International Journal of Trend in Scientific Research and Development (IJTSRD)
Volume 6 Issue 5, July-August 2022 Available Online: www.ijtsrd.com e-ISSN: 2456 – 6470

Synthesis, Characterization of Borax Crosslinked Guar Gum Hydrogel and its Effect in Treatment of Crude Oil Emulsified Water

Ayman Abu Reid, Yousif. A. Ragab, Salih. MAA, Imad. E. A. Mahajoub

Faculty of Engineering and Technology Department of Chemical Engineering,
University of Gezira, Wad Madani, Sudan

ABSTRACT

Nonionic nature of guar gum provides an acceptable material for modification, guar gum based hydrogel were synthesized by crosslinking purified guar gum with borax (disodium tetraborate, decahydrate) at different percentage. Estimation of crosslinked borax has carried out titrimetrically and occurrence of the reaction was confirmed using FT-IR spectroscopy, elemental analysis, thermal analysis, scanning electron microscopy and rheologically. To examine reverse demulsification efficiency of different grades of hydrogel, a (crude oil-in-water emulsion) sample was studied by a modified jar test method. The efficiency of the test materials was compared with the Industrial Reverse Demulsifier (IRD). The best grades of hydrogel didn't outperformed industrial reverse demulsifier but at low concentration (GCB6) diluted to about 0.3% in a dose of 50ppm a maximum efficiency of 66% was reached. The single step synthesized green product thus exhibited a potential as reverse demulsification agent which can be developed.

KEYWORDS: *guar gum, borax crosslinked guar gum, hydrogel, oil-in-water emulsion, water purification*

How to cite this paper: Ayman Abu Reid | Yousif. A. Ragab | Salih. MAA | Imad. E. A. Mahajoub "Synthesis, Characterization of Borax Crosslinked Guar Gum Hydrogel and its Effect in Treatment of Crude Oil Emulsified Water" Published in International Journal of Trend in Scientific Research and Development (ijtsrd), ISSN: 2456-6470, Volume-6 | Issue-5, August 2022, pp.1543-1556, URL: www.ijtsrd.com/papers/ijtsrd50675.pdf

IJTSRD50675

INTRODUCTION

Guar gum is an excellent starting material for modified polymers. Research work on guar and its derivatives during last few decades has revealed its tremendous potential for application in various industries. The ability of guar gum to hydrate quickly to produce viscous solution even in cold water has been its specialty, which proves its superiority over other counterparts. Guar gum is in fact the powdered endosperm of the seeds of the Cyamopsis tetragonolobus which contains a complex polysaccharide called galactomannan. This hydroxyl group is rich high molecular weight polymer consisting of d-mannose backbone on which d-galactose units are attached as side-chains. These functional groups form hydrogen bonding with water to impart viscosity and thickening to the solution [1]

Crosslinking may result either through the polymerization of monomers with functionality greater than two (by condensation mode) or by covalent bonding between preformed polymer molecules accomplished by irradiation, sulfur vulcanization, or miscellaneous chemical reactions. Crosslinking restricts chains from sliding past one another and generates elasticity in an amorphous polymer. It makes a polymer more resistant to heat, light, and other physical agencies, giving it a high degree of dimensional stability, mechanical strength, and chemical and solvent resistance. The effects of crosslinking on the physical properties of the polymers are primarily influenced by the degree of crosslinking, the regularity of the network formed, and the presence and absence of crystallinity in the polymer, this is reflected in an enhancement of glass transition temperature, an improvement in creep behavior also results from crosslinking as it restricts the viscous flow [2].

The inattentive behavior of humans toward environment has majorly affected the condition of water by contaminating it with inadequate quantities of pesticides, drugs, dyes and other pollutants [3].

Hydrogels with controlled biodegradability, stability towards chemicals and biochemical, shape stability, biocompatibility, and high permeability of water-soluble nutrients and metabolites are the novel biomaterials of the present era. The distinct features make them successful polymeric biomaterials in different fields [4].

Hydrogels may act as new emerging technology that is being researched for water remediation [5].

It exhibit most fragile three-dimensional structure, soft nature, high water soaking capacity and porosity which makes it extremely suitable for loading water-soluble bodies [6]

Chemically crosslinked hydrogels are irreversible and stable hydrogels involving chemical crosslinking between polymeric networks and involves the reaction of a polymeric backbone with crosslinking agent. Polysaccharides have functional groups like hydroxyl or amine group which on reaction with crosslinking agent form gel structure. Crosslinking may occur by different mechanisms like condensation, addition, and vulcanization. These hydrogels having strong covalent bonds can attain an equilibrium swelling state which depends upon the (polymer-solvent interactions) and crosslink density. Polysaccharides like cellulose and its derivatives, chitosan, alginate, dextran, guar gum, and starch can be crosslinked in the presence of chemical reagents and in the presence of radiations to form chemical hydrogels [7].

Crosslinked guar gum derivatives have revealed tremendous potential to be exploited as flocculating agents [8] in drug delivery [9] as dye removing agent [10] as metal sorbent [11].

During production, crude oil is usually produced commingled with water. This mingling production with water creates many problems during oil production. Water produced during oil production is either produced as free water and so it will settle out fairly quickly, or the water may be combined with oil in the form of emulsions [12].

Even when the proportion of water is small relative to the oil, when they flow together in a line, the lower viscosity of the water causes it to flow much faster and more turbulently past the oil. This causes the oil to emulsify into the water forming a reverse emulsion. The water also becomes emulsified into the oil. When that obverse emulsion breaks, the oil between the water droplets becomes a reverse emulsion [13].

We chose to carry out a study to investigate the effect of crosslinking of borax onto purified guar gum and compare the resulted hydrogels efficiency with the industrially used reverse-demulsifiers used in the treatment of oil in water emulsions.

EXPERIMENTAL

A. Materials

Extracted polysaccharide, Borax, A.R. (di-sodium tetraborate, decahydrate, 99.0% pure), Acetone for synthesis (99.0%) from CDH Ltd. New Delhi.

B. Measurements

Fourier transform infrared (FT-IR)

FT-IR spectra were acquired on Shimadzu FT-IR 8400 S.CE instrument using potassium bromide (KBr) discs prepared from a powder samples mixed with dry KBr in the ratio 1: 100. FT-IR spectra were analyzed in the Central Research laboratories, University of Khartoum, all FT-IR spectrums were used as a digital forms except for (PGG 15) which transformed into a plot form using Microsoft excel 2013.

Elemental analysis (C, H, N)

(C, H, and N) elemental content of purified guar gum were analyzed using Vario EL III instrument, Micro Analytical Center (MAC) Cairo University.

Thermal gravimetric analysis (TGA) and Differential thermal gravimetric analysis (DTG)

Thermo-gravimetric analysis (TGA) was carried out with the help of Thermo Gravimetric and Differential Thermal Analyzer (TGA-50 SHIMADZU/DTG-60H SHIMADZU), Micro Analytical Center (MAC) Cairo University.

Samples were placed in a platinum crucible, weight loss was recorded while the temperature increasing from 0.00 to 600 C° at a rate of 10 C°/min without hold time under a nitrogen flow.

Scanning electron microscopy (SEM)

The microstructure of the sample was characterized by Scanning Electron Microscope at magnification 2000x and 5000x using (SEM, JSM-6610, JEOL, Japan), Micro Analytical Center (MAC) Cairo University.

Rheological behavior

The rheological behavior of diluted aqueous solutions was studied at 25C° using Brookfield DV-III ultra-programmable rheometer, Spindle-21, Density for the same solution was measured using density bottle. National Research Center, Cairo, Egypt.

Reverse demulsification evaluation (Bottle Test)
Samples obtained from (Block6 - Bleela oilfield – CPF - sedimentation tank 1501B – level 5 meter), after flushing thoroughly, a 5L sample taken where the stream is well mixed, sample temperature about 60C°.

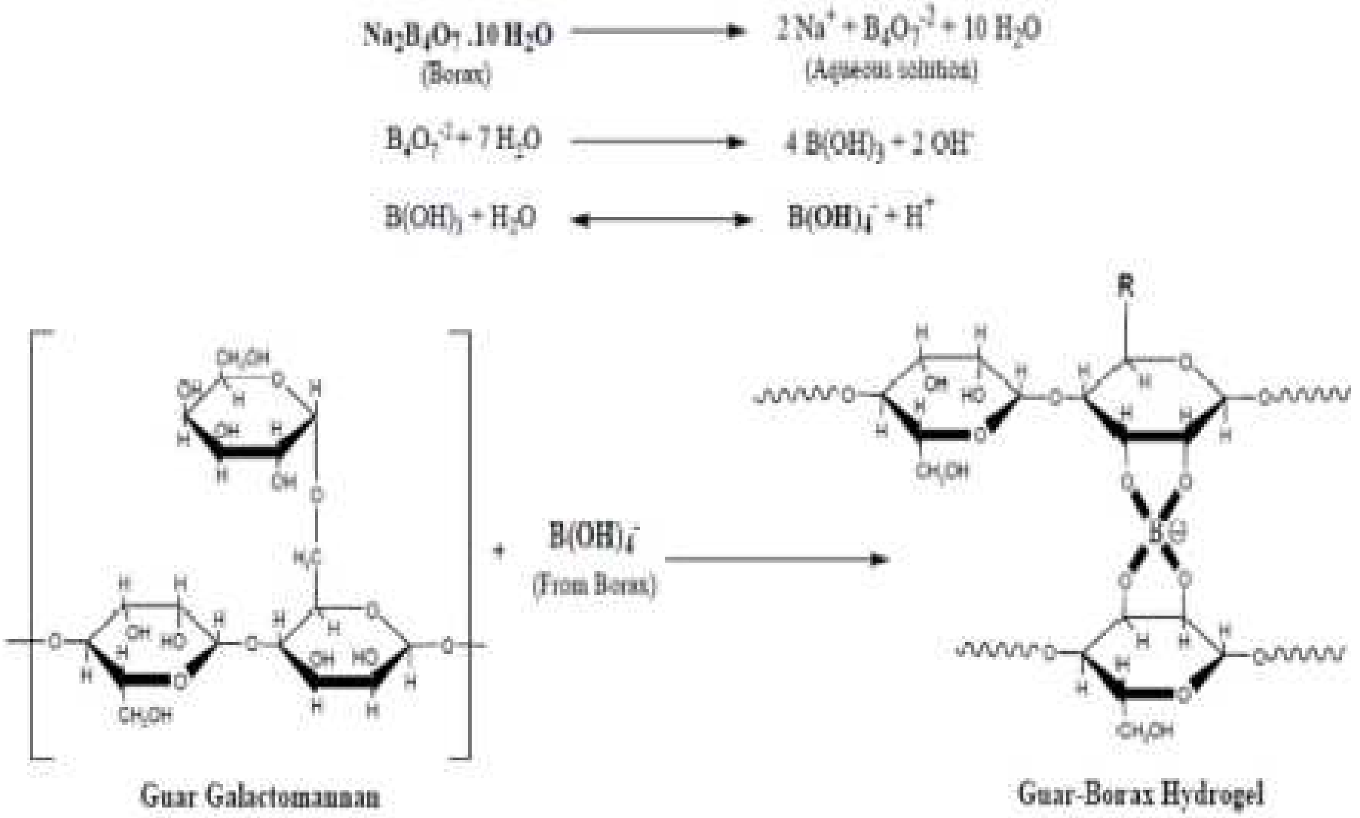
Capped glass measuring cylinders 100ml filled with the emulsified water sample to the mark, the made-up solutions of purified guar gum, synthetically modified guar gum, industrial reverse demulsifier and blank (PGG, GCB1, GCB2, GCB3, GCB4, CGB5, CGBB6, Industrial reverse demulsifier, Blank) respectively.

The flocculation efficiency of various polymer samples was carried out using modified Jar Test. An emulsified water samples in the prescribed tubes are dosed with (2.5, 5, and 10) µL i.e. (25, 50, and 100) ppm. all tubes were shacked 50 slow rolls and transferred to 60C° water bath and rated immediately after 30min (visually), screening test and its assessment recorded as (Oily, Not oily and Clear) based on best water quality, well-coalesced oil layer indicating good treatment. This was done for each of purified guar gum, synthetically modified guar gum products, Industrial reverse demulsifier and blank sample. After that, using a long needle 10 ml aliquot taken from the bottom of each tube and tested for oil in water using UV-VIS spectrophotometer.

Method

A. Synthesis of borax cross-linked guar gum hydrogels

To get a uniform and clod/fisheyes-free solution of purified guar gum, 0.505g, 0.500g, 0.502 of purified guar gum powder was taken in an absolute dry 250 ml beaker. 50 ml of distilled water was added to the beaker and rapidly dispersed by stirring for 2 minutes. The solution was allowed to hydrate and attain maximum viscosity by keeping it as such for 2 hours at room temperature. This process could ensure a uniform solution of purified guar gum. Cross-linker borax was used at three different proportions i.e. 5, 15, and 25% of the weight of the purified guar gum, to get hydrogels with a range of properties (abbreviated as GGB 1, GGB 2, and GGB 3 respectively). Accordingly, 25, 75, and 125 mg of borax, separately dissolved in 30 ml of distilled water, was added to the purified gum solutions. The mixture was then stirred for 30 min with the help of a mechanical stirrer and kept aside for another 4 hours without stirring. The synthesized hydrogels were gently rinsed with distilled water to get rid of unreacted borax, crushed, and soaked overnight in acetone for dehydration. Dehydrated hydrogels were filtered through a nylon sieve and oven-dried at 45°C to a constant weight.



Mechanism of synthesis of borax cross-linked guar gum hydrogels

Figure 1 Reaction of borax and guar gum

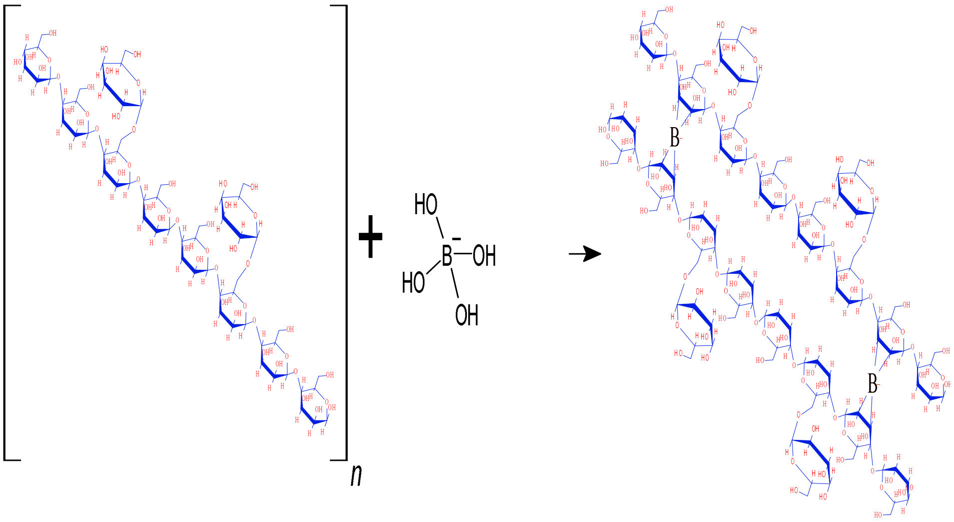


Figure 2 Crosslinking hydrogel

RESULTS AND DISCUSSION

The final product was involved in calculations for the purpose of fining the final product weight, estimation of reacted and unreacted borax and characterization using different techniques.

% of Borax crosslinked guar gum hydrogels = weight of purified polysaccharide ÷ weight of Borax cross-linked guar gum hydrogels × 100 ………………. (1)

Table 1 weights, percent and codes for the produced hydrogel in different grades

Hydrogel weight in gram	% of hydrogel	Product code
0.43	85.15	GCB1
0.46	92.00	GCB2
0.53	105.4	GCB3
0.504	100.8	GCB4
0.513	102.6	GCB5
0.499	99.8	GCB6

A. Estimation of unreacted and reacted borax

Estimation of unreacted borax was done by method given by [14], with some modifications. For this, 100 mg of finely powdered hydrogel was dispersed in 100 ml distilled water at room temperature. 2-3 drops of methyl red indicator. The dispersion was then titrated against 0.01M HCl solution. Color change from yellow to pink was recorded as an endpoint. The reading obtained for guar gum solution was considered blank and subtracted from the readings for hydrogels.

For estimation of crosslinked borax, the dispersion was pre-heated at 70°C to facilitate the breaking of crosslinked bonds, followed by similar titration. Considering one mole of borax equivalent to 2 moles of HCl, the amount of unreacted and crosslinked borax was calculated from the volume of HCl solution consumed.

$$2HCl + Na_2B_4O_7.10H_2O \longrightarrow 2NaCl + 4H_3BO_3 + H_2O$$

Table 2 Estimation of unreacted and crosslinked

Sample	A	V1	B	C	V2	D
Guar gum	3.6	-	-	-	-	-
GGB 1	1.51	1.15	0.219	1.85	0.34	0.065
GGB 2	2.65	2.29	0.436	3.46	0.45	0.086
GGB 3	4.39	4.03	0.768	6.58	1.83	0.26
Guar gum	0.4	-	-	-	-	-
GCB 4	8.86	8.46	1.61	9.31	0.45	0.086
GCB 5	9.5	9.1	1.74	10.2	0.7	0.13
GCB 6	11.65	11.25	2.15	12.47	0.82	0.16

borax in GGB by back titration
A= Vol. of HCl consumed before heating
B= Weight of Unreacted Borax in 100mg
C= Vol. of HCl consumed after heating
D = Weight of crosslinked Borax in 100mg

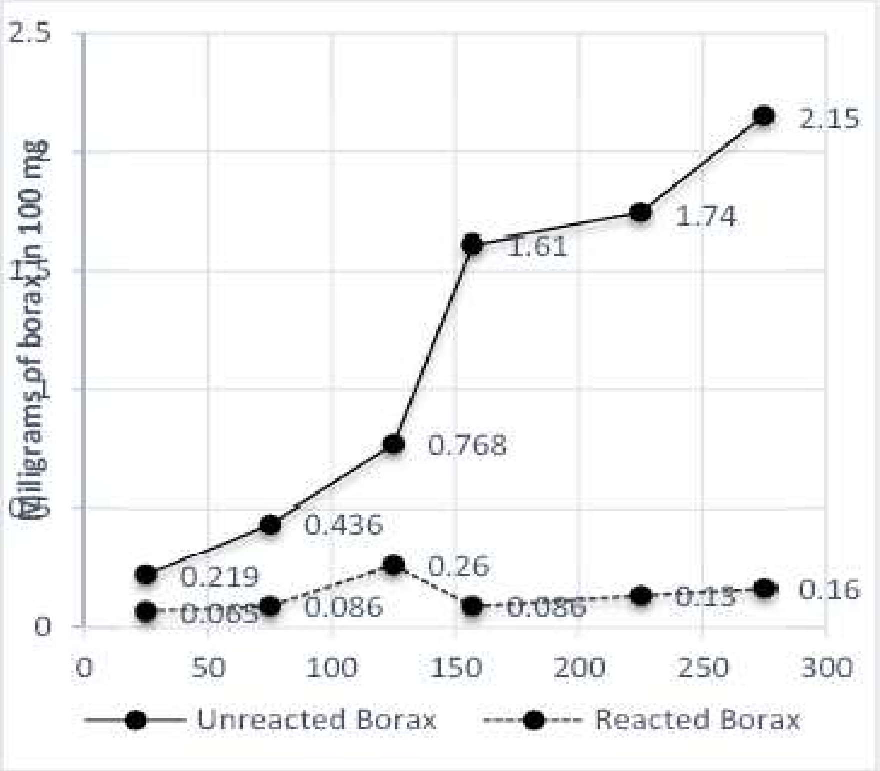


Figure 3 Reacted and unreacted borax

B. Characterization

Fourier transform infrared (FT-IR) spectroscopy

Generally, FTIR spectrum of borax crosslinked guar gum products show a broad peak, related to purified guar gum at around (3400 cm^{-1}) and it has reduced gradually across products (GCB1- GCB6) as expected, which is very clear in GCB3, also there is a slight shift towards a higher wavelength (3420 cm^{-1}), this is attributed to the consumption of hydroxyl groups of galactomannan backbone and the formation of covalent bonds with borax, also the absorption bands around (1645 cm^{-1}) (H-OH bending) and the band around (1400 cm^{-1}) (C-H bending) drastically weakened across products but it didn't disappear, which noticed clearly in the high amount of reacted borax in GCB3 also, that with the higher crosslinking percent.

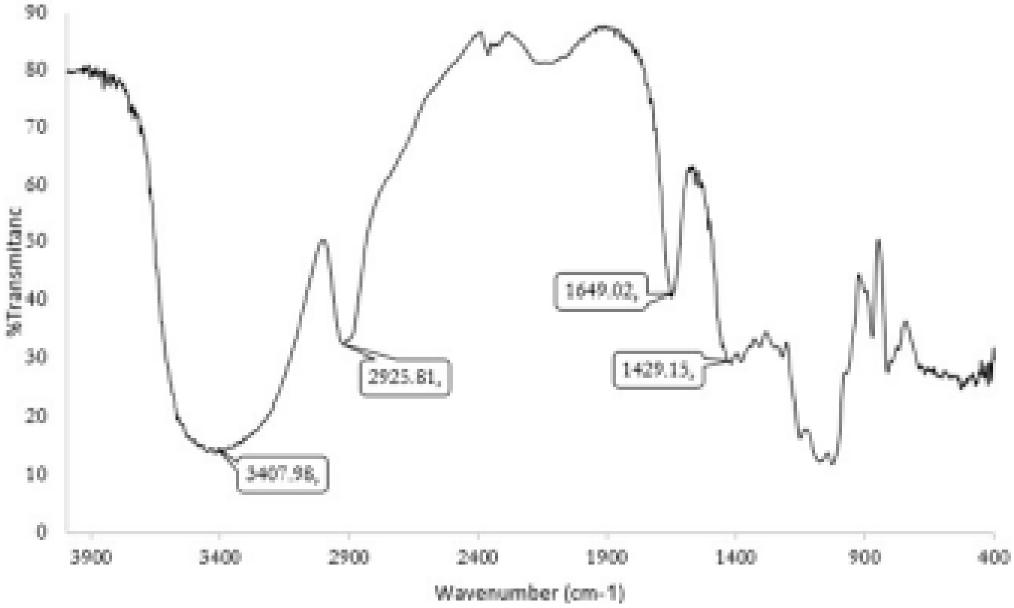


Figure 4 FT- IR spectrum for PGG

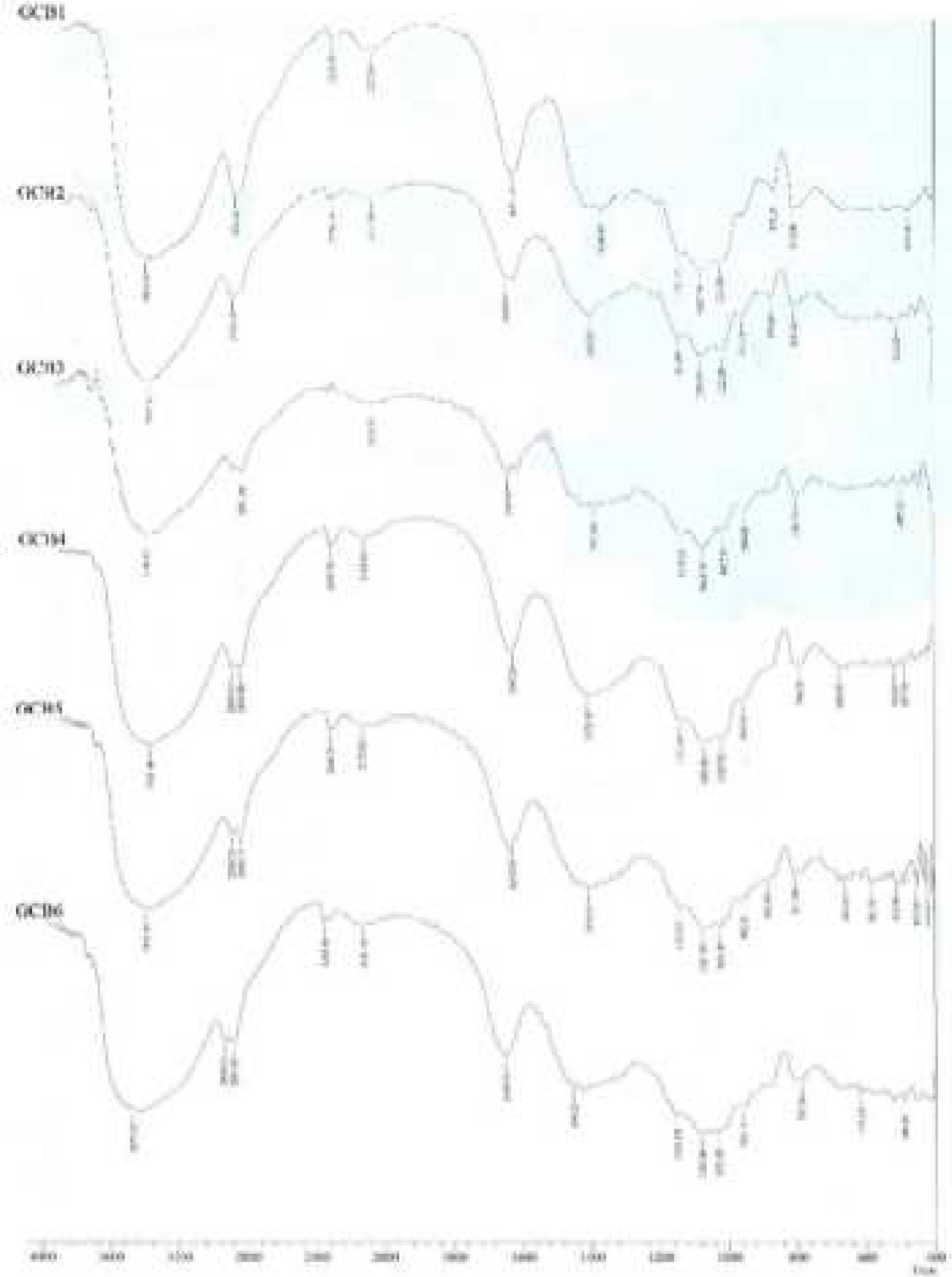


Figure 5 FT- IR spectrum for (GCB) products

Elemental analysis

The table below show the elemental content of borax crosslinked guar gum. It can be seen in the table that the contents of (C, H) decreased slightly compared to starting purified guar gum due to incorporation in crosslinking. Meanwhile, the content of (O) increased due to the borate group presence, on the other hand (N) content was very small. These result indicated that borax crosslinked guar gum, might have took place on (OH) sites of purified polysaccharide.

Table 3 Elemental analysis result for borax crosslinked guar gum hydrogel (GCB) products

Sample	C,%	H,%	N,%	O,%
PGG	38.98	7.49	1.14	52.39
GCB1	-	-	-	-
GCB 2	36.71	7.65	1.97	53.67
GCB 3	35.75	7.14	0.900	56.21
GCB 4	32.04	6.90	1.38	59.68
GCB 5	32.41	6.67	1.39	59.53
GCB6	32.91	6.78	1.5	58.81

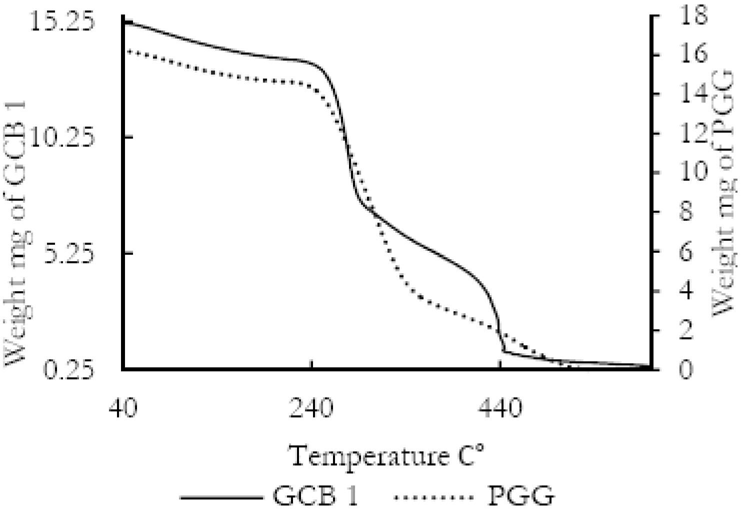


Figure 6 Thermogram of GCB1

Thermal gravimetric analysis (TGA)

According to literature for borax crosslinked guar gum hydrogel, four zones of weight loss were observed. First region (25–280 C°) was assigned to initial moisture loss and second zone from (280–330C°) related to degradation of main backbone of guar gum. The third zone (330–508C°) can be attributed to degradation of the crosslinked bonds between borax and hydroxyl bonds present in the hydrogel network which was followed by the fourth region of complete combustion [1].

Thermogram of PGG show three zones of decomposition while GCB products thermogram reflects three, four and five zones put two major zones appeared in all six GCB products (~270, ~450) C° with major weight losses which were missing in PGG thermogram, also GCB products started to decompose at higher temperature (~ 35) C° and mainly GCB exhibit lower weight loss percentage which give rise to an assumption of more thermal stability.

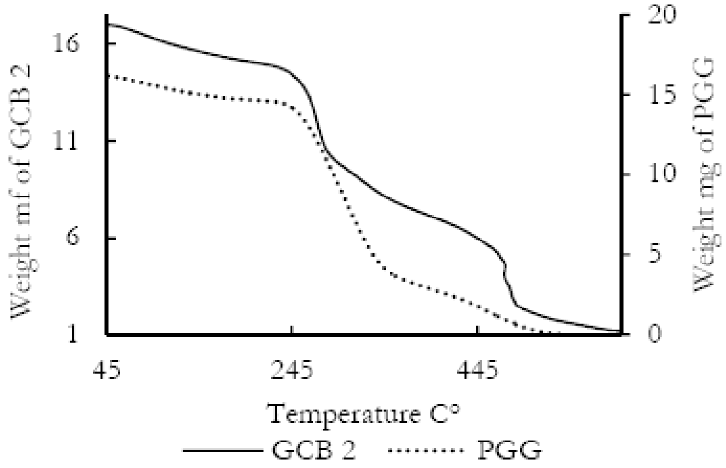


Figure 7 Thermogram of GCB2

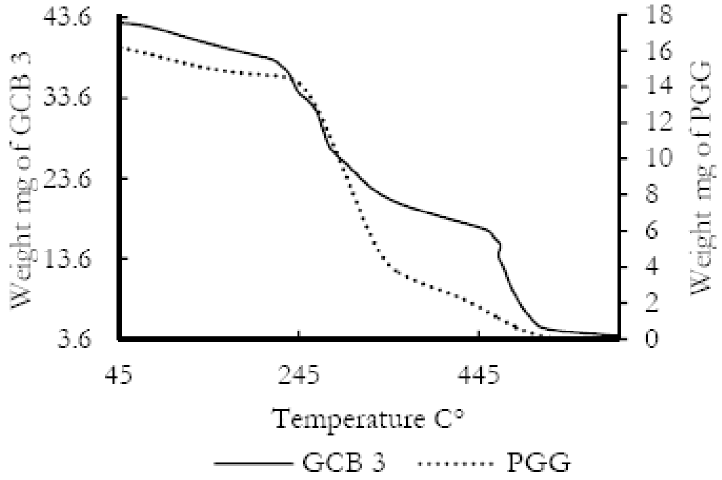


Figure 8 Thermogram of GCB3

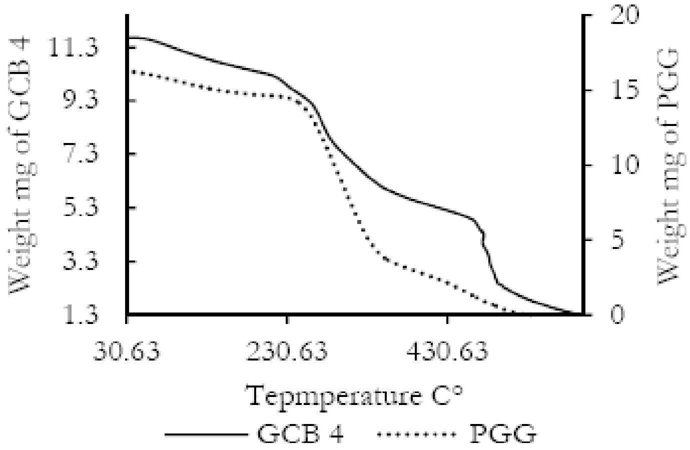


Figure 9 Thermogram of GCB4

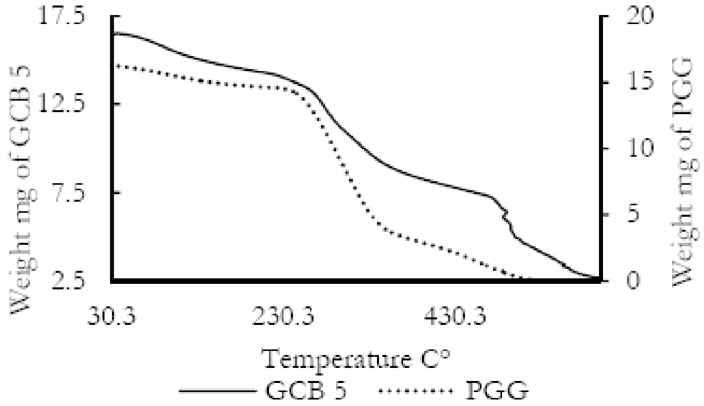


Figure 10 Thermogram of GCB5

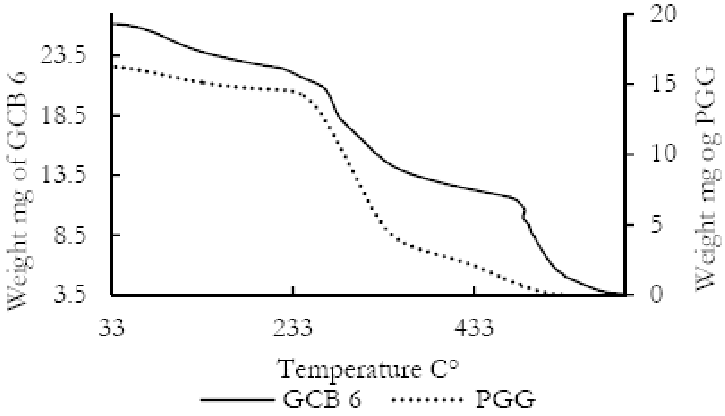


Figure 11 Thermogram of GCB6

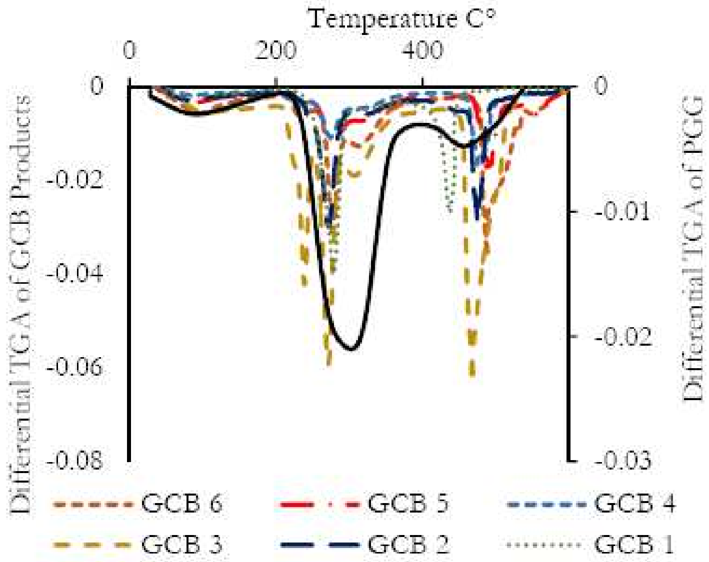


Figure12 Differential thermogram of (GCB) products

Scanning electron microscopy (SEM)

Borax crosslinked guar gum hydrogel (GCB) products is shown in Fig.45 (b, c, d, e and f) at 2000x and investigated for its morphological features against purified guar gum Fig.45 (a). Borax crosslinked guar gum hydrogels showed remarkable change in surface morphology, its uniformed but rough, scaly and fractured surface with considerable pore spaces and cracks in between, unlike its starting material purified guar gum, on the other hand, porous structure and roughness decreases with the decrease of hydrogen content from GCB2-CGB6 which accompanied with higher crosslinking (reacted borax) values. Crosslinking contributes to aqueous fluid diffusion in the polymeric network, producing higher swelling capacity in the final hydrogel. Also the distributed pore spaces in the surface morphology of the GCB hydrogel suggest a homogeneous composition of the absorbent material [1].

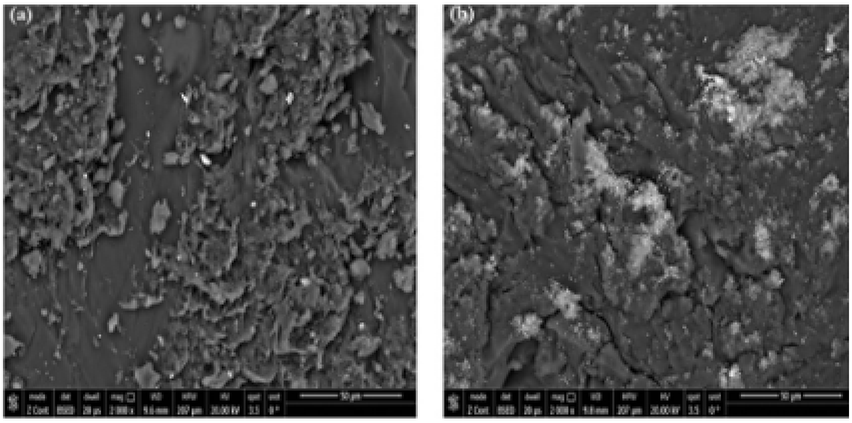


Figure 13 Scanning electron micrographs of purified guar gum (a), GCB2 (b) at (2000x).

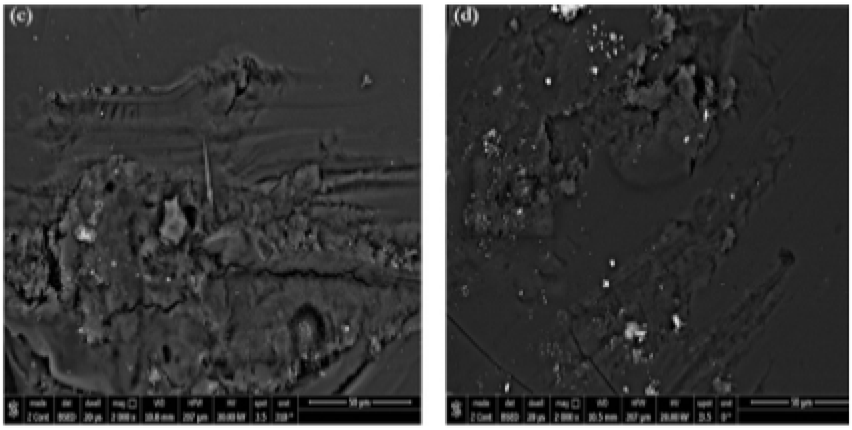


Figure 14 Scanning electron micrographs of GCB (c), GCB4 (d) at (2000x).

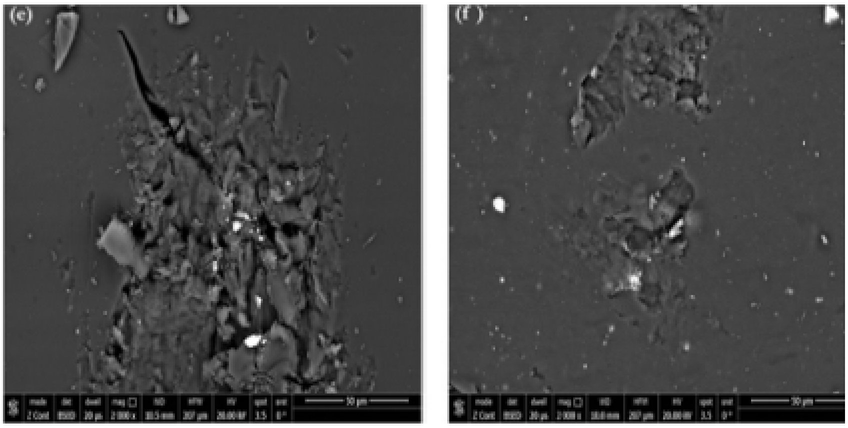


Fig.15 Scanning electron micrographs of GCB5 (e), GCB6 (f) at (2000x).

Rheological behavior

Borax crosslinking guar gum causes the structure of the long branched chain of purified guar gum to produce gel but on dilution a lower viscosity product solutions prepared for evaluating its rheological behavior. Respectively (GCB1, GCB4, GCB5, GCB6) and (GCB2, GCB3) solution diluted to about 0.3% was studied at 25C°, Spindle-21, (%Torque –24.1, 24, 26.7, 22.4 at 250 rpm and 27.3, 27.7 at 150 rpm). Density for the same solutions was measured using density bottle and gave the values below.

Table 4 Viscosity and density of (GCB) product and PGG

Sample	Viscosity in centipoise(cP) at 25C°	Density g\cm3
PGG	34	0.3757
GCB1	4.54	1.009
GCB 2	8.5	1.002
GCB 3	8.6	1.007
GCB 4	4.5	1.017
GCB5	5.01	1.002
GCB6	4.2	1.009

From the above results we can conclude that diluting borax crosslinking products to about (0.3%) generate a low viscosity values comparing to PGG viscosity value and "GCB3" the product of highest reacted borax percent also exhibits highest viscosity value.

Visual and spectrophotometric evaluation

With a dose of 2.5µL or 25ppm, visual evaluation reveled a slight difference could be seen across samples except for the sample (Industrial reverse demulsifier) which show no oil droplets on the glass sides and looked clear, samples show some oil droplets on the glass sides are described as not-oily like (GCB5 and GCB6), on the other hand, very high oil-in-water concentrations appeared in UV-VIS spectrophotometer readings for almost all samples, but not higher than the blank sample which might not give a sing for emulsion formation.

With a dose of 5µL or 50ppm, visual evaluation reveled there is a slight difference could be seen across samples except for sample (Industrial reverse demulsifier) which show no oil droplets on the glass sides and looked clear, samples show some oil droplets on the glass sides are described as not-oily like (GCB1 and GCB6) but high oil-in-water concentrations appeared in UV-VIS spectrophotometer readings for samples (GCB4, GCB3, and PGG) but not higher than blank which might give a sing for no emulsion formation but in contrast almost all products developed some emulsion breaking characteristics.

With a dose of 10µL or 100ppm, visual evaluation reveled there is a slight difference could be seen across samples except for sample except for sample (Industrial reverse demulsifier) which show no oil droplets on the glass sides and looked clear, samples show some oil droplets on the glass sides are described as not-oily like (GCB3, GCB4 and GCB5) but a high oil-in-water concentrations appeared in UV-VIS spectrophotometer readings of sample (GCB2) and even higher than the blank sample which might give a sing for emulsion formation.

Figure 16 visual evaluation of 25 ppm dose deoiling efficiency

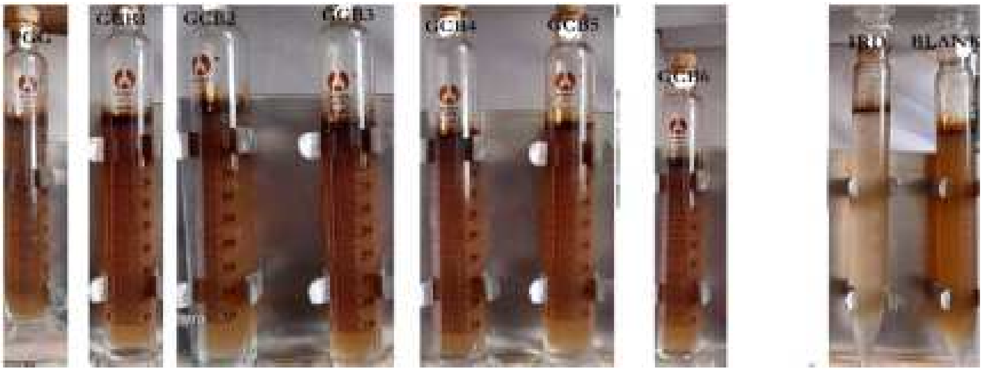


Figure 17 visual evaluation of 50 ppm dose deoiling efficiency

Figure 18 visual evaluation of 100 ppm dose deoiling efficiency

Table 5 Spectrophotometric evaluation of purified guar gum and its derivatives deoiling efficiency

Sample code - Dilution %	Doses in µL	O/W in PPm	De-oiling Efficiency
PPG - 0.04%	2.5	320	13.3
	5	277.5	18.4
	10	244.8	0
GCB1 - 0.3%	2.5	286.6	22.3
	5	218.6	35.8
	10	303	2.2
GCB2- 0.3%	2.5	308	16.5
	5	217.5	36.2
	10	330.5	0
GCB3- 0.3%	2.5	326.6	11.5
	5	282	17
	10	251	19
GCB4- 0.3%	2.5	362.2	1.8
	5	296	12.9
	10	258	16.8
GCB5- 0.3%	2.5	349	5.4
	5	205	39.7
	10	255	17.7
GCB6- 0.3%	2.5	352	4.6
	5	115	66.2
	10	201	35.2
IRD	2.5	40	89.1
	5	58.8	82.8
	10	44	85.5
Blank	2.5	369	0
	5	340	0
	10	310	0

Generally, its worthy to say that best results of oil in water concentration and hence highest deoiling efficiency was accomplished with the Industrial reverse demulsifier, but for guar gum products the efficiency have been noticed with a dose of 2.5µL or 25ppm of (GCB1) product which gives a deoiling efficiency of (22.3) and with a dose of 5µL or 50ppm of (GCB6) product, the deoiling efficiency increased to reach (66.2) and finally with a dose of 10µL or 100ppm of (GCB6) which gave an efficiency of (35.2). Hence we can conclude that (GCB6) are the synthetically modified guar gum product with the best reverse demulsification (deoiling) properties.

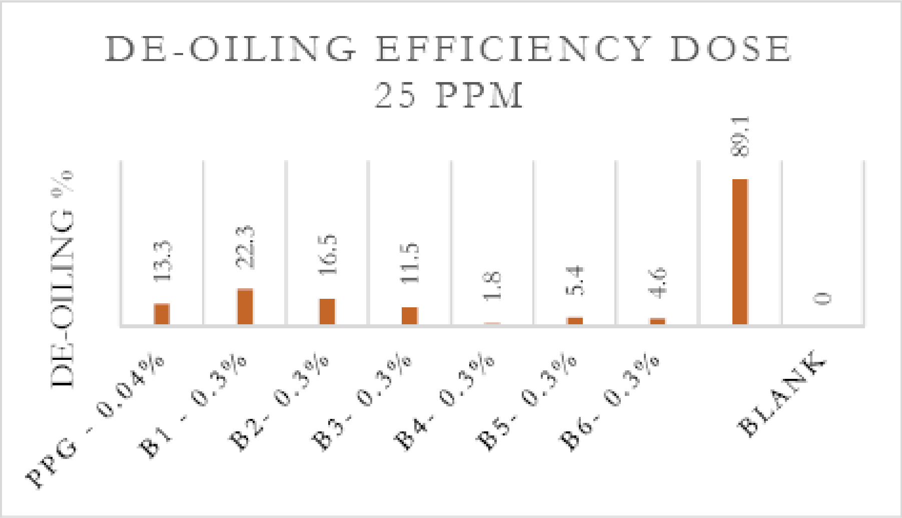


Figure 19 De-oiling efficiency of 25 ppm

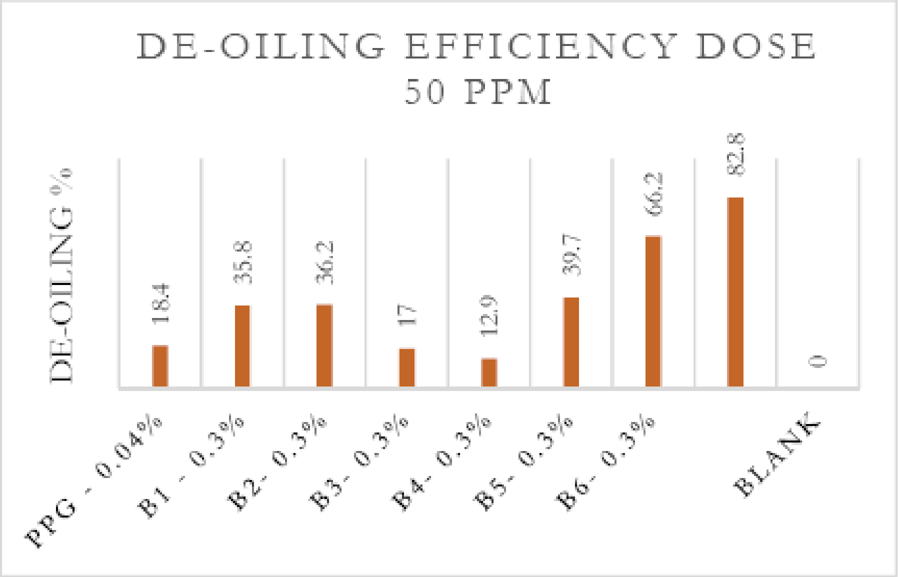


Figure 20 De-oiling efficiency of 50 ppm

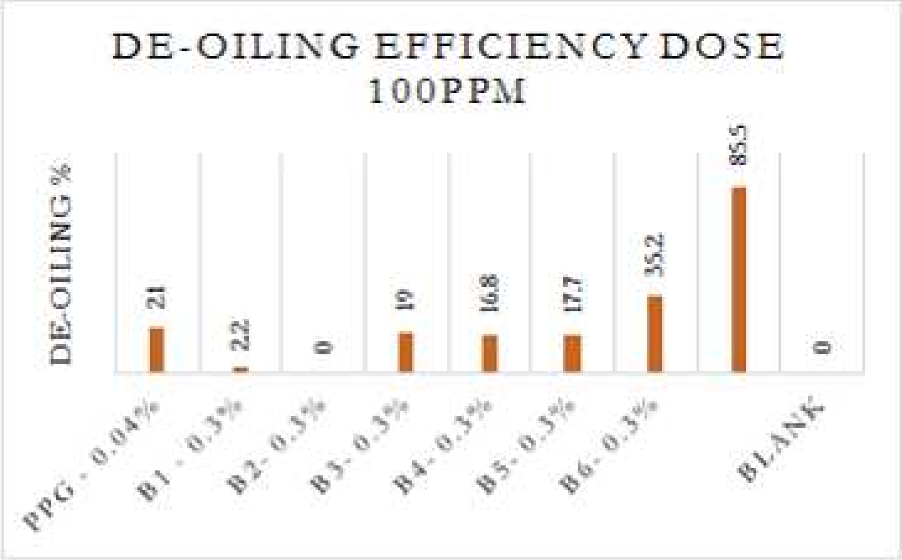


Fig.21 De-oiling efficiency of 100 ppm

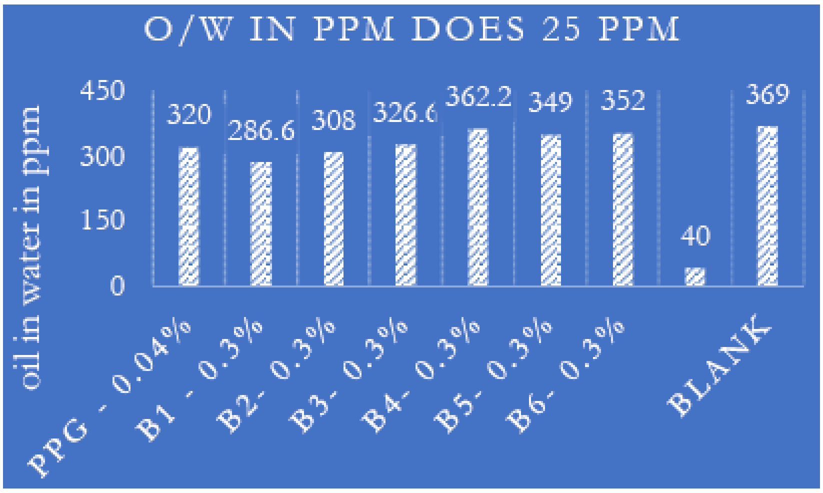


Figure 22 (oil in water) results 25ppm

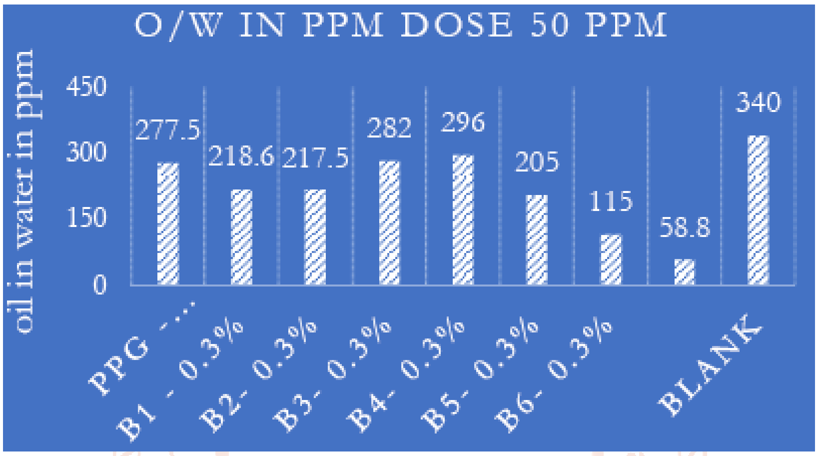


Figure 23 (oil in water) results 50 ppm

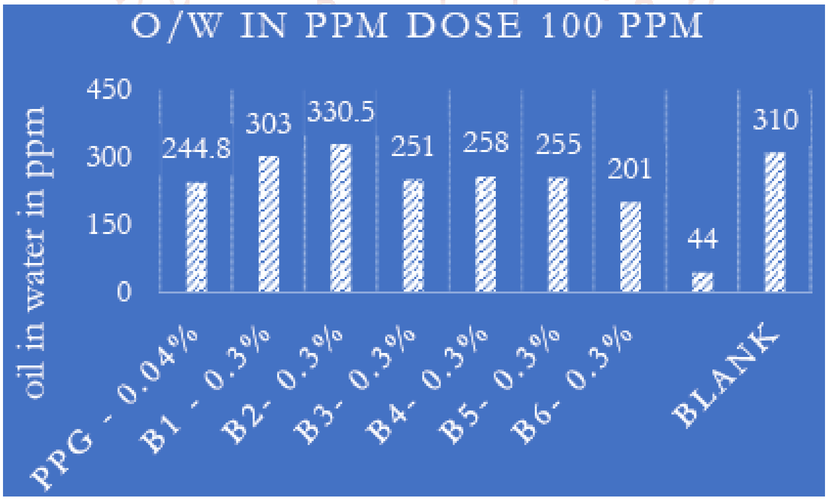


Figure 24 (oil in water) results 100 ppm

Conclusion

In this article, we have tried to explore structure, chemical and physical aspects of guar gum crosslinked product to prove crosslinking accomplishment. Easy to form products and much diluted solutions for reverse-demulsification is favored which is the case with borax crosslinked guar gum. With this method, these hydrogels can be conveniently and economically produced at larger scale. Cross-linking with borax has improved flocculation efficiency of guar gum noticeably. Out of different grades of GGB hydrogels, GGB6 at 50ppm dose has emerged as best dose.

It is a natural, harmless, renewable, low cost, highly viscous, and pH compatible polysaccharide which may easily blend into supportable products. Fine grades of guar gum or preferentially purified guar gum can be more developed and utilized in reverse-demulsification (deoiling) processes throw borax crosslinking and benefited from some of its preserved biodegradability. Also guar gum can act as the preferred choice for different areas of research interests.

References

[1] N. Thombare, U. Jha, S. Mishra, and M. Z. Siddiqui, "Borax cross-linked guar gum hydrogels as potential adsorbents for water purification," *Carbohydr. Polym.*, vol. 168, pp. 274–281, 2017, doi:10.1016/j.carbpol.2017.03.086.

[2] P. Bhattacharya, Amit Rawlins, James W. Ray, A. Bhattacharya, J. W. Rawlins, and P. Ray, *POLYMER GRAFTING AND CROSSLINKING*. 2009.

[3] S. Thakur, P. P. Govender, M. A. Mamo, S. Tamulevicius, Y. K. Mishra, and V. K. Thakur, "Progress in lignin hydrogels and nanocomposites for water purification: Future perspectives," *Vacuum*, vol. 146, pp. 342–355, 2017.

[4] M. Leonardis, A. Palange, R. F. V Dornelles, and F. Hund, "Use of cross-linked carboxymethyl cellulose for soft-tissue augmentation: preliminary clinical studies," *Clin. Interv. Aging*, vol. 5, p. 317, 2010.

[5] A. S. Patra, S. Ghorai, D. Sarkar, R. Das, S. Sarkar, and S. Pal, "Anionically functionalized guar gum embedded with silica nanoparticles: an efficient nanocomposite adsorbent for rapid adsorptive removal of toxic cationic dyes and metal ions," *Bioresour. Technol.*, vol. 225, pp. 367–376, 2017.

[6] V. K. Thakur, M. K. Thakur, P. Raghavan, and M. R. Kessler, "Progress in green polymer composites from lignin for multifunctional applications: a review," *ACS Sustain. Chem. Eng.*, vol. 2, no. 5, pp. 1072–1092, 2014.

[7] B. S. Kaith, R. Jindal, H. Mittal, and K. Kumar, "Temperature, pH and electric stimulus responsive hydrogels from Gum ghatti and polyacrylamide-synthesis, characterization and swelling studies," *changes*, vol. 25, p. 27, 2010.

[8] D. Banerjee and J. C. Williams, "Perspectives on titanium science and technology," *Acta Mater.*, vol. 61, no. 3, pp. 844–879, 2013.

[9] M. Prabaharan, "Prospective of guar gum and its derivatives as controlled drug delivery systems," *Int. J. Biol. Macromol.*, vol. 49, no. 2, pp. 117–124, 2011.

[10] S. Pal *et al.*, "Efficient and rapid adsorption characteristics of templating modified guar gum and silica nanocomposite toward removal of toxic reactive blue and Congo red dyes," *Bioresour. Technol.*, vol. 191, pp. 291–299, 2015.

[11] V. Singh, P. Kumari, S. Pandey, and T. Narayan, "Removal of chromium (VI) using poly (methylacrylate) functionalized guar gum," *Bioresour. Technol.*, vol. 100, no. 6, pp. 1977–1982, 2009.

[12] C. H. M. Roberts, "Use of treating compounds for oil field emulsions in the mid-continent field," *Trans. AIME*, no. 01, pp. 321–334, 1926.

[13] T. Balson *et al.*, "The development and application of dithiocarbamate (DTC) chemistries for use as flocculants by North Sea operators," in *Chemistry in the Oil Industry VII*, 2002, pp. 149–162.

[14] P. J. Subrahmanyam, "Design and development of guar gum and borax crosslinked guar gum matrix tablets of theophylline for colon specific drug," *J. Chem. Pharm. Res.*, vol. 4, no. 2, pp. 1052–1060, 2012.