Arabinose based gelators: Rheological characterization of the gels and

phase selective organogelation of crude-oil

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1. Gelation Tests

Gelation test were carried out by adding exact weights of compounds **3a-3g** individually to 1 ml of appropriate solvent in a vial. The vial was sealed and suspension was heated to dissolve the compound to get a clear solution. The solution was allowed to cool after which gelation was tested by inverting the sample vial. If the inverted vial was able to hold the system, it was considered as a gel. Apart from **3a** and **3g** the other triazoylarabinoside derivatives **3b**, **3c-f** were also tested in the same manner but they were not able to form gels in any solvent.

2. Determination of MinimumGelation Concentration(MGC)

1 ml of solvent was taken in 5 ml of sample vial, 1 mg of gelator **3a** and **3g** was added to the sovent which was then heated till aclear solution was obtained. Then the solution was cooled and the vial inverted to confirm gelation. If partial or no gelation was observed, the cycle was repeated adding 1 mg of **3a** or **3g** at the beginning of each heating cooling cycle till complete gelation of the solvent was observed by inversion of the vial.

3. Gel characterization

3.1 Optical Microscopy

An optical microscope (Olympus - CH20i) equipped with a digital camera (Nikon - Eclipse E200 MV Pole) for digital imaging was used for analyzing the microstructure of the organogels. The experiments were carried out by placing a small amount of the gel sample at a particular concentration on a 3 inch x 2 inch glass slide and viewing it with the microscope.

3.2 Field Emission Scanning Electron Micrographs (FESEM)

The experiments were performed by using a Zeiss supra-55 FESEM. The xerogels of the samples were prepared by dropcasting the hot 1% (w/v) solution of gelator **3a**or**3g** in gelling solvent on a glass slide (2mm x 2mm) and drying them overnight in air inside a vacuum dessicator. The xerogelwas then placed on a stub which was then coatedwithgold by a Quorum -Q150RES sputter coater undervacuum of 5×10^{-5} milibar and a current of 20 mA for 2 minutes.

3.4 Atomic force microscopy (AFM)

The experiments were performed by using a Bruker Dimension Icon instrument. The samples were prepared by dropcasting adilute solution of gelator **3a** and **3g** in *m*-xylene solvent on a

glass slide (2mm x 2mm) and drying them overnight under vacuum inside a dessicator.AFM images of the samples were obtained usingTapping Mode at 1 Hz scanning rate with a silicon cantilever tip (RFESP-MPP-21100-10) at a resonance frequency of 75 kHz and a spring constant of 3 Nm⁻¹.

3.5 Wide Angle X-ray Diffraction (WXRD)

The xerogels of the sample were prepared by dissolcing 3a (100 mg) and 3g (100 mg) in 10 mL of Benzene in a beaker and drying them overnight in a vacuum dessicator. The WXRD diffractogram of the samples were recorded on a ProroAXRD diffractometer. X-rays of wavelength 1.54 A° were used.

| Compound | 3a (MGC, ^b T _g) | 3b | 3c | 3d | 3e | 3f | 3g(MGC, ^b T _g) | | |
|--|--|----|----|----|----|----|---------------------------------------|--|--|
| | | | | | | | | | |
| Solvent | | | | | | | | | |
| Benzene | G, (1.0%, 44-45 °C) | S | S | S | S | S | G, (0.7%, 49-50 °C) | | |
| Toluene | G, (1.0%, 46-47 °C) | S | S | S | S | S | G, (0.5%, 53-54 °C) | | |
| o-xylene | G, (0.9%, 47-48 °C) | S | S | S | S | S | G, (0.5%, 57-58 °C) | | |
| <i>m</i> -xylene | G, (0.7%, 48-49 °C) | S | S | S | S | S | G, (0.5%, 52-53 °C) | | |
| <i>p</i> -xylene | G, (0.9%, 46-47 °C) | S | S | S | S | S | G, (0.5%, 51-52 °C) | | |
| Chlorobenzene | G, (1.0%,45-46 °C) | S | S | S | S | S | G, (0.7%, 53-54 °C) | | |
| Hexane | 1 | I | Ι | Ι | I | Ι | 1 | | |
| <i>n</i> -heptane | 1 | I | Ι | Ι | I | Ι | 1 | | |
| Cyclohexane | 1 | I | I | Ι | I | Ι | 1 | | |
| Methylcyclohexane | 1 | I | Ι | Ι | I | Ι | 1 | | |
| DCM | S | S | S | S | S | S | S | | |
| Chloroform | S | S | S | S | S | S | S | | |
| Ethyl acetate | S | S | S | S | S | S | S | | |
| Aceto nitrile | S | S | S | S | S | S | S | | |
| Ethanol | G, (1.0, 48-49 °C) | S | S | S | S | S | S | | |
| Methanol | S | S | S | S | S | S | S | | |
| n-butyl alcohol | S | S | S | S | S | S | S | | |
| Amyl alcohol | S | S | S | S | S | S | S | | |
| Water | I | I | I | Ι | I | I | 1 | | |
| DMF | S | S | S | S | S | S | S | | |
| DMSO | S | S | S | S | S | S | S | | |
| Kerosene | G (0.3%, 69-70 °C) | S | S | S | S | S | G (0.3%, 71-72 °C) | | |
| Petrol | G, (0.3%, 63-64 °C) | S | S | S | S | S | G, (0.3%, 61-62 °C) | | |
| Diesel | G, (0.3%, 66-67 °C) | S | S | S | S | S | G, (0.3%, 68-69 °C) | | |
| ^a G = Gel; S = Solution; I = Insoluble. ^b (w/v). | | | | | | | | | |

4 Table S1. Gelation ability of **3a-g** with various solvents.^a

5 Figures and graphs

5.1 Figures of gels at various concentrations



Figure S1.Gels from Benzene and **3a** at various concentrations of **3a**(a) at 1.0% (w/v) (b) at 1.1% (w/v) (c) at 1.2% (w/v) (d) at 1.3% (w/v) (e) at 1.4% (w/v).



Figure S2.Gels from tolueneand**3a** at various concentrations of **3a** (a) at 1.0% (w/v) (b) at 1.1% (w/v) (c) at 1.2% (w/v) (d) at 1.3% (w/v) (e) at 1.4% (w/v).



Figure S3.Gels from *o*-xylene and**3a** at various concentrations of **3a** (a) at 0.9% (w/v) (b) at 1.0% (w/v) (c) at 1.1% (w/v) (d) at 1.2% (w/v) (e) at 1.3% (w/v).



Figure S4.Gels from *m*-xylene and **3a** at various concentrations of **3a** (a) at 0.7% (w/v) (b) at 0.8% (w/v) (c) at 0.9% (w/v) (d) at 1.0% (w/v) (e) at 1.1% (w/v).



Figure S5.Gels from *p*-xylene and**3a** at various concentrations of **3a** (a) at 0.9% (w/v) (b) at 1.0% (w/v) (c) at 1.1% (w/v) (d) at 1.2% (w/v) (e) at 1.3% (w/v).



Figure S6.Gels from chlorobenzene and 3a at various concentrations of 3a (a) at 1.0% (w/v) (b) at

1.1% (w/v) (c) at 1.2% (w/v) (d) at 1.3% (w/v) (e) at 1.4% (w/v).



Figure S7.Gels from ethanol and **3a** at various concentrations of **3a** (a) at 1.0% (w/v) (b) at 1.1% (w/v) (c) at 1.2% (w/v) (d) at 1.3% (w/v) (e) at 1.4% (w/v).



Figure S8.Gels from petrol and **3a** at various concentrations of **3a** (a) at 0.3% (w/v) (b) at 0.4% (w/v) (c) at 0.5% (w/v) (d) at 0.6% (w/v) (e) at 0.7% (w/v).



Figure S9.Gels from diesel and 3a at various concentrations of 3a (a) at 0.3% (w/v) (b) at 0.4% (w/v) (c)

at 0.5% (w/v) (d) at 0.6% (w/v) (e) at 0.7% (w/v).





Figure S10.Gels from Crude oil of **3a** at minimum gelation concentrations at 0.5% (w/v). (a) 1 ml Crude oil (b) Crude oil gel



Figure S11.Gels from benzene and **3g** at various concentrations of **3g**(a) at 0.7% (w/v) (b) at 0.8% (w/v) (c) at 0.9% (w/v) (d) at 1.0% (w/v) (e) at 1.1% (w/v).



Figure S12.Gels from toluene and **3g** at various concentrations of **3g**(a) at 0.5% (w/v) (b) at 0.6% (w/v) (c) at 0.7% (w/v) (d) at 0.8% (w/v) (e) at 0.9% (w/v).



Figure S13.Gels from *o*-xylene and**3g** at various concentrations of **3g**(a) at 0.5% (w/v) (b) at 0.6% (w/v) (c) at 0.7% (w/v) (d) at 0.8% (w/v) (e) at 0.9% (w/v).



Figure S14.Gels from *m*-xylene and **3g** at various concentrations of **3g**(a) at 0.5% (w/v) (b) at 0.6% (w/v) (c) at 0.7% (w/v) (d) at 0.8% (w/v) (e) at 0.9% (w/v).



Figure S15.Gels from *p*-xylene and **3g** at various concentrations of **3g**(a) at 0.5% (w/v) (b) at 0.6% (w/v) (c) at 0.7% (w/v) (d) at 0.8% (w/v) (e) at 0.9% (w/v).



Figure S16.Gels from chlorobenzene and**3g** at various concentrations of **3g**(a) at 0.7% (w/v) (b) at 0.8% (w/v) (c) at 0.9% (w/v) (d) at 1.0% (w/v) (e) at 1.1% (w/v).



Figure S17.Gels from petrol and **3g** at various concentrations of **3g**(a) at 0.3% (w/v) (b) at 0.4% (w/v) (c) at 0.5% (w/v) (d) at 0.6% (w/v) (e) at 0.7% (w/v).



Figure S18.Gels from diesel and **3g** at various concentrations of **3g**(a) at 0.3% (w/v) (b) at 0.4% (w/v) (c) at 0.5% (w/v) (d) at 0.6% (w/v) (e) at 0.7% (w/v).



Figure S19. Gelation of crude-oil with gelator 3g (a) Crude-oil (b) Gelled crude-oil.



5.2 Tables for variation of Tg with concentration

Figure S20. Variation of T_g with concentration for organogels of 3a.



Figure S21. Variation of T_g with concentration for organogels of 3g.

5.3 Optical microscopy images



Figure S22.Optical microscopy image for **(a) 3a** in *m*-xylene at 1% (w/v) **(b)3g** in *m*-xylene at 1% (w/v) concentration.

5.4 FESEM micrographs



Figure S23.FESEM image of xerogel of 3a at 1% (w/v) concentration in benzene.



Figure S24.FESEM image of xerogel of 3a at 1% (w/v) concentration in toluene



Figure S25.FESEM image of xerogel of 3a at 1% (w/v) concentration in o-xylene



Figure S26.FESEM image of xerogel of 3a at 1% (w/v) concentration in*m*-xylene



Figure S27.FESEM image of xerogel of 3a at 1% (w/v) concentration in *p*-xylene



Figure S28.FESEM imageofxerogel of 3a at 1% (w/v) concentration in chlorobenzene



Figure S29.FESEMimage of xerogel of 3a at 1% (w/v) concentration inethanol



Figure S30.FESEM image of xerogel of 3a at 1% (w/v) concentration inpetrol



Figure S31.FESEM image of xerogel of 3a at 1% (w/v) concentration in diesel



Figure S32.FESEM image of xerogel of 3g at 1% (w/v) concentration in benzene



Figure S33.FESEM image of xerogel of 3g at 1% (w/v) concentration in toluene



Figure S34.FESEM image of xerogel of 3g at 1% (w/v) concentration in o-xylene



Figure S35.FESEM image of xerogel of 3g at 1% (w/v) concentration in *m*-xylene



Figure S36.FESEM image of xerogel of 3g at 1% (w/v) concentration in *p*-xylene



Figure S37.FESEM image of xerogel of 3gat 1% (w/v) concentration chlorobenzene



Figure S38.FESEM image of xerogel of 3g at 1% (w/v) concentration petrol



Figure S39.FESEM image of xerogel of 3g at 1% (w/v) concentration diesel

5.5 AFM images



Figure S40.AFM image of 3a in *m*-xylene (a) 2-D image (b) 3-D image



Figure S41. AFM image of 3g in *m*-xylene (a) 2-D image (b) 3-D image

5.6 Rheology



Figure S42. (a) DSS cruve of **3a** gel with benzene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3a** gel with benzene at 1% (w/v) at strain 0.001% and temperature 25 °C (c) DTS curve of **3a** gel with benzene at 1% (w/v) at frequency 1 Hz and temperature 25°C.



Figure S43. (a)DSS cruve of **3a** gel with toluene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3a** gel with toluene at 1% (w/v) at strain 0.001% and temperature 25 °C (c)DTS curve of **3a** gel with toluene at 1% (w/v) at frequency 1 Hz and temperature 25°C



Figure S44. (a)DSS cruve of **3a** gel with *o*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3a** gel with *o*-xylene at 1% (w/v) at strain 0.001% and temperature 25 °C (c) DTS curve of **3a** gel with *o*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25°C.



Figure S45. (a)DSS cruve of **3a** gel with *m*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3a** gel with *m*-xylene at 1% (w/v) at strain 0.002% and temperature 25 °C (c)DTS curve of **3a** gel with *m*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25°C.



Figure S46. (a)DSS cruve of **3a** gel with *p*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3a** gel with *p*-xylene at 1% (w/v) at strain 0.001% and temperature 25 °C (c)DTS curve of **3a** gel with *p*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25°C.



Figure S47. (a)DSS cruve of **3a** gel with Chloro-benzene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3a** gel with Chloro-benzene at 1% (w/v) at strain 0.001% and temperature 25 °C (c)DTS curve of **3a** gel with Chloro-benzene at 1% (w/v) at frequency 1 Hz and temperature 25 °C.



Figure S48. (a) DSS cruve of **3a** gel with Ethanol at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3a** gel with Ethanol at 1% (w/v) at strain 0.003% and temperature 25 °C (c)DTS curve of **3a** gel with Ethanol at 1% (w/v) at frequency 1 Hz and temperature 25°C.



Figure S49. (a)DSS cruve of **3a** gel with Petrol at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3a** gel with Petrol at 1% (w/v) at strain 0.002% and temperature 25 °C (c)DTS curve of **3a** gel with Petrol at 1% (w/v) at strain 0.002% and temperature 25 °C (c)DTS curve of **3a** gel with Petrol at 1% (w/v) at frequency 1 Hz and temperature 25°C



Figure S50. (a)DSS cruve of **3a** gel with Diesel at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3a** gel with Diesel at 1% (w/v) at strain 0.002% and temperature 25 °C (c)DTS curve of **3a** gel with Diesel at 1% (w/v) at strain 0.002% and temperature 25 °C (c)DTS curve of **3a** gel with Diesel at 1% (w/v) at frequency 1 Hz and temperature 25°C



Figure S51. (a)DSS cruve of **3a** gel with Crude oil at 2% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3a** gel with Crude oilat 1% (w/v) at strain 0.002% and temperature 25 °C (c)DTS curve of **3a** gel with Crude oil at 1% (w/v) at frequency 1 Hz and temperature 25°C.



Figure S52. (a)DSS cruve of **3g** gel with benzene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3g** gel with benzene at 1% (w/v) at strain 0.001% and temperature 25 °C (c)DTS curve of **3g** gel with benzene at 1% (w/v) at frequency 1 Hz and temperature 25°C.



Figure S53. (a)DSS cruve of **3g** gel with toluene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3g** gel with toluene at 1% (w/v) at strain 0.001% and temperature 25 °C (c)DTS curve of **3g** gel with toluene at 1% (w/v) at frequency 1 Hz and temperature 25°C.



Figure S54. (a)DSS cruve of **3g** gel with *o*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3g** gel with *o*-xylene at 1% (w/v) at strain 0.002% and temperature 25 °C (c)DTS curve of **3g** gel with *o*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25°C.



Figure S55. (a)DSS cruve of **3g** gel with *m*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3g** gel with *m*-xylene at 1% (w/v) at strain 0.001% and temperature 25 °C (c)DTS curve of **3g** gel with *m*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25 °C.



Figure S56. (a)DSS cruve of **3g** gel with *p*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3g** gel with *p*-xylene at 1% (w/v) at strain 0.001% and temperature 25 °C (c)DTS curve of **3g** gel with *p*-xylene at 1% (w/v) at frequency 1 Hz and temperature 25 °C.



Figure S57. (a)DSS cruve of **3g** gel with chlorobenzene at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3g** gel with chlorobenzene at 1% (w/v) at strain 0.001% and temperature 25 °C (c)DTS curve of **3g** gel with chlorobenzene at 1% (w/v) at frequency 1 Hz and temperature 25°C.



Figure S58. (a)DSS cruve of **3g** gel with petrol at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3g** gel with petrol at 1% (w/v) at strain 0.001% and temperature 25 °C (c)DTS curve of **3g** gel with petrol at 1% (w/v) at strain 0.001% and temperature 25 °C (c)DTS curve of **3g** gel with petrol at 1% (w/v) at frequency 1 Hz and temperature 25°C.



Figure S59. (a)DSS cruve of **3g** gel with diesel at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3g** gel with diesel at 1% (w/v) at strain 0.002% and temperature 25 °C (c)DTS curve of **3g** gel with diesel at 1% (w/v) at frequency 1 Hz and temperature 25 °C.



Figure S60. (a)DSS cruve of **3g** gel with crude oil at 1% (w/v) at frequency 1 Hz and temperature 25 °C (b) DFS cruve of **3g** gel with crude oil at 1% (w/v) at strain 0.001% and temperature 25 °C (c)DTS curve of **3g** gel with crude oil at 1% (w/v) at frequency 1 Hz and temperature 25 °C.



Figure S61. Curves for thixotropic experiment for the meta-xylene gel at 1% (w/v) concentration for (a) 3a and (b) 3g.

5.7 PSOG of petrol, diesel and crude-oil



Figure S62. PSOG of petrol and recovery of congealed petrol using gelator **3a**. (a) Water (b) Biphasic mixture of petrol and water (c) congealed petrol layer (d) separated congealed petrol layer (e) transfer of congealed petrol into a flask (f) residual water after removal of congealed petrol (g) distillation set-up for recovery of petrol (h) petrol distillate (i) residual organogelator.



Figure S63. Phase selective gelation of diesel using **3a** in diesel (a) 4 ml of water. (b) Biphasic mixture of water and diesel (c) Congealed diesel layer (d) separated congealed diesel layer (e) transfer of congealed diesel gel into a flask (f) residue water after removal of congealed diesel (g) distillation setup for recovery of diesel (h) diesel distillate (i) residual organogelator.



Figure S64. Phase selective gelation of petrol using **3g** in petrol (a) 4 ml of water. (b) Biphasic mixture of water and diesel (c) Congealed petrol layer (d) separated congealed petrol layer (e) transfer of congealed petrol gel into a flask (f) residue water after removal of congealed petrol (g) distillation set- up for recovery of petrol (h) petrol distillate (i) residual organogelator.



Figure S65. Phase selective gelation of diesel using **3g** in diesel (a) 4 ml of water. (b) Biphasic mixture of water and diesel. (c) Congealed diesel layer (d) separated congealed diesel layer (e) transfer of congealed diesel gel into a dish. (f) residue water after removal of congealed diesel (g) distillation setup for recovery of diesel. (h) diesel distillate (i) residual organogelator.



Figure S66. Phase selective gelation of crude oil using **3g** (a) Water (b) Biphasic mixture of water and crude-oil (c) congealed crude-oil layer after addition of gelator (d) floating congealed crude-oil layer (e) removed congealed crude-oil (f) residual water

6 Spectra of 3a-3g







Figure S68. ¹³C NMR spectra of 3a in CDCl₃.



Figure S69. ¹H NMR spectra of 3b in CDCl₃.



Figure S70. ¹³C NMR spectra of 3b in CDCl₃.



Figure S71. ¹H NMR spectra of 3c in CDCl₃.



Figure S72. ¹³C NMR spectra of 3c in CDCl₃.



Figure S73. ¹H NMR spectra of 3d in CDCl₃.



Figure S74. ¹³C NMR spectra of 3d in CDCl₃.



Figure S75. ¹H NMR spectra of 3e in CDCl₃.



Figure S76. ¹³C NMR spectra of 3e in CDCI₃.



Figure S77. ¹H NMR spectra of 3f in CDCl₃.



Figure S78. ¹³C NMR spectra of 3f in CDCl₃.



Figure S79. ¹H NMR spectra of 3g in CDCl₃.

