume weighted mean diameter; $d_{3,2}$: surface weighted mean diameter; D (0.1), D (0.5) and D (0.9) cumulative distribution of particle size at 10%, 50% and 90%, respectively

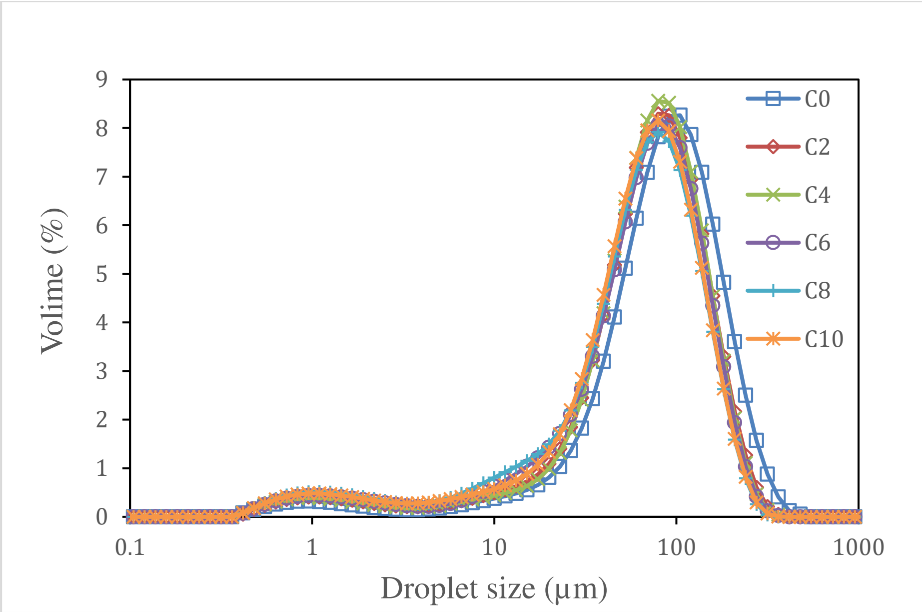


Figure 4: **Particle size distributions of oil-in-water emulsions stabilized by microfibrillated celluloses prepared from different cycles of homogenizer.** (C0 represented cycle 0, C2 represented cycle 2, C4 represented cycle 4, C6 represented cycle 6, C8 represented cycle 8, C10 represented cycle 10)

3.3.2 Accelerated stability

Table 4 shows the accelerated stability of the emulsion stabilized by MFC prepared from cycle 0 to cycle 10 under three different temperatures whereas Figure 5 illustrates the Lumifuge transmission profile of emulsion. CI reduced with the increase in high pressure homogenization cycle. MFC produced at cycle 10 was the most stable where no oiling off was observed showing all emulsions were capable of holding 10% of oil. All emulsions stored under 5 °C had a lower CI as compared to 25 °C and 45 °C.

Table 4: Accelerated creaming index of emulsions prepared from MFC produced by different high pressure homogenization cycles at different temperatures.

Homogenization cycle	Creaming index (%)		
	Temperature (°C)		
	5	25	45
0	$40.76\pm3.92^{a*}$	$39.70\pm2.78^{a*}$	$44.85\pm3.03^{a'}$
2	$36.97\pm2.33^{b*}$	$36.52\pm1.84^{b*}$	$42.42\pm1.84^{a'}$
4	$32.27\pm0.79^{c*}$	$32.73\pm1.14^{cd*'}$	$35.61\pm1.31^{b'}$
6	$30.45\pm1.57^{cd*}$	$35.15\pm0.52^{bc'}$	$37.27\pm1.64^{b'}$
8	$27.73\pm1.20^{d*}$	$33.94\pm1.14^{bc'}$	$35.15\pm0.69^{b'}$
10	$24.55\pm1.20^{e*}$	$30.76\pm0.69^{d'}$	$31.82\pm0.45^{c'}$

The C0 represented emulsion produced using MFC from homogenization cycle 0, so on and so forth. Test were performed in triplicates. Mean value±standard deviation followed by same letter in each column are not significantly different (P>0.05). For pairwise comparison of different temperatures for each individual HPH cycle the same symbol (*’) in each row are not significantly different (P>0.05).

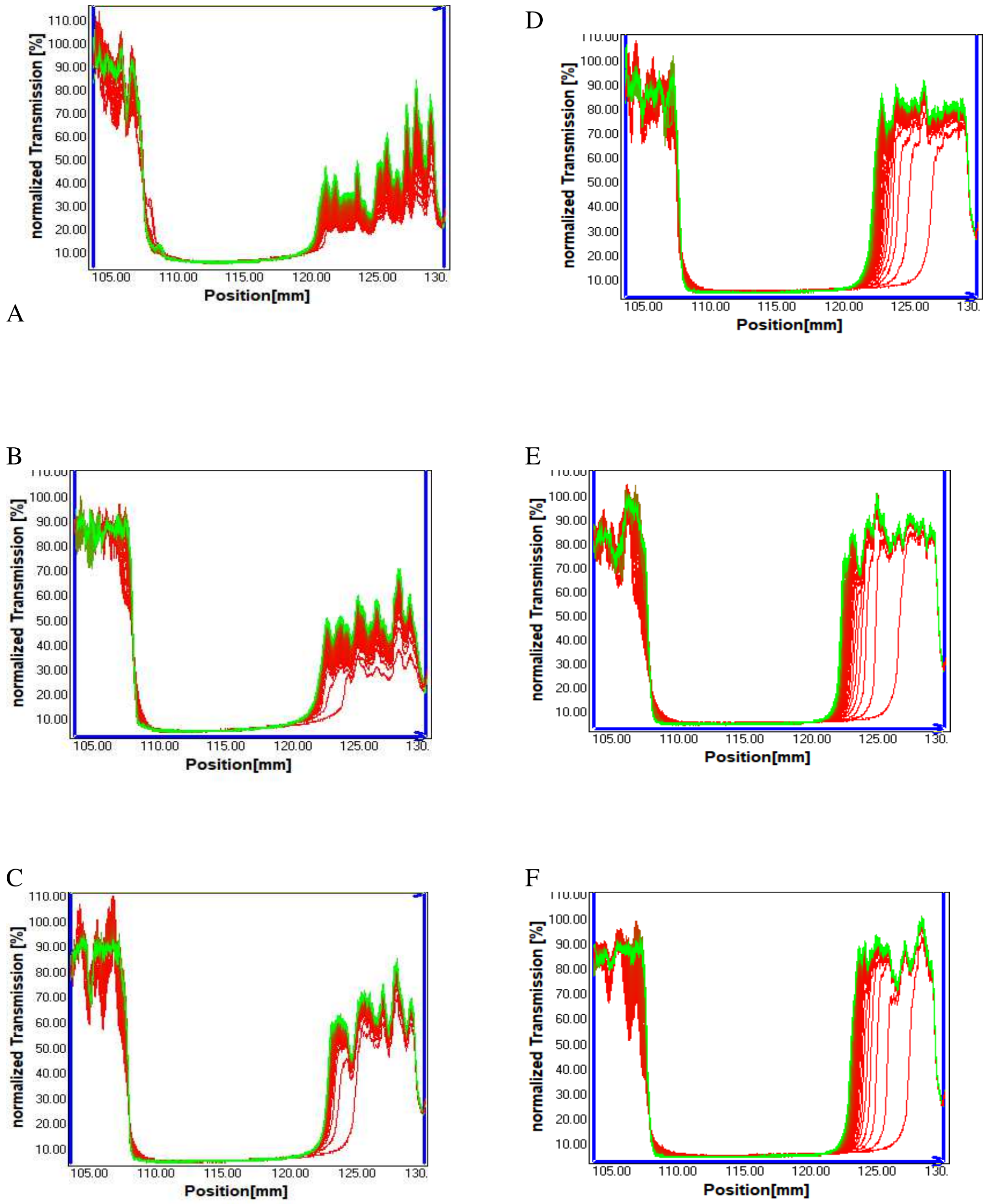


Figure 5: Lumifuge transmission profile over time of emulsions prepared using MFC from different cycles of high pressure homogenization at 25 °C.

(A) cycle 0, (B) cycle 2, (C) cycle 4 (D) cycle 6 (E) cycle 8 (F) cycle 10. Photograph of emulsion using MFC cycle 4 and cycle 10 at 25 °C.

3.3.3 Rheology

3.3.3.1 Flow behavior

Figure 6 A shows the steady flow behavior of emulsions stabilized by MFC produced from cycle 0 to cycle 10. All emulsions prepared from MFC showed shear thinning (pseudoplastic) behavior. Similar observation was found in many emulsion systems that is stabilized by polysaccharides such as chitosan, gum Arabic and microcrystalline cellulose (Burr et al. 2018; Jia et al. 2015). However, the shear thinning effect only reduced slightly when with the cycles of homogenization. This may be due to more MFC being released under high HPH cycles that creates a stronger fiber network causing the emulsion to be more resistant to shear.

Zero shear viscosity is viscosity of sample when shear approaches zero (no shear) that demonstrates the viscosity of sample at stand still condition when no force is being applied. A higher zero shear viscosity value is important in regards to the product stability. An emulsion with higher zero shear viscosity will have higher stability as it restricted the movement of molecules and reduced their overall kinetic energy. Therefore, lesser molecules can overcome the energy barrier to coalesce. The emulsions produced using MFC from cycles 0 to 10 had zero shear viscosity around 3 Pa.s (Figure 5 A). It implies that the emulsion system may have similar stability. Nevertheless, MFC produced from higher cycles performed better in stabilizing emulsion since its overall shear viscosity is the highest.

The degrees of thixotropy of emulsions produced were tabulated in Table 5. A sample is called *thixotropy* (time-dependent shear thinning property) when it is tested with ascending shear rate followed by descending shear rate and create a hysteresis loop. The area of the hysteresis loop is the energy consumed for structural breakdown. Despite significant difference was observed between C2 to C10 emulsions ($P>0.05$), thixotropy of the emulsion gradually increased from 350 to around 450 with exception of emulsions C6. Emulsions made up of more MFC like C8 and C10 is believed to possess more complex network structure that required longer time for structure restoration.

3.3.3.2 Phase angle and G'G'' crossover

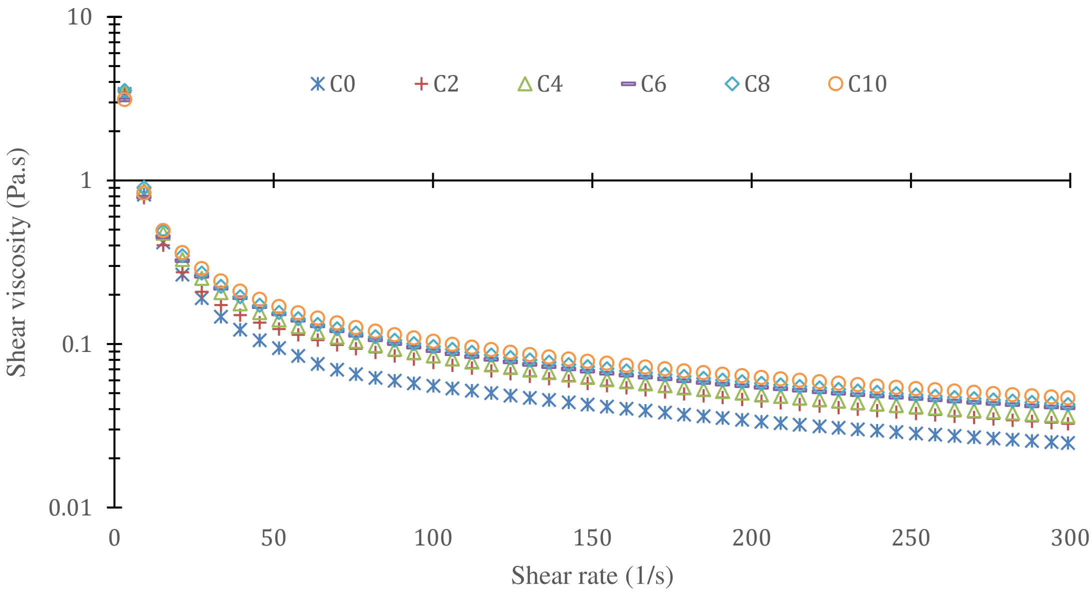
Figure 6 B illustrates the phase angle distributions of emulsions stabilized by MFC produced from cycle 0 to cycle 10. Phase angle provide information related to whether

sample deform elastically like a solid in low phase angle (0°) or flow like a liquid at high phase angle (90°). All emulsions had a plateau phase angle around 8° showing that the emulsions have intact elastic-like structure at low stress (amplitude). However, when the stress forces increased, the phase angle shifted to the region that belongs to liquid dominant (90°).

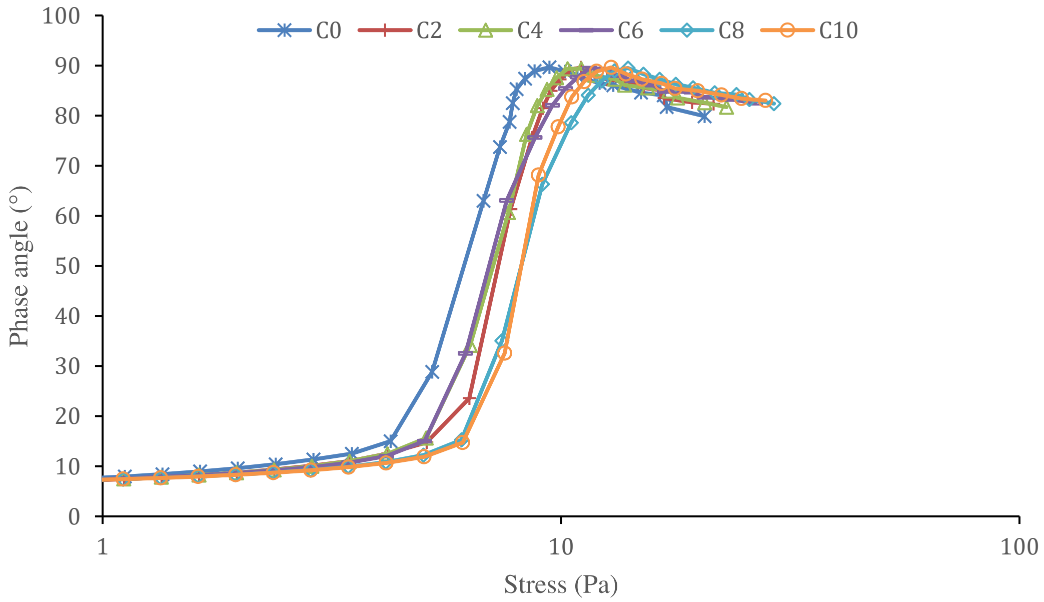
Table 5 shows the as single point yield stress. A yield is when a certain force is applied to a certain extent that exceeded a threshold that caused a gel or viscoelastic solid start to flow. It should be noted that yield is a process. The transition of solid to liquid occur (G'G'' crossover) is interpolated by the phase angle 45° (Shih et al. 1999). The crossover occurred at a higher Pa corresponding to MFC produced at C10 which is 7.83Pa. Under high HPH cycles, more MFC are released in the fiber suspension to create a strong 3D network for supporting the emulsion system. Therefore, a high yield stress (force) is required to disrupt the network in order to collapse the emulsion system allowing it to flow like a liquid. This property is applicable in many products like mayonnaise which stand still like a solid when no external forces were applied, but starting to flow like a liquid when pressure was applied. Hence, yield stress for a product should be conditional not absolute.

As for the frequency sweep test, the storage modulus G' for all emulsions were always higher than loss modulus G'' independent to the frequency (Figure 6 C-D). It showed that the emulsions possesses gel-like behavior. The magnitude of G'G'' increased with HPH cycles demonstrating that the gelation properties were greatly influenced by the HPH cycle. Emulsion with a greater gelation properties can be obtained if MFC from higher HPH cycles were used.

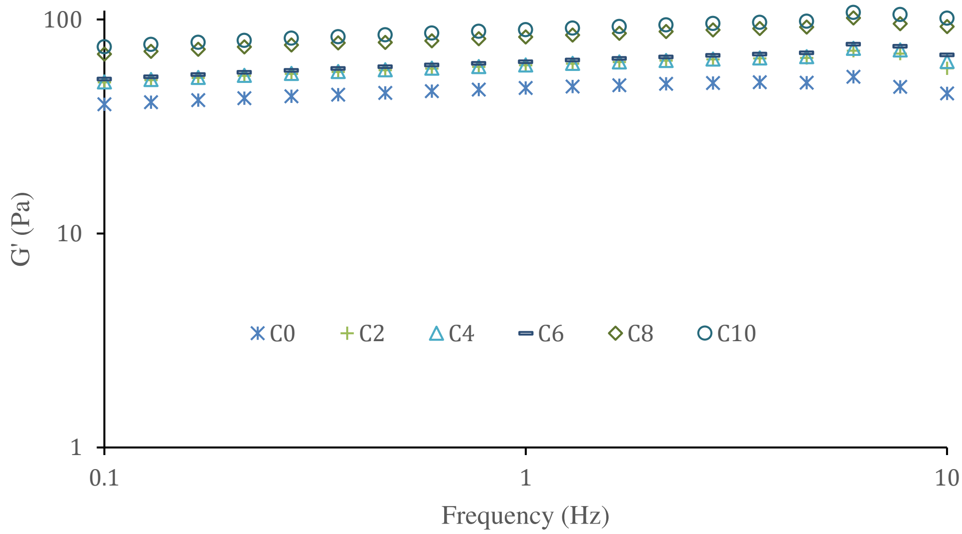
A


C0
C2
C4
C6
C8
C10
Shear viscosity (Pa.s)
10
1
0.1
0.01
0
50
100
150
200
250
300
Shear rate (1/s)

B


C0
C2
C4
C6
C8
C10
Phase angle (°)
100
90
80
70
60
50
40
30
20
10
0
1
10
100
Stress (Pa)

C



D

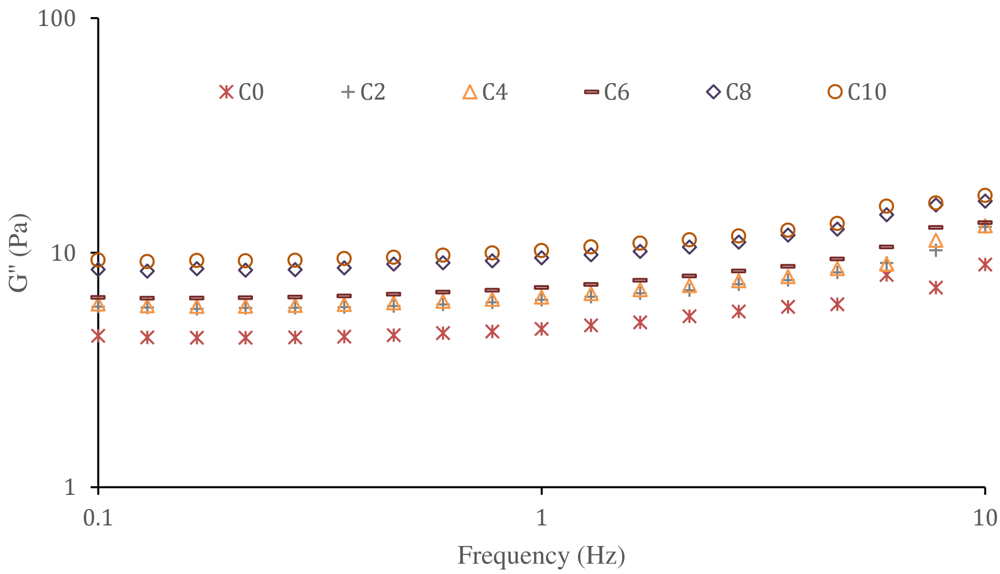


Figure 6: Rheological behavior of emulsions produced using different cycles of microfibrillated cellulose. (A) shear stress as a function of shear rate, (B) phase angle as a function shear stress (C) storage modulus G' and (D) loss modulus G'' as a function of angular frequency measured at temperature of 25 °C. The C0 represented emulsion produced using MFC from homogenization cycle 0, so on and so forth. The final MFC amount in the emulsions were 0.45%.

Table 5: Thixotropy and G'G'' crossover of emulsion stabilized by using MFC produced from different cycles of high pressure homogenization

Homogenization cycle of MFC	Hysteresis area (Pa/s)	G'G" crossover (Pa)
C0	350.47±13.45^{a}	6.03±0.26^{a}
C2	424.87±26.70^{b}	7.15±0.22^{bc}

C4	453.30±7.46^{b}	6.91±0.79^{ab}
C6	420.77±45.42b	6.70±0.41^{ab}
C8	477.93±44.63^{b}	7.85±0.47^{c}
C10	465.63±54.90^{b}	7.83±0.66^{c}

The C0 represented emulsion produced using MFC from homogenization cycle 0, so on and so forth. The final MFC amount in the emulsion were 0.45%. Test were performed in triplicates. Mean value±standard deviation followed by same letter in each column are not significantly different (P>0.05)

3.4 Emulsion stabilized by 0%-1% of MFC

As MFC produced after undergoing 10 cycle of homogenization was the most stable, it was in different amount from 0% to 1% to assess the amount of MFC on emulsion stability.

3.4.1 Particle size

Table 6 shows the of particle size distribution of emulsion stabilized by 0% to 1% of MFC. Volume weighted diameter $d_{4,3}$ of the emulsion significantly increased (P<0.05) with MFC amount. In general, emulsion with smaller particles had higher stability. However, it should be noted that this theory only applies to spherical particle. Hence, MFC-stabilized emulsions might be different since MFC is usually in elongated rod shape in structure. The bimodal model distribution observed in present study showed the possibility that small particle size (~1μm) might be contributed by the oil droplets that is not trapped by MFC while a bigger particle size may be attributed to the emulsion where the oil was entrapped in the three dimensional network of MFC (Figure 7). Hence, it proved that a stable MFC-stabilized emulsion will have a larger particle size $d_{4,3}$.

Table 6: Particle size profiles of emulsions produced using MFC with different final amounts of MFC.

	Droplet mean diameter					
Amoun (%)	Span index	$d_{4,3}$ (μm)	$d_{3,2}$ (μm)	D (0.1)	D (0.5)	D (0.9)
0.00	2.14±0.04^{a}	3.05±0.05^{a}	1.74±0.00^{a}	0.91±0.00^{a}	2.05±0.01^{a}	5.29±0.07^{a}
0.25	2.50±0.02^{b}	27.18±0.81^{b}	6.09±0.11^{b}	1.86±0.04^{a}	22.52±0.57^{b}	58.15±1.83^{b}
0.50	1.91±0.03^{b}	71.50±4.14^{c}	13.15±0.59^{c}	13.25±1.04^{b}	64.53±3.04^{c}	136.61±8.57^{c}
0.75	2.03±0.03^{c}	73.96±3.01^{cd}	12.48±0.32^{d}	10.84±0.52^{b}	65.54±2.03^{cd}	143.77±6.66^{cd}
1.00	1.98±0.03^{c}	77.81±2.71^{d}	14.21±0.16^{e}	15.48±0.32^{b}	68.55±1.87^{d}	150.96±6.43^{d}

MFC used were from high pressure homogenization cycle 10. Test were performed in triplicates. Mean value±standard deviation followed by same letter in each column are not significantly different (P>0.05). (span index; $d_{4,3}$: volume weighted mean diameter; $d_{3,2}$: surface weighted mean diameter; D (0.1), D (0.5) and D (0.9) cumulative distribution of particle size at 10%, 50% and 90%, respectively.

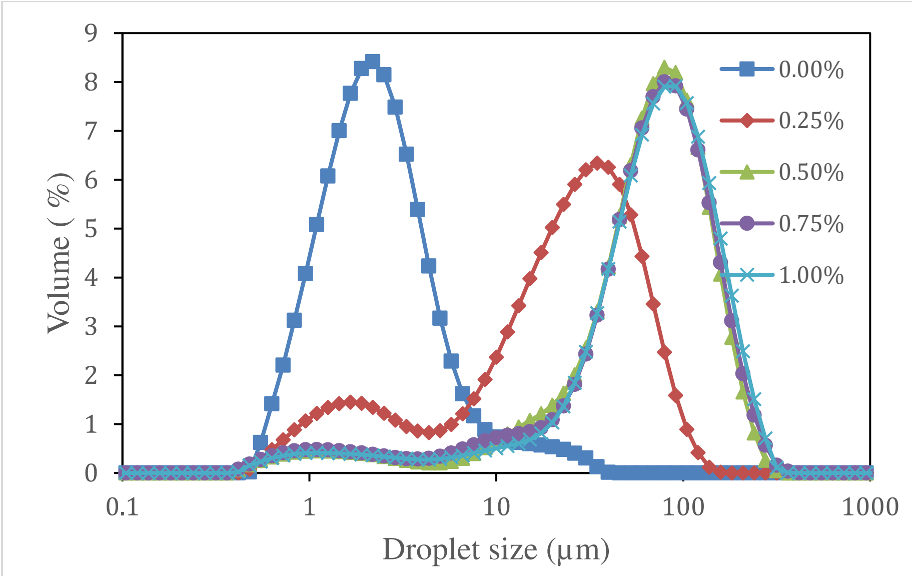


Figure 7: **Particle size distributions of oil-in-water emulsions stabilized by 0%, 0.25%, 0.50%, 0,75%, 1.00 % amount of microfibrillated celluloses**.The final MFC amount in the emulsions were 0.45% in the cycle study. MFC used in the amount study were from high pressure homogenization cycle 10.

3.4.2 Accelerated stability

Table 7 shows the accelerated stability of the emulsions stabilized by 0% to 1% of (w/w) of MFC under different thermal environments. Figure 8 shows the lumifuge profile of the emulsion. Emulsion produced tended to have a high stability and lower creaming behavior when high amount of MFC was used. For instance, CI of MFC emulsions reduced from 57.42% to 2.73% when MFC amount increased from 0.25% to 1.0%. A high amount of MFC was postulated to restrict the movement of oil droplets thereby preventing coalescence.

Table 7: Accelerated Creaming index of emulsions prepared by different amount MFC at different temperatures.

MFC amount (w/w) %	Creaming index (%)		
	Temperature (°C)		
	5	25	45
0.00	21.67±0.95	38.18±0.45	55.45±0.45
0.25	42.12±0.95^{a*}	57.42±0.95$^{a'}$	54.39±0.69$^{a'}$
0.50	24.85±0.69^{b*}	31.21±3.47$^{b'}$	33.18±1.36$^{b'}$
0.75	4.70±1.14^{c*}	12.73±1.20$^{c'}$	15.15±1.60$^{c'}$
1.00	4.55±1.64^{c*}	2.73±0.45^{d*}	3.03±0.26^{d*}

MFC used were from high pressure homogenization cycle 10. Tests were performed in triplicates. Mean value±standard deviation followed by same letter in each column are not significantly different (P>0.05). For pairwise comparison of different temperatures for each individual HPH cycle the same symbol (*') in each row are not significantly different (P>0.05).

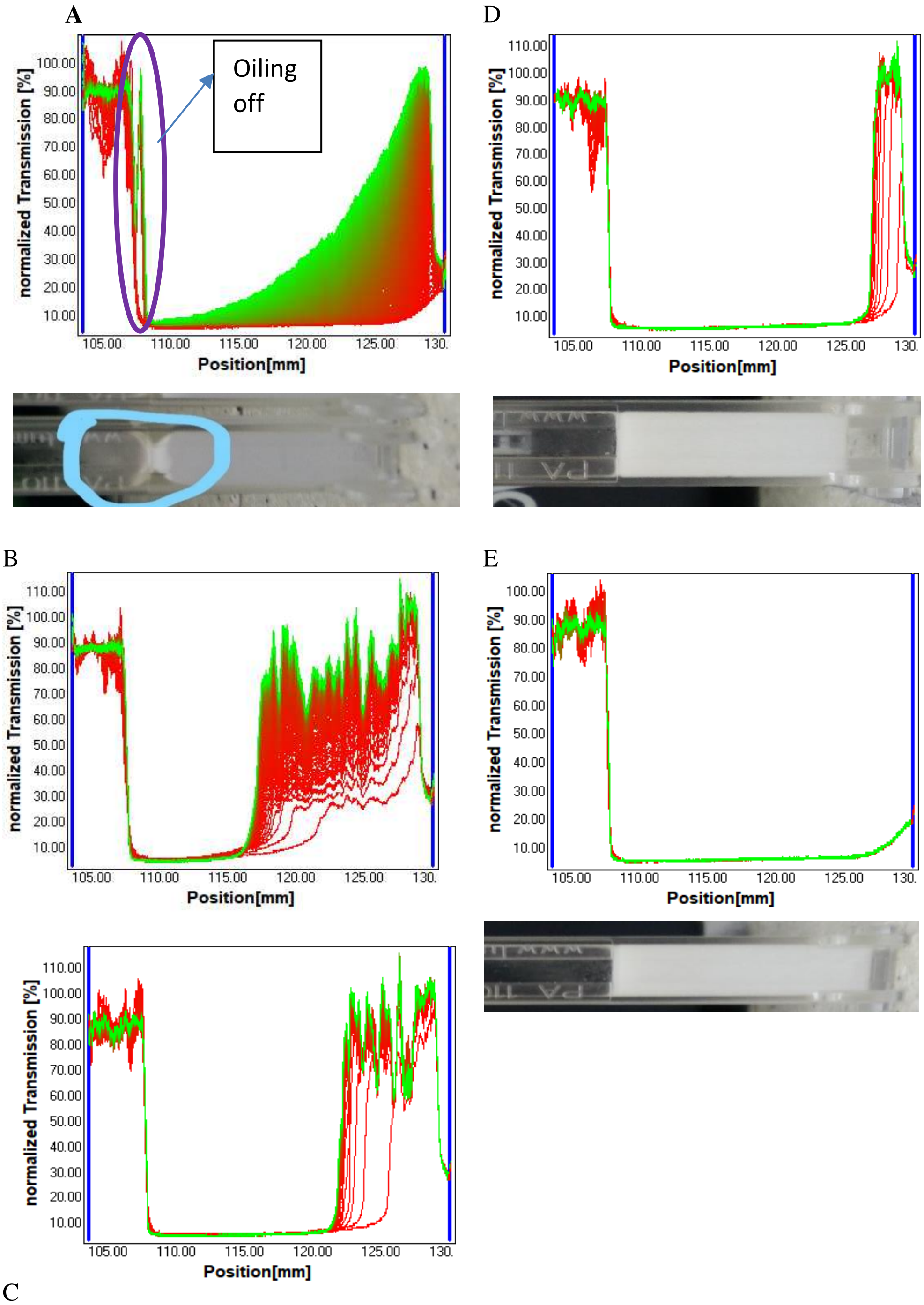


Figure 8: **Lumifuge transmission profile over time for emulsions stabilized prepared with different final amounts of MFC at 25 °C.**
(A) 0.00%, (B) 0.25%, (C) 0.5% (D) 0.75% and (E) 1.00% MFC with (A), (D) and (E) having photo of emulsions containing tubes with its respective final amount of MFC.

3.4.3 Rheology

3.4.3.1 Flow behavior

Figure 9 A shows the steady flow curve of emulsions stabilized by 0%-1% of MFC. Emulsion stabilized by 0.25% MFC possessed the lowest shear viscosity. Shear viscosity increased with the amount of MFC up to 0.75% and plateau at 1% of MFC. A slight

increase in MFC amount increase the apparent viscosity drastically. It was found that the shear thinning behavior was more pronounced at higher concentrations of MFC. A high MFC concentration creates a compact flocculated droplets entrapped by MFC network. Hence, the flocs became more sensitive to shear force when exists in a closed packed arrangement. When shear force is exerted, it causes rearrangement of shape, disintegration of flocs and disruption of MFC networks. As a result, the droplets moved in a parallel position causing the viscosity to reduce (Quemada and Berli 2002). It should be noted that the reduction of shear viscosity for 1.00% MFC stabilized-emulsion was from ~100 at 0 Pa.s to ~0.1 Pa.s at 300 s^{-1} shear rate. However, 1.00% of MFC stabilized emulsion still possesses higher shear viscosity as compared to other emulsions.

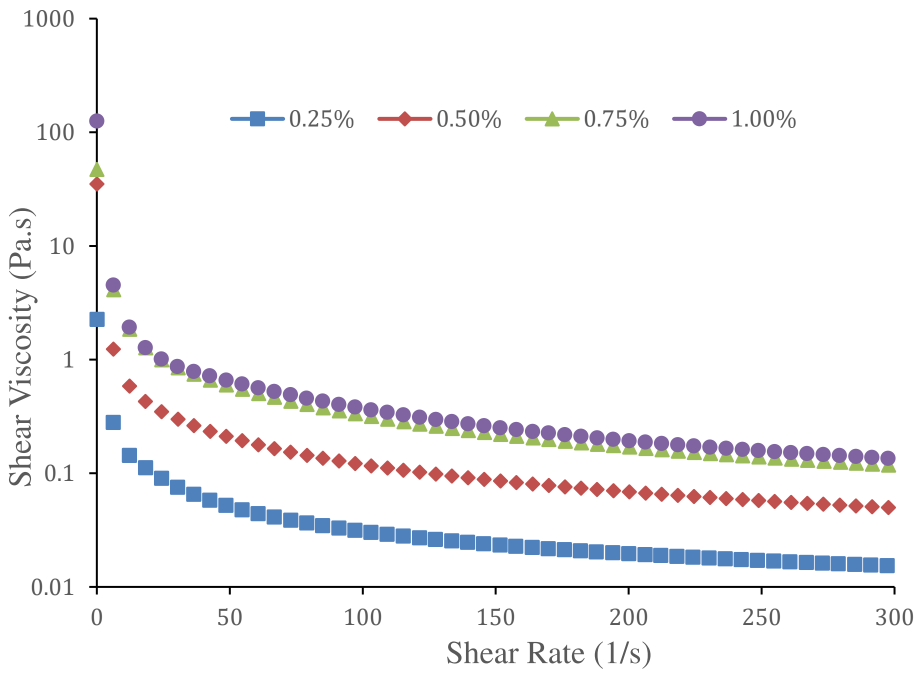
The thixotropy of emulsions stabilized by 0% to 1% of MFC is tabulated in Table 8. The result showed that thixotropy value in emulsion significantly ($p<0.05$) increased from 90.89 (0.25% MFC) to 2657 (1.00% MFC) with a slight increase in the MFC amount. Similar property was also exhibited in many polysaccharide stabilizers due to the presence of large amounts of available binding sites for hydrogen bonding. A slight increase in MFC amount can create more entanglement and binding region between the molecules which further promote the development of a strong network. Hence, it takes a longer time for the structure to recover thereby creating a larger thixotropy areas.

3.4.3.2 Phase angle and G'G'' crossover

The phase angle distribution of the emulsion stabilized by 0%-1% of MFC is shown in Figure 9 B. When the amount of MFC increased from 0.25% to 1.00%, the phase angles plateau area extended significantly ($P<0.05$) from 0.01 Pa to ~10 Pa (Figure 9 B). Emulsion stabilized by 1.00% of MFC had yield stress increased from 5.65Pa to 28.77 Pa, when MFC concentration increases from 0.5% to 1.0%. It implies that 1.00% MFC stabilized emulsion is two times harder to push for it to move/flow then emulsion prepared from 0.5% MFC. A higher yield stress value also indicates a more stable emulsion. Interestingly, unlike the effect of HPH cycle of MFC on the stability of emulsion, the yield stress were not as significant as compared to amount of MFC. A slight increment in the percentage of MFC was able to provide a strong structural network of MFC to stabilize emulsion due to the extremely high water holding capability of MFC (Siró and Plackett 2010).

Figure 9C-D shows the study of frequency sweep test to assess the viscous-elastic behavior of the material. From Figure 5 C, it was observed that emulsion with higher amounts of MFC had a larger G’ values. A larger G’ value from the frequency sweep test indicates a stronger of MFC network present in the emulsion and therefore less susceptible to deformation without losing its elasticity. For a strong gel, the G’ and G’’ are independent of the test frequency, and the G’ was always higher than the G’’. The emulsion that stabilized by high amount of MFC which was 1.00% and 0.75% exhibited a longer frequency independency of G’G” as compared to the emulsion having a lower concentration of MFC. It implies that 1.00% and 0.75% of MFC-stabilized emulsion possessed better gelling properties. The emulsion was able to sustain similar magnitude of disturbance (fixed amplitude) for many times (Hz) and still having a good restoration property (G’ and G’ ’value were not influenced).

A



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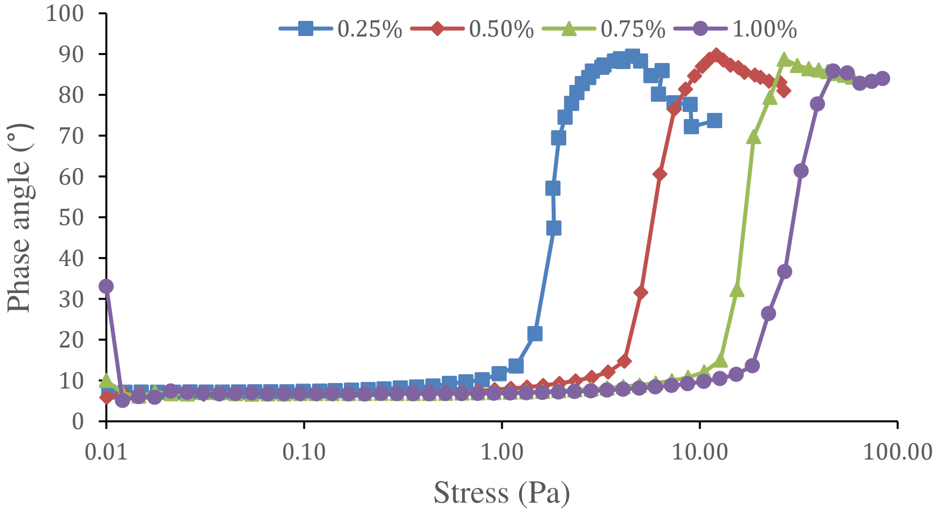

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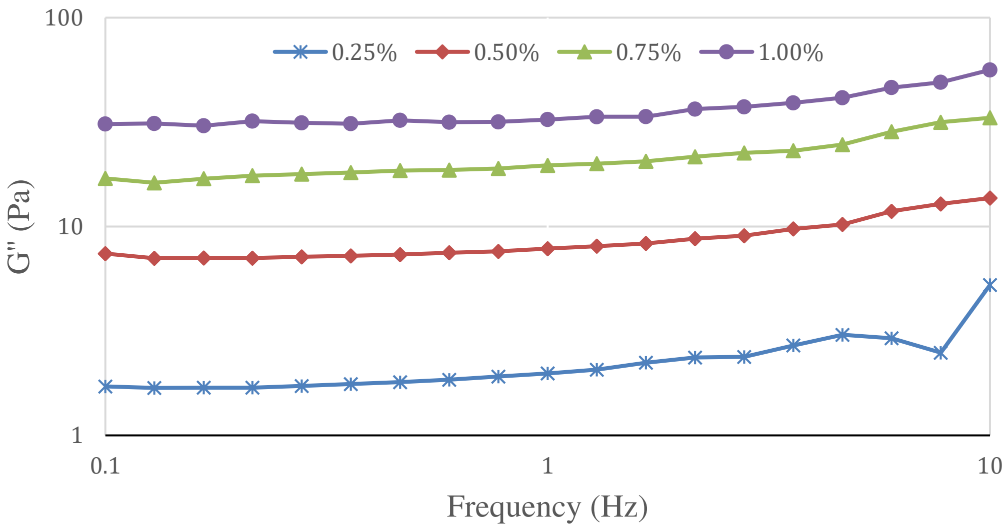

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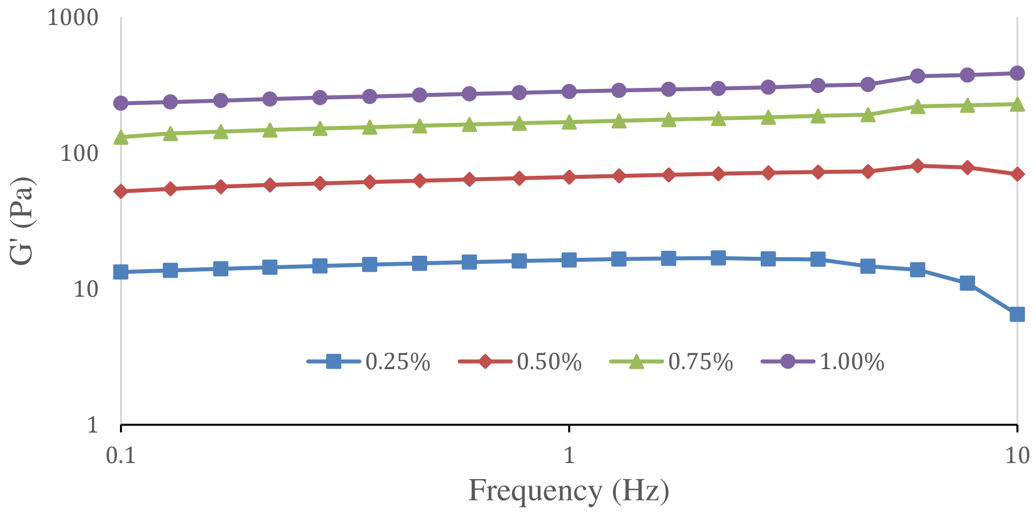

Figure 9 Effect of different amounts of microfibrillated cellulose on the rheological behavior of emulsions. The amount of MFC refers to the final amount of MFC in the emulsion and the MFC used were cycle 10.

(A) shear stress as a function of shear rate, (B) phase angle as a function shear stress (C) storage modulus G' and (D) loss modulus G'' as a function of angular frequency measured at temperature of 25 °C.

Table 8: Thixotropy and G'G'' crossover of emulsion stabilized by using different amounts of MFC.

MFC amount (%, w/w)	Hysteresis area (Pa/s)	G'G" crossover (Pa)
0.00	-	-
0.25	90.89±6.19[a]	1.87±0.40[a]
0.50	569.60±26.55[a]	5.65±0.68[a]
0.75	2299.67±54.45[b]	16.46±0.34[b]
1.00	2657.00±550.90[b]	28.76±6.22[c]

The amount of MFC refers to the final amount of MFC in the emulsion and the MFC used were cycle 10. Tests were performed in triplicates. Mean value±standard deviation followed by same letter in each column are not significantly different (P>0.05)

3.5 Morphologies of the MFC-stabilized emulsions

Figure 10 illustrates the photomicrographs of MFC-stabilized emulsions prepared using HPH at cycle 2 and cycle 10 viewed under field emission analytical electron microscope (FEEM). The MFC network structure showed to possess thread-like network structure whereas oil droplets was observed as whitish spot with irregular shapes. The images confirmed the entrapment of oil droplet in the MFC network. TEM also revealed that the MFC had diameter in in nanosize range and length that is estimated up to several microns in length.

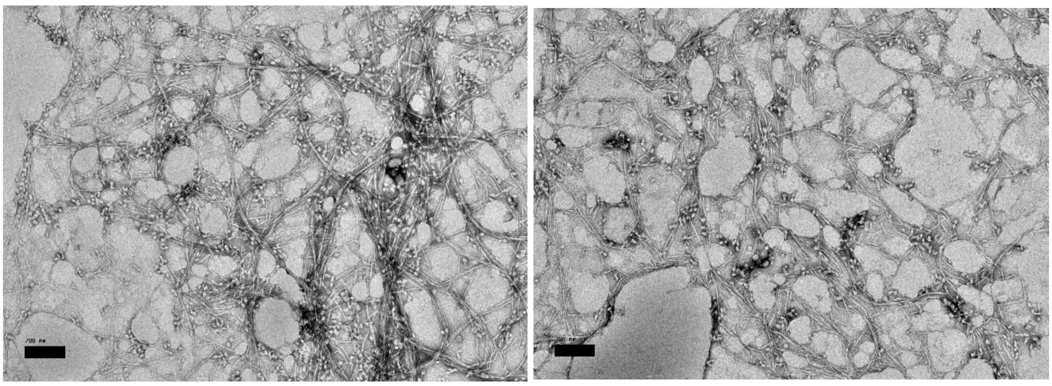

Figure 10: **Morphologies of the microfibrillated cellulose-stabilized oil-in-water emulsions prepared using MFC produced from high pressure homogenization cycle 2 (left) and 10 (right) when observed under field emission analytical electron microscope with magnification power of 10000x.**

4. Conclusion

Underutilized corn cob can be converted into MFC using high pressure homogenization aided with endoglucanse Fibercare R enzyme. Addition of 0.02% of enzyme

endoglucanse is sufficient to facilitate the softening of fiber and prevent blockage of high pressure homogenizer for MFC production. MFC released from the fiber bundle increased with the number of cycles of homogenization. MFC showed to possess gel like consistency with high water holding capacity. MFC was able to stabilize 10% oil-in-water emulsion owning to its ability to form three dimensional network structures that can restrict the movement of emulsion droplet. Emulsion stabilized by MFC produced at high cycle of homogenization had a smaller particle sizes and better stability than those produced at low cycle of homogenization. Similarly, a higher amount of MFC resulted in a more stable emulsion even though the emulsion droplet size is bigger due to interconnected fibrous network. All emulsions stabilized by MFC was relatively more stable than those without MFC. The gel like property of the MFC is important in food industry to act as stabilizer for food emulsion. It allows food manufacturers to manipulate the viscosity of the food products particularly for high fat food products such as mayonnaise which commonly need a high percentage of oil to achieve the desirable apparent viscosity. Incorporation of MFC not only can lower the amount of oil used but also and help to maintain viscosity of the products. This approach is important particularly in producing low calorie product that is deemed to be much healthier.

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Ethics Declaration (Conflict of Interest)

The authors decale that they have no conflicts of interest.

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Figures

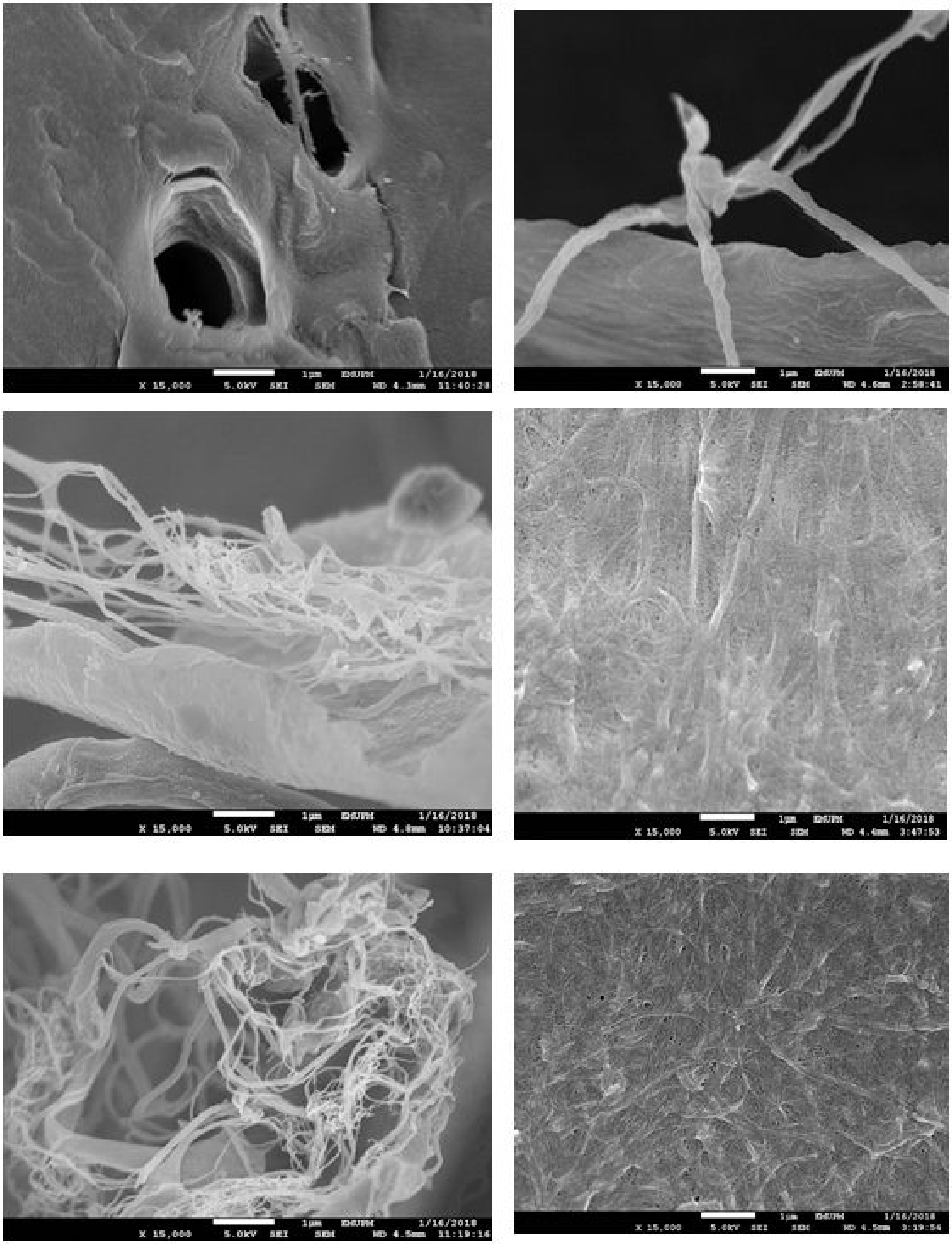

Figure 1

SEM photograph of MFC produced from different cycles of HPH. (A) Pretreated corn cob pulp-cycle 0, (B) cycle 2, (C) cycle 4, (D) cycle 6, (E) cycle 8 and (F) cycle 10

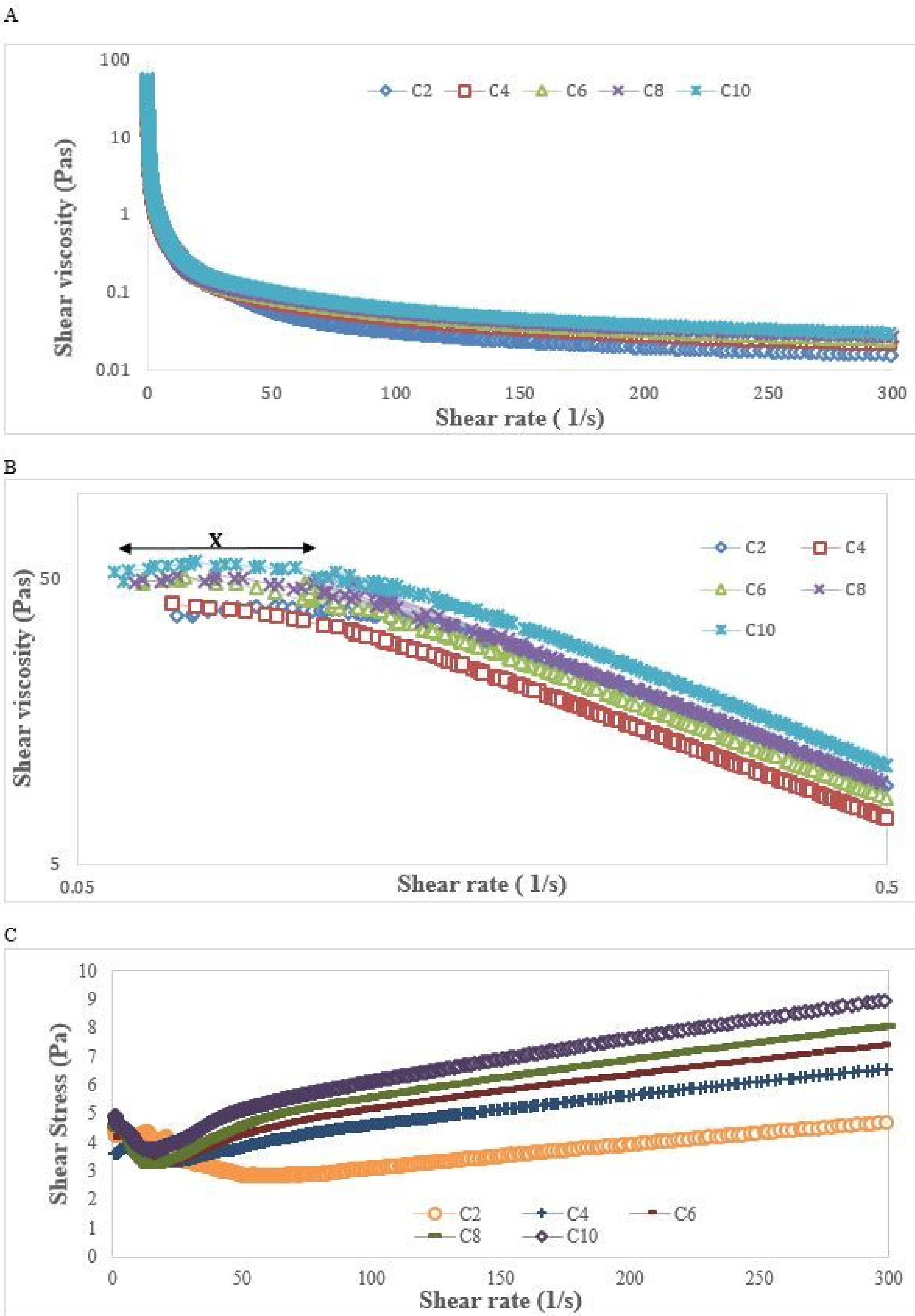

Figure 2

Flow properties of MFC from different cycles of HPH. (A) shear stress as a function of shear rate, (B) shear stress as a function of shear rate log scale (C) Shear stress as function of shear rate. C0 represented cycle 0, C2 represented cycle 2, C4 represented cycle 4, C6 represented cycle 6, C8 represented cycle 8, C10 represented cycle 10

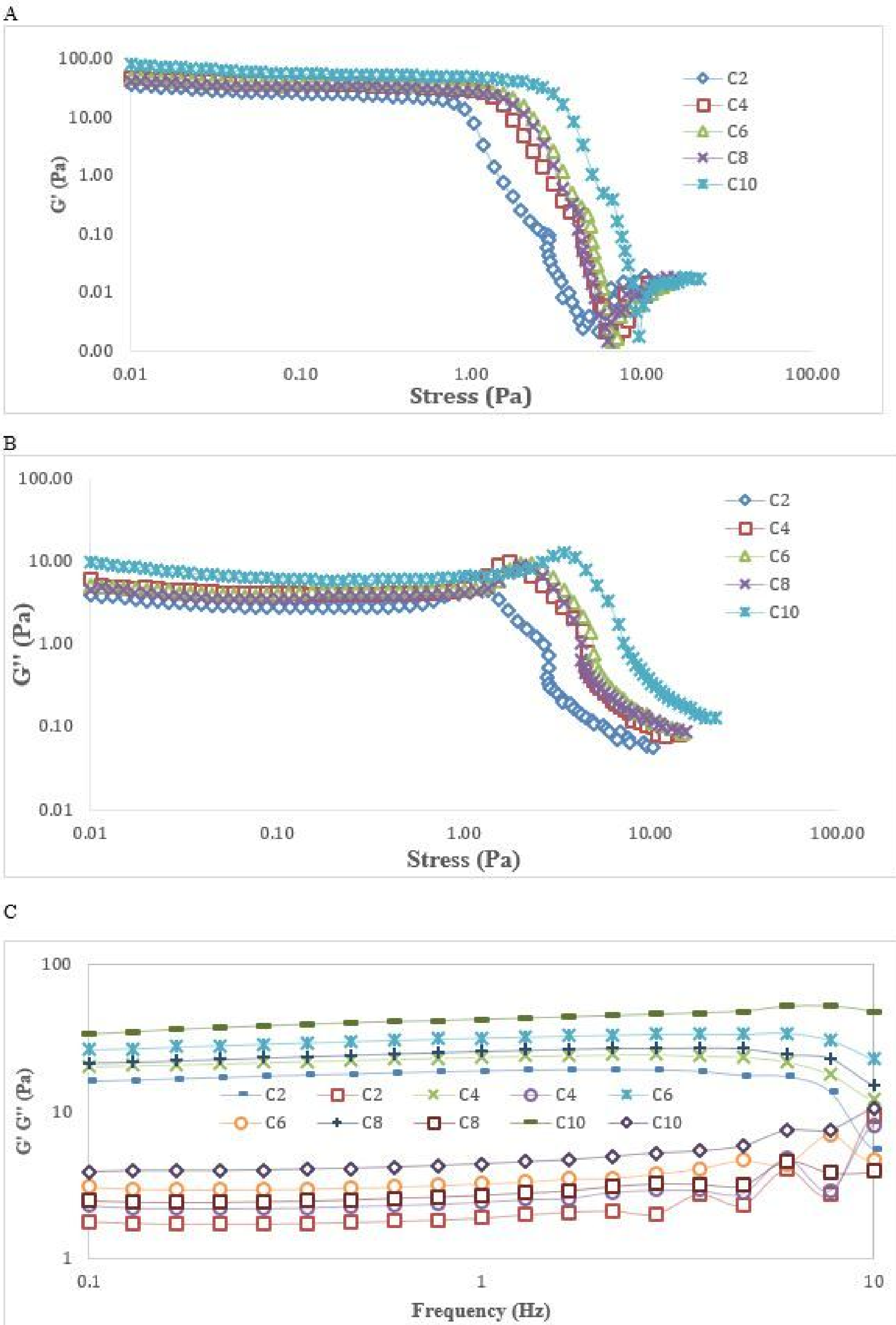

Figure 3

Viscoelastic properties of MFC produced from different cycles of HPH (A) storage modulus G’ as a function of stress, (B) loss modulus as a function of stress log scale (C) G’ (closed symbol) G” (open symbol) as function of frequency. C0 represented cycle 0, C2 represented cycle 2, C4 represented cycle 4, C6 represented cycle 6, C8 represented cycle 8, C10 represented cycle 10.

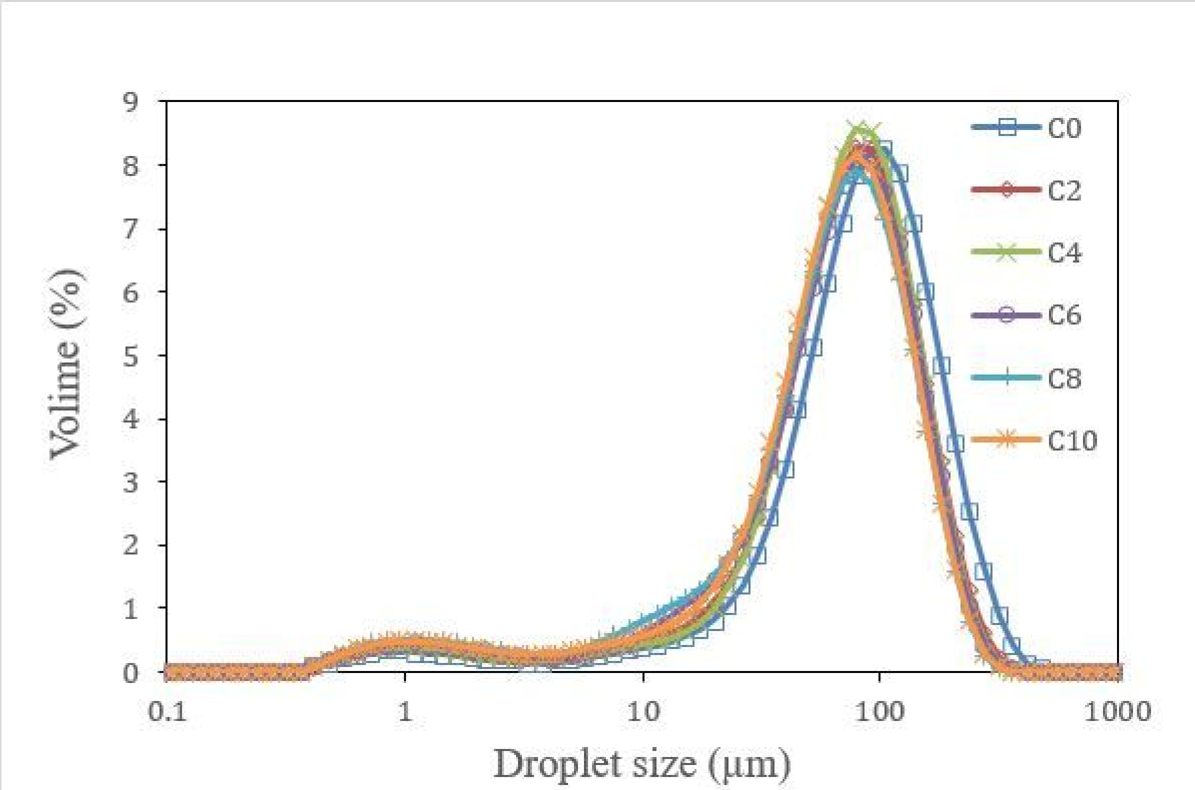

Figure 4

Particle size distributions of oil-in-water emulsions stabilized by microfibrillated celluloses prepared from different cycles of homogenizer. (C0 represented cycle 0, C2 represented cycle 2, C4 represented cycle 4, C6 represented cycle 6, C8 represented cycle 8, C10 represented cycle 10)

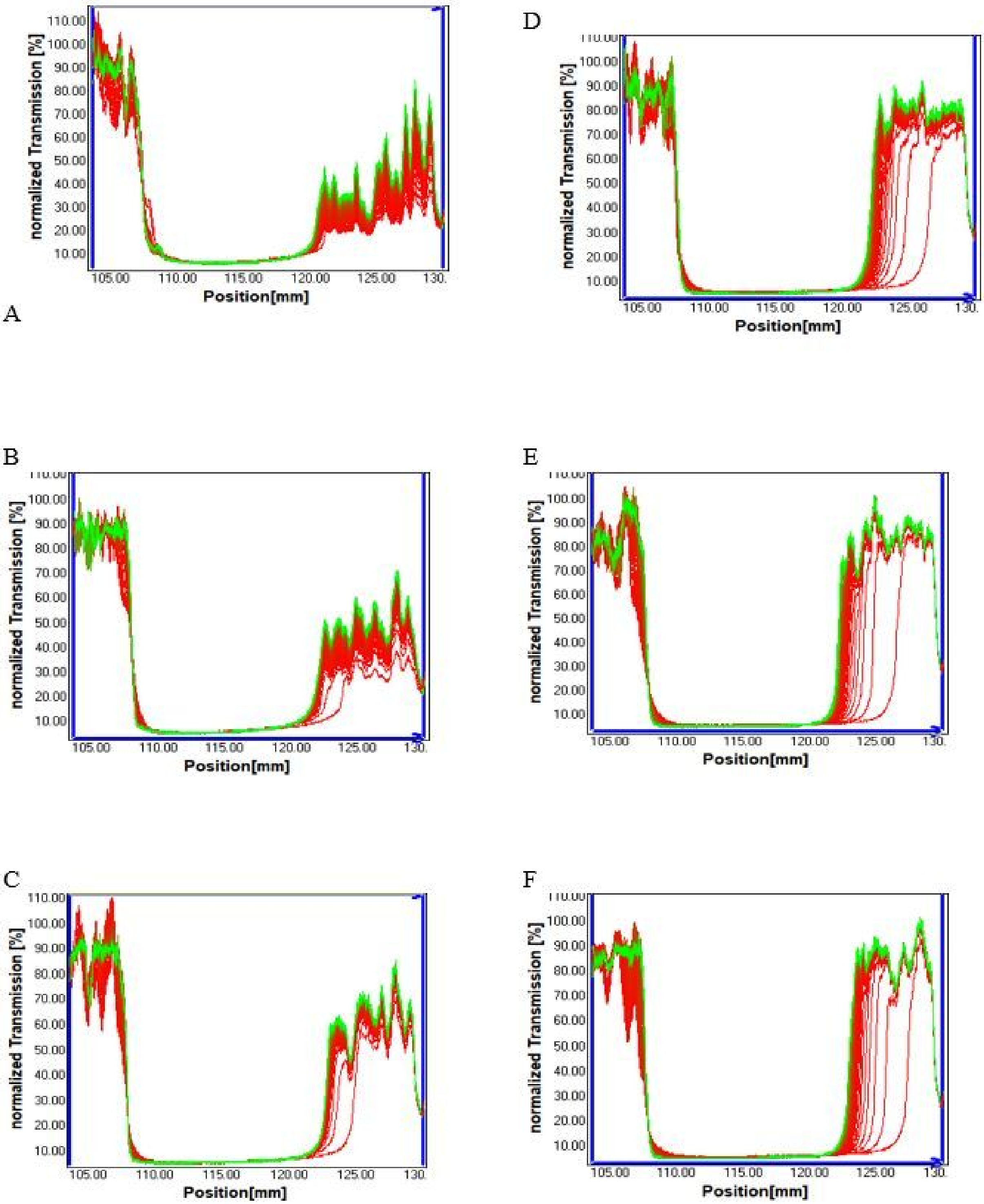

Figure 5

Lumifuge transmission profile over time of emulsions prepared using MFC from different cycles of high pressure homogenization at 25 °C. (A) cycle 0, (B) cycle 2, (C) cycle 4 (D) cycle 6 (E) cycle 8 (F) cycle 10. Photograph of emulsion using MFC cycle 4 and cycle 10 at 25 °C.

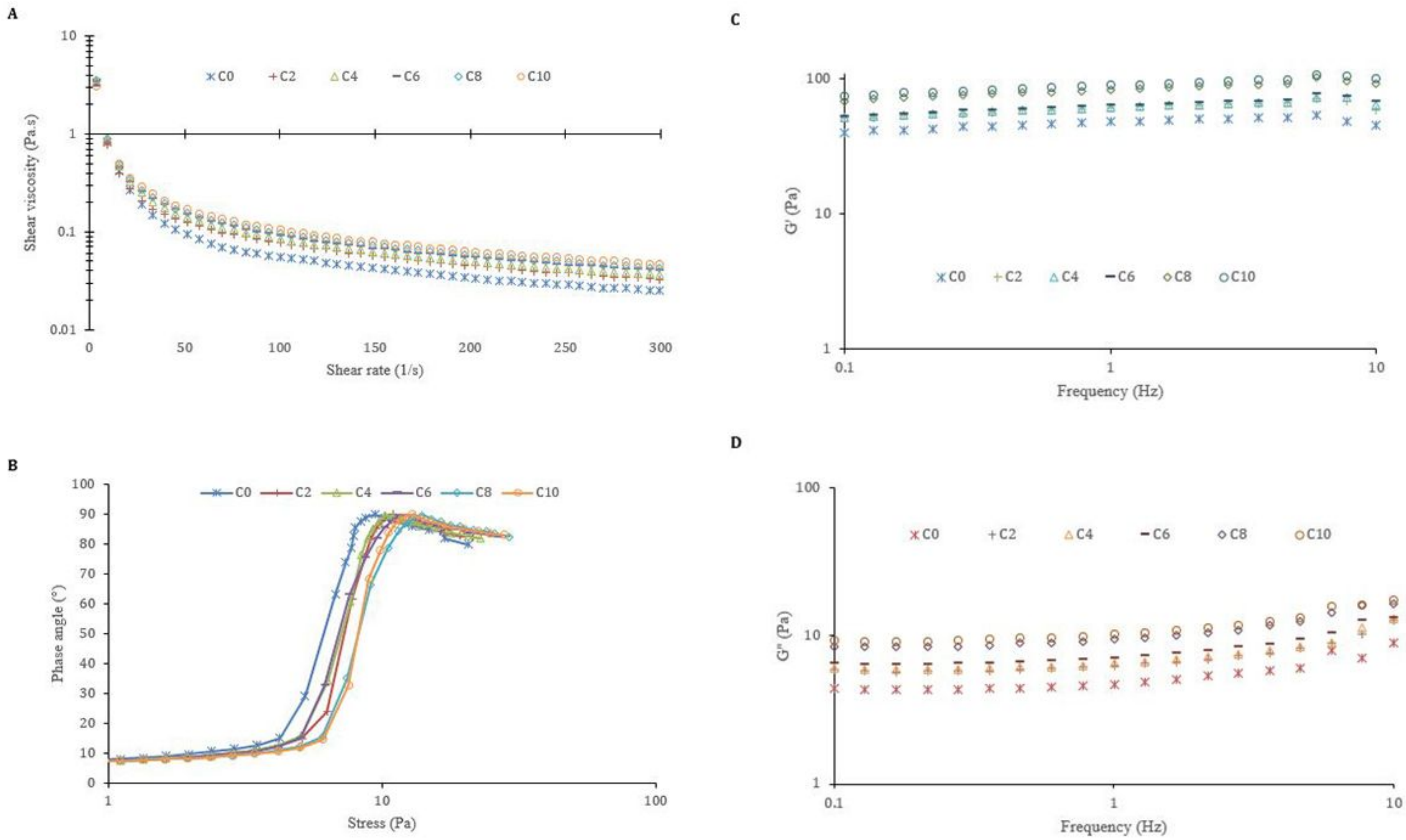

Figure 6

Rheological behavior of emulsions produced using different cycles of microfibrillated cellulose. (A) shear stress as a function of shear rate, (B) phase angle as a function shear stress (C) storage modulus G’ and (D) loss modulus G” as a function of angular frequency measured at temperature of 25 °C. The C0 represented emulsion produced using MFC from homogenization cycle 0, so on and so forth. The final MFC amount in the emulsions were 0.45%.

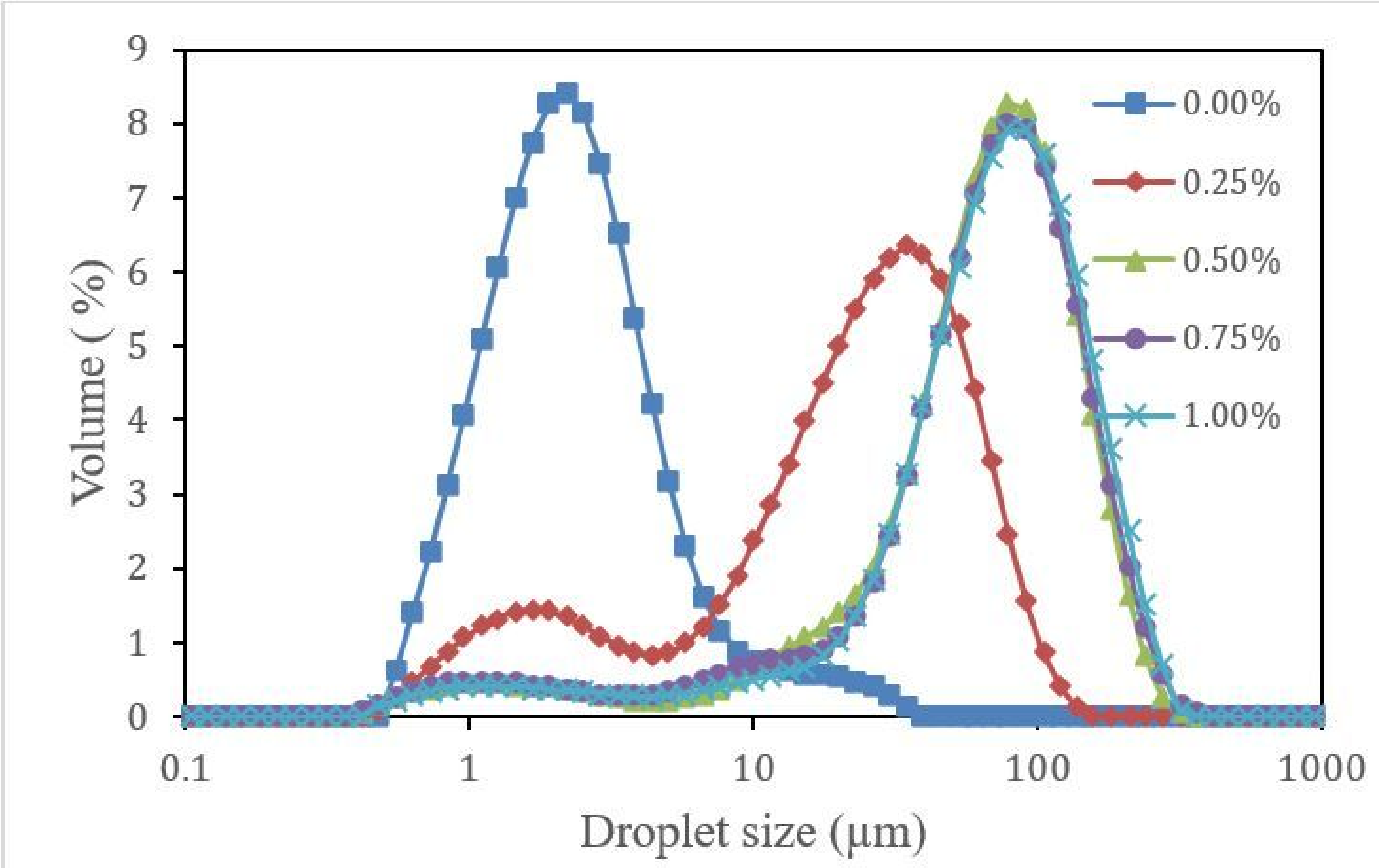

Figure 7

Particle size distributions of oil-in-water emulsions stabilized by 0%, 0.25%, 0.50%, 0,75%, 1.00 % amount of microfibrillated celluloses.The final MFC amount in the emulsions were 0.45% in the cycle study. MFC used in the amount study were from high pressure homogenization cycle 10.

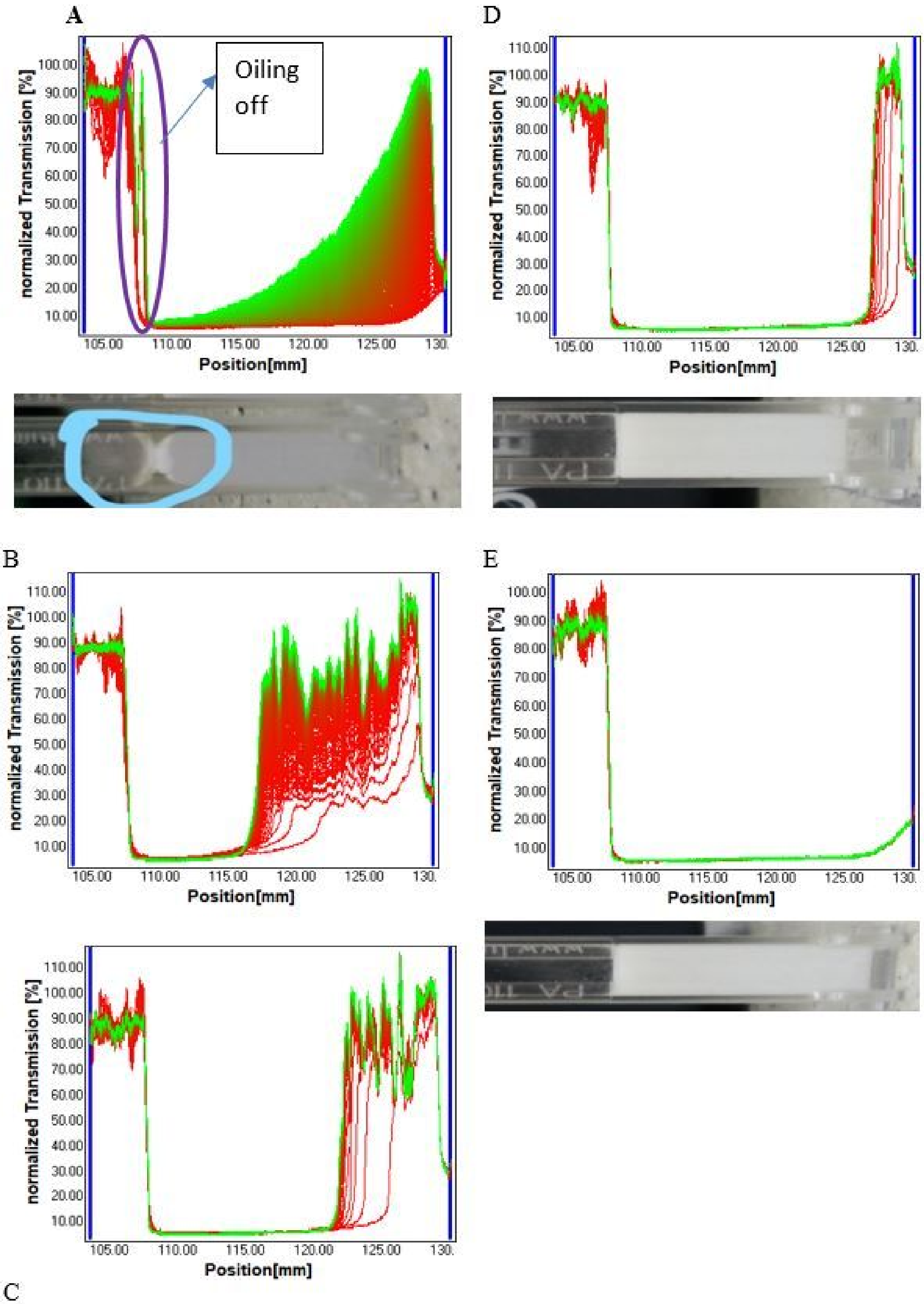

Figure 8

Lumifuge transmission profile over time for emulsions stabilized prepared with different final amounts of MFC at 25 °C. (A) 0.00%, (B) 0.25%, (C) 0.5% (D) 0.75% and (E) 1.00% MFC with (A), (D) and (E) having photo of emulsions containing tubes with its respective final amount of MFC.

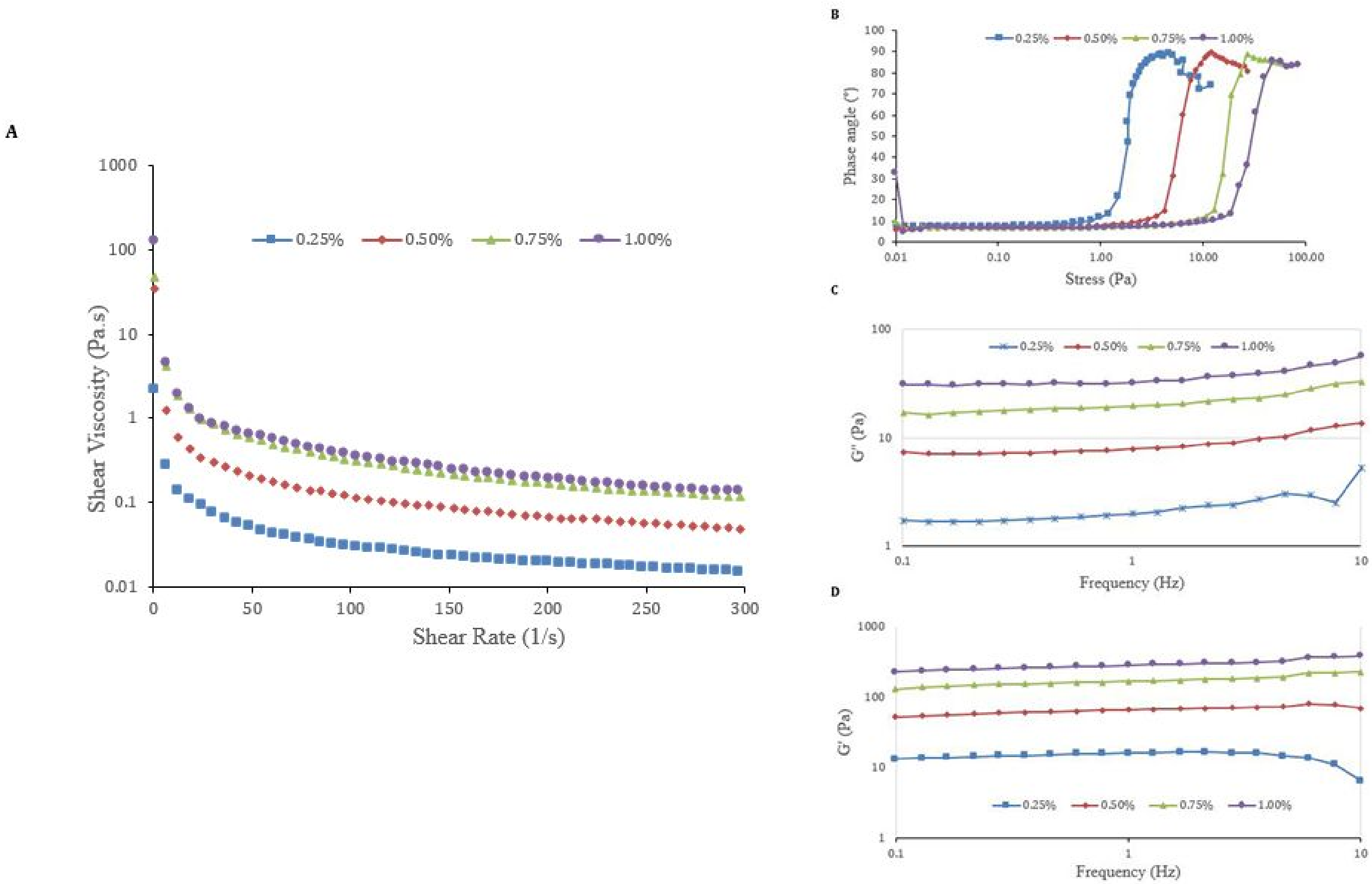

Figure 9

Effect of different amounts of microfibrillated cellulose on the rheological behavior of emulsions. The amount of MFC refers to the final amount of MFC in the emulsion and the MFC used were cycle 10. (A) shear stress as a function of shear rate, (B) phase angle as a function shear stress (C) storage modulus G' and (D) loss modulus G" as a function of angular frequency measured at temperature of 25 °C.

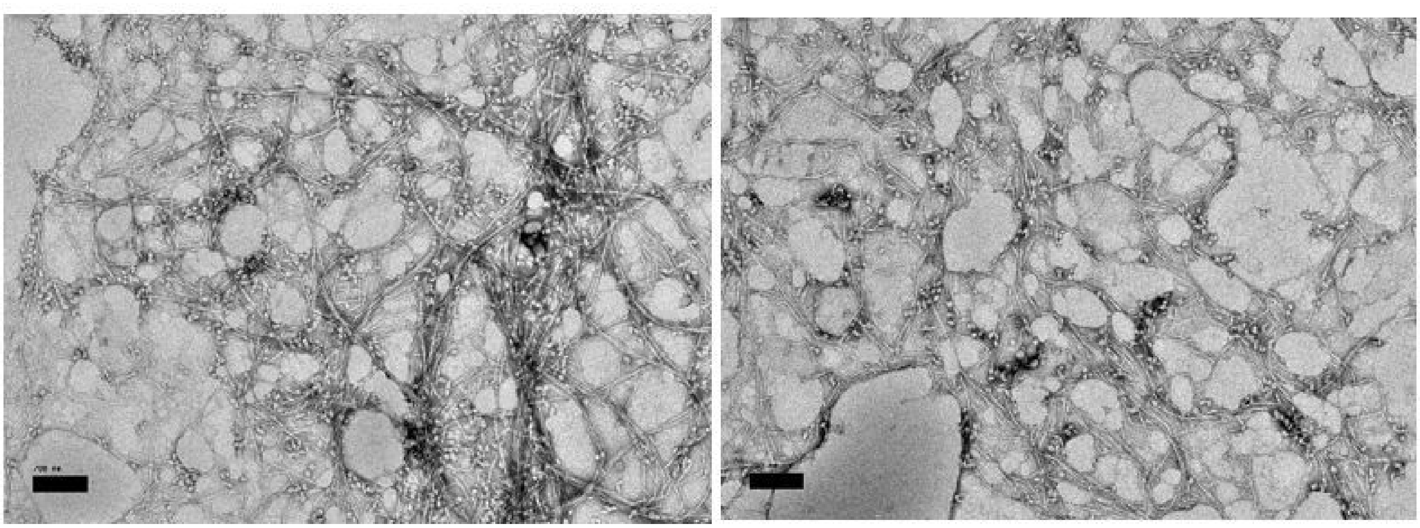

Figure 10

Morphologies of the microfibrillated cellulose-stabilized oil-in-water emulsions prepared using MFC produced from high pressure homogenization cycle 2 (left) and 10 (right) when observed under field emission analytical electron microscope with magnification power of 10000x.