



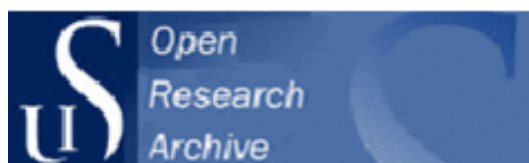
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1 **Investigation of a New Application for Cellulose Nanocrystals – A Study of the**
2 **Enhanced Oil Recovery Potential by use of a Green Additive**

3

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20 Oil recovery

21 CNC

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23 Heat aging

24 **Abstract**

25 Cellulose nanocrystals (CNC) has been investigated for a potential new application, enhanced
26 oil recovery (EOR), by performing core flooding experiments with CNC dispersed in low salinity
27 brine (CNC-LS) in outcrop sandstone cores. Experiments on 100 % water saturated cores
28 confirmed that most of the viscosity generating CNC particles were able to travel through the
29 cores at temperatures ranging from 60, to 120 °C. Oil recovery experiments on crude oil
30 saturated sandstone cores showed that when CNC-LS was used in tertiary mode, the ultimate
31 oil recovery could be increased, both at 90 and 60 °C. During tertiary CNC-LS injection, the
32 CNC particles increased fluctuations in differential pressure, an effect that can be linked to log
33 jamming in pore throats leading to remobilisation of oil in the pore space. The results from this
34 work indicate that CNC dispersed in low saline brine might have a certain potential for use in
35 enhanced oil recovery.

36

37 **1. Introduction**

38 The world is experiencing an ever-increasing energy demand, and although there is a lot of
39 focus on the “green shift”, with energy originating from renewable sources, one cannot avoid
40 the fact that petroleum-derived products will continue to be an important part of everyday life.

41 Many of the oilfields around the world are approaching, or is in the phase where the
42 rate of production is declining (Hendraningrat et al., 2013). The average oil recovery from
43 mature oilfields is between 20 and 40 %, meaning that as much as 60 – 80 % of the original
44 oil in place (OOIP) remains in the reservoirs after conventional recovery methods are spent.
45 Even with the current situation, with relatively low oil prices, an increase of a few percent of
46 OOIP might be economically viable compared to exploration and drilling of new wells. It is
47 becoming more and more difficult to discover new oilfields suitable for production, and many
48 of these unexplored fields are located in remote and/or environmentally vulnerable areas
49 (Muggeridge et al., 2014).

50 Oil is recovered by creating pressure gradients, which causes the oil to flow towards a
51 production well. In primary or first-line recovery, this process is mostly driven by natural flow,
52 sometimes assisted by artificial lift. Secondary oil recovery is usually assisted by gas-injection
53 or water-flooding, through injection wells for offshore reservoirs, and most often with seawater
54 (Muggeridge et al., 2014; Thomas, 2008; Wei et al., 2016). The injection fluid has low viscosity
55 compared to the oil phase which reduces the total sweep efficiency (Liu et al., 2012). The
56 ultimate oil recovery is determined by two different efficiencies; the microscopic displacement
57 efficiency, which is a measure of oil recovery at pore level. Due to wetting and capillary
58 trapping of oil in pore spaces, this efficiency rarely exceeds 70 % OOIP (Hu et al., 2016). The
59 second determinant is the macroscopic sweep efficiency, which refers to the amount of oil the

60 flooding fluid can contact. Technologies that improve these two parameters are usually
61 referred to as tertiary, or enhanced oil recovery (EOR) (Taber et al., 1997).

62 The term EOR comprises many different techniques, but in this paper the focus will be
63 on chemical methods, through alteration of the ion composition of the injection brine, and
64 nanoparticle flooding. The addition of polymers to the flooding fluid increases the viscosity of
65 the aqueous phase, thus lowering the mobility ratio between the water and oil phases. This
66 could be a favourable situation, by improving both the vertical- and area sweep efficiency
67 (Raney et al., 2012). Polymer flooding has been thoroughly researched and implemented in
68 fields for the last 40 years (Kamal et al., 2015). Two types of polymers are more extensively
69 utilised than others. These are the synthetic hydrolysed polyacrylamides (HPAMs) and the
70 polysaccharide biopolymer xanthan gum. HPAMs is most often used due to low cost and
71 improved viscoelastic properties compared to xanthan gum (Sheng et al., 2015). Both
72 polymers have some drawbacks though; HPAMs are generally not very stable in high salinity
73 dispersions and at elevated reservoir temperatures, in addition to being susceptible to shear
74 degradation (Raney et al., 2012). Xanthan gum tolerates high salinity and shear forces, but
75 has relatively low temperature stability (Seright & Henrici, 1990) and problems with formation
76 plugging, viscosity loss and bacterial degradation has been experienced (Wellington, 1983).
77 The problem with biodegradation is that biocides need to be added to the polymer injection
78 brine, which may cause an otherwise environmentally friendly polymer to become harmful to
79 the environment. HPAM also has some problems due to poor biodegradation properties,
80 making it necessary with post-flooding water treatment, which might be costly, time consuming
81 and poses an environmental threat if it fails (Guo, 2013). There is also some suspicion
82 regarding its breakdown into toxic acrylamide after slow natural degradation, which can cause
83 harm to local ecosystems (Aguilar & Mansur, 2016; Bao et al., 2010).

84 Cellulose nanocrystals (CNC) is here introduced as a potential green alternative to
85 water assisted polymer flooding. CNC is rod-like particles derived from cellulose of various
86 sources, most often wood, through controlled acid hydrolysis (Klemm et al., 2011). Coming
87 from an abundant and completely renewable source, CNC is both non-toxic and
88 biodegradable, making it a green flooding chemical according to the OSPAR Commissions
89 PLONOR list (OSPAR, 2016). CNC particles from wood are usually 3 – 5 nm wide and have
90 lengths ranging from 100 – 200 nm (Habibi et al., 2010). They do not alter the viscosity of the
91 injection brine significantly, and are added to improve the microscopic and macroscopic sweep
92 efficiencies through flow diversion. As is the case with the particles used in the experiments
93 in this article, CNC is often produced using 64 wt. % sulphuric acid at a temperature of 45 °C,
94 with reaction times depending on the temperature used (Reiner & Rudie, 2013). The acid
95 reacts with the hydroxyl groups on the surface of the cellulose, which removes the amorphous
96 part of the cellulose and yields crystalline CNC particles with charged sulphate half esters on

97 the surface. This leaves the particles anionic, promoting their dispersion behaviour in water
98 (Revol et al., 1992). The use of cellulose nanocrystals for petroleum industry applications is
99 not a complete novelty, it has been used as cement strengthener, by adding it to well fluids,
100 and it has been used to increase the viscosity of water-based well fluids for fracturing and
101 gravel packing (Rincon-Torres & Hall, 2015). Proposals have also been made to utilise CNC
102 in well treatment fluids as a substitute for conventional polymers (Lafitte et al., 2014). No
103 reports have been found of the use of CNC in enhanced oil recovery, but cellulose derivatives
104 like variations of hydrophobically modified hydroxyethyl cellulose (HM-HEC) have been
105 investigated, although these, like HPAMs have shown to be relatively salt sensitive (Kjøniksen
106 et al., 2008; Wever et al., 2011). Cellulose nanocrystals are believed to be less vulnerable to
107 shear and biological degradation due to their crystalline morphology (Aadland et al., 2016),
108 and has a thermal stability within the limits for oilfield applications (Heggset et al., 2017;
109 Molnes et al., 2017).

110 Presented in this paper is a lab scale study of the EOR potential in sandstone of
111 negatively charged cellulose nanocrystals used in combination with low salinity (LS) brine.
112 The stability of CNC in dispersions with low salinity (1000 ppm NaCl) brine has been
113 investigated and proved earlier (Molnes et al., 2016). Flooding procedures are performed
114 using CNC in combination with low saline brines which changes the wettability and
115 redistributes the residual oil within the pore spaces of the sandstone cores. CNC has also
116 been subjected to core flooding procedures at lab scale to investigate the injectivity and
117 eventual retention of CNC inside sandstone cores. These investigations have shown that the
118 CNC is injectable in sandstone and are able to travel through the core, but some filtering and
119 retention of larger particles was observed. These effects were enhanced when the CNC
120 concentration or the brine injection rate was increased (Aadland et al., 2016; Molnes et al.,
121 2016).

122 A proposed mechanism for CNC in a porous media is log-jamming, where the particles
123 block pore throats (larger than the particle size) and thus cause microscopic diversion in the
124 pore matrix. The most important factors regarding log-jamming are pore size distribution,
125 particle concentration and effective hydrodynamic size, as well as the injection flow rate
126 (Bolandtaba et al., 2009; T. Skauge et al., 2010).

127 The pore-jamming effect can partly be explained by the mass difference between the
128 particles and the dispersion medium. Pore throats are smaller than the pores, and combined
129 with the constant differential pressure, the flow velocity will increase at the pore throats
130 compared to inside the pores. Water molecules will accelerate faster than the particles at the
131 entrance of a pore throat, due to a significant difference in mass between a water molecule
132 and a particle. The particles will then start to accumulate at the pore throat and slowly reduce
133 the diameter of the pore throat and eventually block it.

134 The initial wetting properties of an oil reservoir is related to the chemical equilibrium
135 between rock surface, oil phase and brine phase, which is established over millions of years.
136 The extremes are completely oil or completely water wet, and a typical sandstone reservoir is
137 usually mixed wet. Improvement of the oil recovery from a reservoir in equilibrium is possible
138 through alteration of the ionic composition of the injected brine, which will destabilise the
139 system and increase the oil flow through the porous formation (Strand et al., 2016). The effect
140 of low saline brine flooding on enhanced oil recovery has been known for many years, and it
141 has also been confirmed experimentally and in the field (Lager et al., 2007; Seccombe et al.,
142 2010; Tang & Morrow, 1999a, 1999b). It is generally accepted that the effect of the LS brine
143 flooding in sandstone reservoirs is caused by a wettability alteration of the sandstone, but
144 there is still some debate regarding how this wettability modification takes place (Strand et
145 al., 2016). Over the years, a variety of mechanisms has been proposed: 1) Migration of fines
146 from clay (Tang & Morrow, 1999a), 2) Increase in pH due to impact from alkaline flooding
147 (McGuire et al., 2005), 3) Multi-component ion exchange (MIE) at the clay surface (Lager et
148 al., 2007), 4) Migration of fines causing microscopically diverted flow (A. Skauge, 2008), and
149 5) Ionic double layer expansion at the rock surface (Ligthelm et al., 2009). Austad et al. (2010)
150 described a new mechanism, based on, and in agreement with existing experimental data. At
151 reservoir conditions, the pH value of the formation water (FW) is slightly acidic, due to
152 dissolved acidic gases like H₂S and CO₂. Negatively charged mineral surfaces like clay work
153 as a cation exchange material, and will at this pH adsorb acidic and protonated basic
154 components from the crude oil, as well as cations like Ca²⁺ from the formation water. When
155 low salinity brine is injected into this system, a desorption of Ca²⁺ from the clay surface is
156 promoted, which will lead to a local pH increase in the interface between the clay and LS brine
157 because Ca²⁺ is substituted by H⁺ from the injected fluid. A fast reaction between OH⁻ and the
158 adsorbed acidic and protonated basic material leads to a desorption of organic material from
159 the clay, causing an improved water wetness, which generate positive capillary forces and
160 enhance the oil recovery (Austad et al., 2010).

161 It is experimentally verified in oil recovery experiments from restored cores that FW
162 injection, which will not chemically affect the initial core wettability established, gives a low oil
163 recovery, typically ~40 % OOIP (Piñerez Torrijos et al., 2016; Torrijos et al., 2017). The FW
164 injection is successively followed by tertiary LS injection which significantly improves the oil
165 recovery with 8-9 % OOIP, which could only be explained by a wettability alteration towards
166 more water wet conditions and is also confirmed in Spontaneous Imbibition (SI) experiments.
167 Using the same Crude oil-Brine-Rock system (CoBR), a secondary LS injection improved the
168 oil recovery to 66 % OOIP compared to only 40 % OOIP with FW (Piñerez Torrijos et al.,
169 2016). The secondary flooding with LS brine will cause a wettability change in the pore matrix
170 of the sandstone core, and redistribute the residual oil to the middle of the pores making it

171 more accessible for EOR techniques. Tertiary injection of CNC may cause log-jamming of
172 pore throats and divert the water flow into the lesser available, oil containing pores to further
173 increase the oil recovery.

174 This paper describes core flooding tests that have been performed on outcrop sandstone
175 cores, saturated with LS brine, at temperatures from 60 to 120 °C, while the oil recovery tests
176 were performed on outcrop sandstone cores at 60 and 90 °C. Testing at such elevated
177 temperatures is relevant due to the temperatures encountered in oil reservoirs. Reservoir
178 temperatures increases with burial depth, usually with 3 °C per 100 m depth, with
179 temperatures reaching above 150 °C (Beal, 1946; Jahn et al., 2008). The cores in the oil
180 recovery tests were restored with initial water saturation (S_{wi}) of 0.2, saturated and aged in
181 crude oil. The cores have been successively flooded, first with LS brine, followed by LS brine
182 with CNC. Both oil recovery and differential pressure over the core have been monitored
183 during the tests.

184

185 **2. Materials and Methods**

186

187 **2.1 Materials**

188

189 **Cellulose nanocrystals**

190 Cellulose nanocrystals (CNC) were acquired from the Process Development Centre at The
191 University of Maine (USA). The particles were produced by the Forest Products Laboratory
192 (FPL), USDA (US: Department of Agriculture) by acid hydrolysis of softwood pulp using 64 %
193 (by mass) sulphuric acid. CNC from the same provider was analysed by Sacui et al. (2014) by
194 transmission electron microscopy (TEM) and atomic force microscopy (AFM). The charge
195 density of the sulphate ester groups was evaluated by Heggset et al. (2016). All values are
196 shown in Table 2.1.

197

198 **Table 2.1:** CNC characteristics, adapted from (Heggset et al., 2017) and (Sacui et al., 2014).

Sample	Charge density (mmol/g)	Crystallite diameter (nm)^c	Crystallite length (nm)^c	Functional groups
CNC	approx. 0.3 ^{a,b}	5.9 ± 1.8	130 ± 67	-OH, -SO ₃ H

199 ^aAmount of sulphate ester groups

200 ^bMeasured with inductively coupled plasma-atomic absorption (ICP-AA) (Heggset et al., 2017).

201 ^cDetermined with atomic force microscopy (AFM) (Sacui et al., 2014).

202

203 The dispersion stability of CNC particles at elevated temperatures has been investigated at
204 an earlier point (Heggset et al., 2017; Molnes et al., 2017), and the particles are regarded as
205 relatively stable in low saline dispersions at the temperatures and time frames used in the

206 experiments performed for this article. However, the colloidal stability of CNC is to some
 207 degree affected by electrolytes. Zhong et al. (2012) have reported that the zeta potential of
 208 CNC dispersions was reduced to below -30 mV and that the particles started to agglomerate
 209 at Na⁺ concentrations from 10 mM (Zhong et al., 2012). High temperatures will also cause
 210 desulfation of CNC with reduced surface charge and less colloidal stability as a result (Dorris
 211 & Gray, 2012).

212

213 **Brines**

214 Brines were prepared using de-ionized (DI) water and reagent grade chemicals as given in
 215 Table 2.2. After adding the salts, the brine solutions were stirred for approximately one hour,
 216 before being filtrated over 0.22 μm Millipore filter paper. Formation water (FW) brine was
 217 diluted five times with DI water and will from now be denoted d₅FW. The finished brines were
 218 stored at room temperature. CNC dispersions were prepared by adding exact amounts of CNC
 219 to LS brine to achieve a concentration of 0.5 wt. %; before the dispersions were vigorously
 220 shaken until the particles were well dispersed in the brine. The salinity contribution by Na⁺
 221 present in the CNC particles was not considered, as Na⁺ only exists in CNC in minuscule
 222 amounts.

223

224

Table 2.2: Brine compositions. TDS = total dissolved solids.

Ions	FW (mM)	d ₅ FW (mM)	LS (mM)
Na ⁺	1540	308	17
Cl ⁻	1720	356	17
Ca ²⁺	90	24	-
TDS (mg/L)	100 000	20 000	1000

225

226

227 **Core material**

228 Two cylindrical outcrop sandstone cores were utilised in the experiments, and are denoted
 229 SM8 and SM10, respectively. The mineralogical properties have earlier been investigated
 230 through X-ray diffraction analysis (XRD), and are given in Table 2.3.

231

232 **Table 2.3:** Mineralogical properties of the outcrop sandstone cores used in the experiments, given in
 233 wt. %.

Albite	Quartz	Calcite	Apatite	Pyrite	Anatase	Chlorite	Illite	Clays & micas
32.0	57.0	0.3	0.2	0.1	0.5	1.9	8.6	10.5

234

235 The physical properties of the cores are provided in Table 2.4.

236

237 **Table 2.4:** Physical core properties

Core name	Length, cm	Diameter, cm	Dry weight, g	Sat. weight, g	Porosity, %	PV, mL
SM8	7.05	3.79	164.37	180.73	20.33	16.17
SM10	7.04	3.80	165.79	182.33	20.47	16.34

238

239 The pore distribution of the core material has been investigated by mercury injection (MICP),

240 confirming heterogeneous pore distribution, Fig. 2.1.

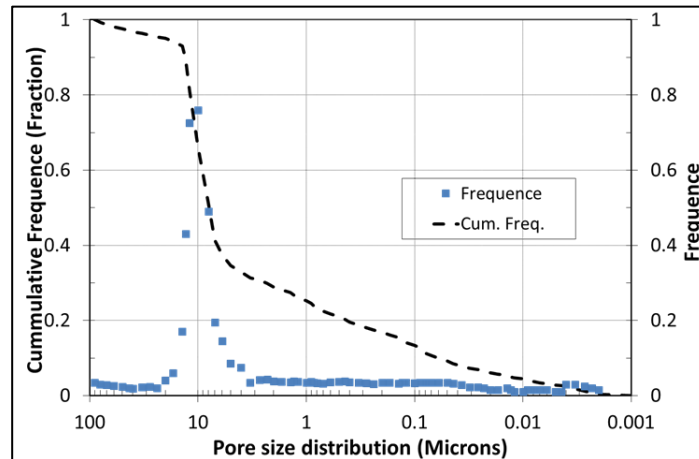


Figure 2.1: Pore size distribution tested by Mercury injection (MICP) into a core from the same block as the tested core material. The core material has a heterogeneous pore size distribution, with pores ranging from ~ 0.01 to $100 \mu\text{m}$. The majority of the pores are in the range of $10 \mu\text{m}$.

241

242 The same outcrop core material has been used in previous EOR and LS brine studies with

243 very good experimental reproducibility between individual cores (Piñerez Torrijos et al., 2016;

244 Torrijos et al., 2017).

245

246 Crude oil

247 Stabilised crude oil was centrifuged at high rotation speed for 1 hour and filtrated through 8

248 μm Millipore filter and at last with a $5 \mu\text{m}$ filter to remove particles and precipitates. The acid

249 number (AN) characterises crude oil through the total concentration of strong and weak acidic

250 organic compounds, and is given as the amount of potassium hydroxide (KOH) needed to

251 neutralise the acids in one gram of oil. The base number (BN) is a measure of the alkalinity of

252 crude oil, given in mg KOH/g crude oil. Both values were acquired by potentiometric titration

253 based on modified versions of ASTM (American Society for Testing Materials) D2896 for BN

254 and ASTM D664 for AN ((ASTM), 1988, 1989; Fan & Buckley, 2006). The crude oil properties

255 are given in Table 2.5.

256

257

258

259 **Table 2.5:** Crude oil properties

Acid number (AN) mg KOH/g	Base number (BN) mg KOH/g	Density g/cm ³ at 20 °C	Viscosity, cP at 20 °C
0.1	1.8	0.8459	17.6

260

261

262

263 **2.2 Methods**

264

265 **Core cleaning**

266 A mild core cleaning procedure was used. The core was first flooded with kerosene to displace
267 any residual crude oil from the core. Then followed heptane to displace the kerosene, and at
268 last the core was flooded with 1000 ppm NaCl brine for a few pore volumes (PV), to displace
269 brine and easily dissolvable salts. After this, the core was dried at 90 °C to a constant weight.

270

271 **Core restoration**

272 Initial water saturation (S_{wi}) of 20 % was established in the mildly cleaned and dried core. The
273 core was saturated under vacuum with 5 times diluted FW (d_5FW). 20 % S_{wi} with FW was
274 established using the desiccator technique (Springer et al., 2003).

275

276 Oil saturation and core aging. The core with $S_{wi} = 20\%$ was mounted in a core holder. Gas
277 was removed from the pores by vacuum evaporation, and the core was saturated with crude
278 oil. The core was then flooded with 2. PV of the filtrated crude oil in both directions at 50 °C.
279 The core was then placed in an aging cell and aged for 14 days at the test temperature (60 or
280 90 °C) before oil recovery experiments were performed. .

281

282 **Oil recovery experiment**

283 The oil recovery experiment was performed in a computer controlled setup with a Gilson HPLC
284 307 pump, stainless steel piston cells with either LS brine or CNC-LS brine dispersion. The
285 Hassler core holder was placed in a heating cabinet with gauges for monitoring the inlet and
286 differential pressure (ΔP), as well as the temperature. All experiments were performed with a
287 confining pressure of 20 Bar and a back pressure of 10 Bar. A schematic overview of the core
288 flooding setup can be seen in Figure 2.2.

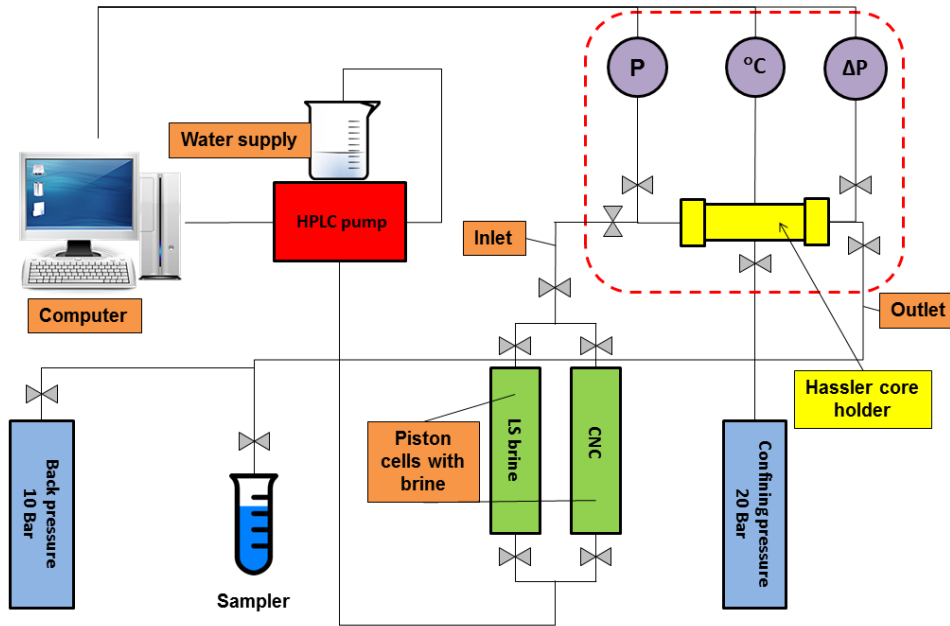


Figure 2.2: Core flooding setup. Line valves are shown in grey, and were used to regulate line flow. The stapled line indicates the heating cabinet.

289

290 The restored core was placed in the core holder. The core was equilibrated at test conditions
 291 overnight with bypass flow (no fluid flooded through the core) to ensure stable temperature
 292 and pressure as well as air tight flood lines prior to testing. The core was successively flooded
 293 with LS brine and CNC-LS dispersion at a rate of 4 PV/day. Produced oil and water was
 294 monitored and collected using a glass burette and a Gilson GX-271 Liquid Handler (Gilson
 295 Inc., Middleton, WI, USA) by night.

296

297 **CNC injectivity and core permeability**

298 The core permeability was measured on a 100% water saturated core by flooding LS brine at
 299 increased flood rate to acquire stable and accurate differential pressure (ΔP) measurements.
 300 This was performed before injecting CNC-LS dispersion and then between every temperature
 301 step.

302 Permeability is calculated using the Darcy equation (Eq. 2.1) and given in milliDarcy
 303 (mD) (Zolotukhin & Ursin, 2000):

304

$$305 \quad k = \frac{\mu \Delta x}{A} \frac{u}{\Delta P} \quad (2.1)$$

306

307

308

309 Where

310

311 μ Viscosity of the injected fluid (for water at 60 °C = 0.47 mPa·s)

312 Δx Length of sandstone core sample (cm)

313 A Cross sectional area of sandstone core sample (cm²)

314 u Flow rate of the injected fluid (mL/min)

315 ΔP Differential pressure over the sandstone core sample (mBar)

316

317 The oil recovery experiments were performed in the same core flooding setup, using crude oil
318 saturated core SM10 with $S_{wi} = 20\%$, at 60 and 90 °C. The core was cleaned and restored
319 prior to each EOR test.

320

321 **Effluent characterisation**

322 The viscosity of brine and the effluent CNC-LS samples was assessed using an Anton Paar
323 MCR 301 rotational rheometer. The instrument was configured with a 50 mm 1° cone and
324 plate geometry with a measuring gap of 0.096 mm. Measurements were performed at 20 °C,
325 with shear rates from 10 – 1000 1/s, and were monitored and logged using the Rheoplus
326 software v3.40.

327 pH values of LS and CNC-LS effluent samples were measured with a Mettler Toledo
328 SevenEasy™ pH meter.

329

330

331 **3. Results and Discussion**

332

333 **3.1 CNC Injectivity study**

334 The injectivity of CNC into sandstone cores was investigated. LS brine with 0.5 wt. % CNC
335 was injected at constant temperature into a 100 % LS saturated outcrop sandstone core SM8
336 at a flooding rate of 4 PV/day. The inlet pressure and differential pressure over the core was
337 monitored throughout the whole experiment and effluent samples were collected. In between
338 each experiment, the core was cleaned by injection of LS brine in the opposite direction, until
339 stable pressure drop. Experiments were performed at constant temperature, 60, 90 and 120
340 °C.

341 The core flooding at 60 °C had an initial ΔP of 6 mBar (100 % LS brine saturation), and
342 the pressure drop gradually increased to 15 mBar after 1 PV injected and continued to
343 increase. After 7 PV, the ΔP had increased to 60 mBar. Viscosity measurement on effluent
344 samples showed only a very small reduction in the viscosity of the CNC-LS dispersion,
345 confirming that mostly all CNC particles are being transported through the porous media, and

346 the main part of the pressure build-up is linked to the filtration of the largest fraction of CNC
347 particles at the inlet surface. The effluent viscosity measurements are shown in Figure 3.1.
348

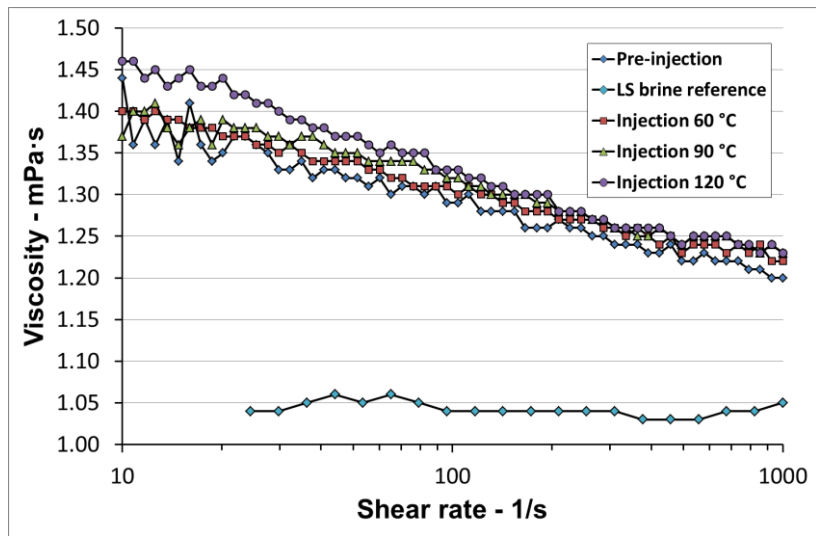


Figure 3.1: Core flooding with 0.5 wt. % CNC-LS brine into core SM8 at 60, 90 and 120 °C. The viscosity of effluent samples was measured and compared with the viscosity of a pre-flooded CNC-LS sample. Measurements were performed at increasing shear rates at 20 °C.

349

350 The core SM8 was regenerated for the next test by flooding the core with LS brine in the
351 reversed direction, to remove CNC particles from inside the core and from the inlet surface.
352 LS brine flooding confirmed only slight changes in core permeability on the regenerated core.
353 The CNC-LS flooding test was repeated at both 90 and 120 °C. The pressure build-up effects
354 are presented in Figure 3.2.

355

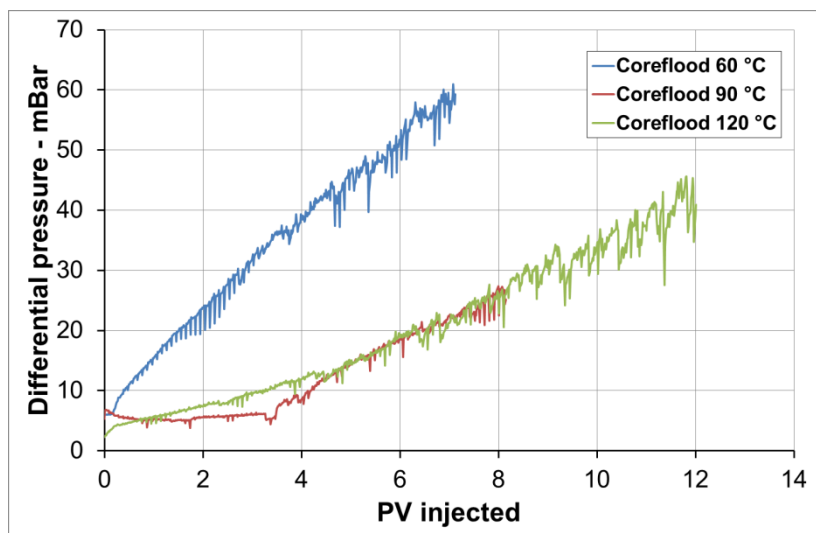


Figure 3.2: Core flooding of a 100 % LS saturated sandstone core SM8. The core was flooded with 0.5 wt. % CNC-LS brine at a rate of 4 PV/Day at 60, 90, and 120 °C. Pressure drop (ΔP) over the core was measured as a function of PV injected.

356

357 Less pressure build-up is observed at 90 and 120 °C compared to 60 °C. The viscosity of
 358 effluent samples at 90 and 120 °C are also in line with the effluent viscosity at 60 °C, confirming
 359 that the major part of the viscosity-contributing CNC particles is transported through the porous
 360 media at all tested temperatures, indicated in Figure 3.1. At a shear rate of 10 1/s, the effluent
 361 viscosity at 60, 90 and 120 °C is close to 1.40 mPa·s, while the viscosity of the bulk dispersion
 362 is 1.45 mPa·s. In a previous study, the viscosity of the CNC-LS solution significantly increased
 363 during long term temperature aging (Molnes et al., 2017). No increase in the viscosity of CNC-
 364 LS effluent viscosity were observed during these experiments. A possible explanation for this
 365 is that the CNC particles are not trapped in the pore matrix long enough for the viscosity
 366 increase to take place. At an injection rate of 4 PV/day, a CNC particle will use approximately
 367 6 hours to travel through the core. The heat aging experiments revealed that this increase in
 368 viscosity is not observed before the dispersion has been aged for at least 20 hours (Molnes et
 369 al., 2017). On a lab scale, the time frame is thus too short to observe these viscosity changes,
 370 but on oil reservoir scale, there is a possibility that this heat viscosifying effect may increase
 371 the viscosity of the CNC-LS solution giving an extra support to the oil recovery process.

372 The core permeability in the different experiments is given in Table 3.1. Permeability
 373 values in mD were acquired using Equation 2.1.

374

375 **Table 3.1:** Permeability of core SM8 after core cleaning/core regeneration, and prior to injectivity tests
 376 (K_1) at 60, 90 and 120 °C. K_2 indicates the calculated values between measurements, after
 377 regeneration.

Temperature, °C	K_1 , mD	K_2 , mD
60	63	44
90	44	75
120	75	68

378

379 As observed after the 90 °C CNC-LS injection, the permeability of the core sample was
 380 increased. This was not expected, as earlier injection studies with CNC-LS generally exhibited
 381 a decrease in permeability, due to particles trapped inside the porous medium (Molnes et al.,
 382 2016). The reason for this effect may be a beginning degradation of CNC trapped in the porous
 383 media. Prolonged exposure to high temperatures can change the surface chemistry of the
 384 CNC, for example by release of sulphate ester groups (Molnes et al., 2017), and this change
 385 may lead to desorption/un-jamming of trapped CNC. Combined with an elevated injection rate
 386 between the measurements, the CNC particles are probably expelled from the core sample.

387 The increase in differential pressure over the core SM8 is mainly caused by filtering at
 388 the core inlet, but some adsorption on pore surfaces or jamming in pore throats cannot be
 389 excluded. As mentioned in the Introduction, the CNC particles are negatively charged, due to
 390 the sulphate half esters substitutions on their surfaces through the production process. Silicate

391 minerals in the pore surfaces are also negatively charged, which excludes electrostatic
392 adsorption.

393 The pH values in the effluent samples were also tested. It is known that brine pH of
394 non-buffered systems can increase when flooded through sandstone, due to a cation
395 exchange reaction with pore surface minerals, where H^+ exchange with cations at mineral
396 surfaces (Austad et al., 2010). The bulk pH of the CNC-LS brine was 5.7, as shown in Figure
397 3.3.

398

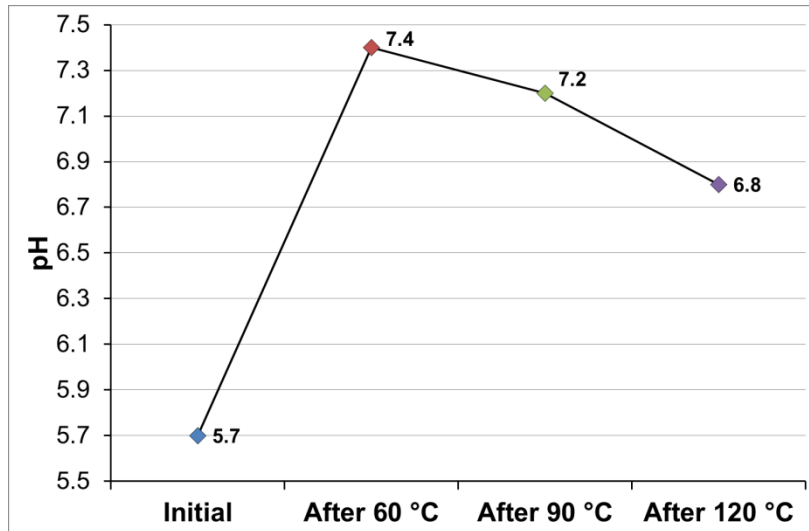


Figure 3.3: pH of 0.5 wt% CNC-LS brine samples, before injection and in effluent samples after core flooding at 60, 90 and 120 °C.

399

400 As seen in the figure, the pH observed in CNC-LS effluent samples at 60, 90 and 120 °C
401 confirmed a ΔpH of 1.7, 1.5 and 1.1 respectively. The results are in agreement with reduced
402 pH changes during LS brine injection in sandstone cores at increasing temperatures (Piñerez
403 Torrijos et al., 2016).

404

405 The observed effluent pH could also be affected by the chemistry of CNC, like sulphate ester
406 groups being split off (Dorris & Gray, 2012; Heggset et al., 2017; Molnes et al., 2017).

407

408 **3.2 Oil recovery experiments**

409 Enhanced oil recovery experiments were performed both at 60 and 90 °C using sandstone
410 core SM10. After a mild core cleaning, the core was restored with $S_{wi} = 20\%$, and saturated
411 and aged in crude oil. The tests were performed by initially LS injection (secondary mode) until
412 ultimate oil recovery plateau was reached, before the CNC-LS solution was injected in tertiary
413 mode.

414 The results from the oil recovery test performed at 90 °C are shown in Figure 3.4.

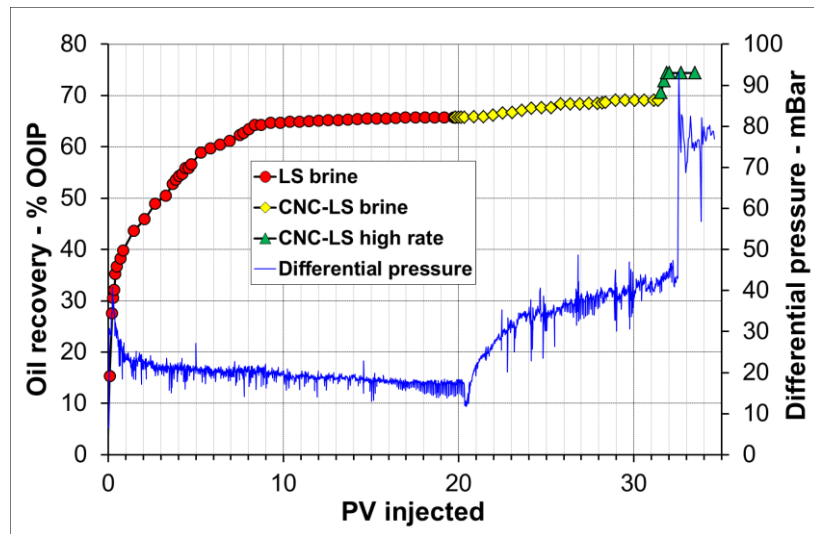


Figure 3.4: Oil recovery test performed on core SM10 at 90 °C. The core with $S_{wi} = 0.2$ and saturated and aged with crude oil, was successively flooded with LS – CNC-LS brines a rate of 4 PV/Day. The oil recovery (% OOIP) and ΔP (mBar) are shown as a function of PV injected (time). At the end, the injection rate was increased to 16 PV/Day.

416

417 During secondary LS brine injection, the oil recovery steadily increased, and an ultimate
 418 recovery plateau of 66 % OOIP was reached after 8 PV injected. The LS recovery result is in
 419 line with the observed results by (Piñerez Torrijos et al., 2016).

420 The ΔP gradually decreased as the water saturation of the core increased. A fluctuation in ΔP
 421 was observed during LS injection, which is an indication that mobile oil is moving through the
 422 pore throats. The ΔP fluctuation decreased when the ultimate recovery was reached. 19 PV
 423 of LS brine was injected to ensure that all mobile oil was produced.

424 When the CNC-LS brine was injected, an increase in differential pressure was observed.
 425 During the next 11 PV, 3.4 % OOIP extra oil was produced. In the same period, there was
 426 also observed an increase in the ΔP fluctuation, which could indicate increased mobilisation
 427 of oil in the pore space due to the introduction of CNC particles. At the end, the injection rate
 428 was increased 4 times to 16 PV/day. Differential pressure increased significantly with
 429 increased injection rate and an extra oil production of 5.4 % OOIP was obtained. The pH of
 430 the bulk CNC-LS dispersion was 5.7, and the pH measured in sampled CNC-LS effluent was
 431 7.4.

432 Oil recovery studies on the same CoBR system, confirms reproducible ultimate recoveries
 433 during secondary FW injections in the range of 35 – 40 % OOIP at 60 – 120 °C, which are
 434 explained by low pH of produced water (PW) which is not promoting wettability alteration and
 435 increased microscopic sweep efficiency (Piñerez Torrijos et al., 2016). Compared to the
 436 ultimate oil recovery plateau of 66% and pH increase observed during secondary LS
 437 injections, the effect of improved microscopic sweep efficiency could have a dramatic effect

438 on the amount of producible oil from heterogeneous pore structures, and the tertiary CNC—
439 LS injection at 90 °C was able to improve the ultimate recovery from 66 – 69.4 % OOIP.

440 A second oil recovery experiment was performed on core SM10 at 60 °C, also using
441 CNC-LS in tertiary mode after LS injection. The results from the test is presented in figure 3.5.

442
443

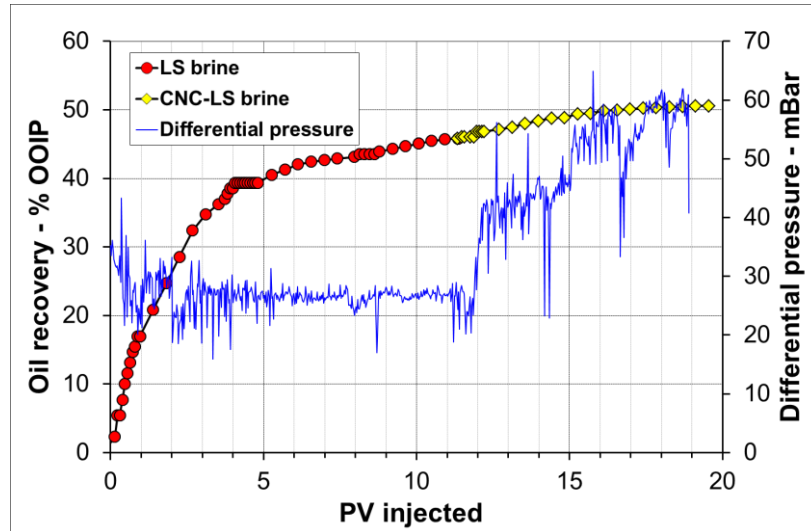


Figure 3.5: Oil recovery test performed on core SM10 at 60 °C. The core with $S_{wi}=0.2$ and saturated and aged in crude oil, was successively flooded with LS – CNC-LS brines a rate of 4 PV/Day. The Oil recovery (%OOIP) and the ΔP (mBar) are reported as a function of PV injected (time).

444
445

446 During the LS injection, the oil recovery gradually increased and reached 45.8 %OOIP after
447 11 PV. Fluctuation in ΔP indicated oil production. After 5 PV injected, the differential pressure
448 fluctuations stabilised as the oil production decreased. Compared to the experiment performed
449 at 90 °C and previous LS EOR experiments performed on the same CoBR system, the LS
450 brine only improved the oil recovery from 40 % OOIP, which are expected as base line
451 recovery without wettability alteration using FW, to 45.8 % OOIP after 11 PV injected. In this
452 experiment, the LS brine failed to contribute with an efficient wettability alteration and
453 increased microscopic sweep efficiency.

454 At 11 PV, the injection brine was changed to CNC-LS. At the changing point the core was
455 less water wet compared to the experiments observed after LS injection at 90 °C and at 60 °C
456 by (Piñerez Torrijos et al., 2016). As the CNC-LS brine was injected, the differential pressure
457 increased with increased fluctuations, Figure 3.4. The introduction of CNC particles into the
458 pores space affects the fluid flow in the pores, and log jamming of CNC particles in pore throats
459 and redistribution of oil could be an explanation of the observations. At less water wet
460 conditions, the CNC-LS injection is not contributing with a significant increased recovery

461 compared to LS injection. The experiment was terminated after 20 PV injected, with an
462 ultimate oil recovery of 51 % OOIP. Table 3.2 shows the measured pH values for the bulk LS
463 brine and CNC-LS dispersion prior to injection, and pH values of the produced water samples
464 during LS and CNC-LS injection.

465

466 **Table 3.2:** pH measured in bulk solutions and produced water samples at 60 °C.

Sample	Bulk pH	Effluent pH	ΔpH
LS brine	5.8	7.6	1.8
CNC-LS	5.6	6.4	0.8

467

468 A less pH increase is observed in PW samples during CNC-LS injection compared to
469 LS brine, and this may indicate that the CNC-LS brine is less efficient as a wettability modifier.
470 It may also be that the pH is influenced by desulfation (Dorris & Gray, 2012).

471

472 For both experiments, a fluctuation in differential pressure over the core was observed during
473 CNC-LS injection in tertiary mode, which may be a sign of log jamming and redistribution of
474 oil within the pore space. The pores are in the range from 0.01 to 100 μm, with the majority at
475 10 μm. According to Zhong et al. (2012), CNC can form agglomerates of approx. 1 μm at a
476 Na⁺ salinity of 50 mM. Agglomeration may also be promoted by reduced surface charge due
477 to desulfation at the elevated temperature with reduced repulsion of particles as a result. Log
478 jamming is thus likely. An increase in pH was seen for the CNC-LS dispersion at both test
479 temperatures, with the largest increase observed at 90 °C, with a Δ pH of 1.7. At 60 °C, the Δ
480 pH was 0.8. The large jump at 90 °C is most likely caused by changes in the reactivity of the
481 CNC particles with increasing temperature. The rock minerals contribute with the same pH
482 increase during LS injection at both 60, 90, 120 °C (Piñerez Torrijos et al., 2016).

483 As discussed, the CNC particles could cause water diversion by jamming up in easily flooded
484 pore throats, which could lead the water flow diversion into less available pores.

485

486

487 **4. Conclusions**

488 The injectivity of CNC-LS brine into sandstone cores have been studied at 60, 90 and 120 °C.
489 The CNC particles were injectable at all studied temperatures. Rheological measurements of
490 effluent samples confirmed that the main part of CNC particles travelled through the core
491 material. The increase in differential pressure was substantially lower at 90 and 120 °C,
492 compared to 60 °C. Core regeneration by injecting low saline brine in reverse confirmed that
493 most of the retained particles were filtered on the inlet of the core, and that only small changes
494 in core permeability was observed. The particle size of the CNC should be optimised to reduce
495 the observed filtration on the core inlet. Oil recovery tests with CNC-LS brine used in tertiary

496 mode after LS injection showed that the CNC dispersion affect fluid flow in the pores. As the
497 CNC dispersion was injected, increased fluctuation in the differential pressure over the core
498 was observed.

499 The oil recovery experiment performed at 90 °C showed a small but significant tertiary CNC-
500 LS EOR effect of 3.4 %OOIP, after a secondary LS injection promoting an efficient wettability
501 alteration towards more water wet conditions. Without an efficient wettability alteration during
502 the LS injection, no significant EOR effect during tertiary CNC-LS was observed in the oil
503 recovery test performed at 60 °C. The preliminary results indicate that CNC particles may
504 have some potential as a green flooding fluid additive, but that more investigation is needed.
505 At this early stage of research, it is difficult to suggest a mechanism behind the extra oil
506 observed. The CNC particles could participate in log jamming and agglomeration in pore
507 throats, as the core floodings showed increased pressure drop fluctuations during CNC-LS
508 injection. Both the CNC particles and the silicate minerals present in the cores are negatively
509 charged at typical reservoir pH, and further studies should include CNC particles with modified
510 surface charges which could affect log jamming and diverted flow. The chemistry of the CNC
511 particles also effected the CoBR chemistry which are important in explaining wettability and
512 wettability alteration processes as observed during FW and LS brine injection.

513

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523

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