UTL Al-IM-12 Si(84), Ge(15), Al(1)

Contributed by Jiri Čejka

Verified by R. Ryoo, A. Puškarić

Type Material: $(SDA)_4[Si_{64}Ge_{11}Al_1O_{152}]$: w H_2O
(SDA = 7-Ethyl-6-azoniaspiro[5.5]undecane hydroxide)

Method: M. V. Shamzhy, O. V. Shvets, M. V. Opanasenko, P. S. Yaremov, L. G. Sarkisyan, P. Chlubná, A. Zukal, V. R. Marthala, M. Hartmann, J. Čejka [1]

Batch Composition: 0.788 SiO_2 : 0.4 GeO_2 : 0.006 Al_2O_3 :0.45 SDAOH/Br : 30 H_2O

Source Materials
deionized water
sodium hydroxide (98%, NaOH)
1,5-dibromopentane (97 %)
2-ethylpiperidine (98%)
chloroform (99%)
sodium sulfate anhydrous (99 %, Na_2SO_4)
diethyl ether (99%)
Dowex® SBR LCNG, hydroxide form (Supelco)
aluminum hydroxide (r.g., $Al(OH)_3$)
germanium oxide (99.99 % GeO_2)
silica (Degussa Aerosil 200, or Cab-O-Sil M5)

Batch Preparation (for 3.5 g dry product)
(1) [33.1 g water + 1.34 g sodium hydroxide + 7.67 g 1,5-dibromopentane], stir in a flask
(2) [(1) + 3.80 g 2-ethylpiperidine], add 2-ethylpiperidine to (1) drop by drop over a period of 30 min under reflux; reflux for 12 h under vigorous stirring [a]; cool with an ice bath
(3) [8.3 g water + 8.3 g sodium hydroxide], stir until dissolved, cool with ice bath
(4) [(2) + (3)], stir, recover solid by filtration
(5) [solid (4) + 150 ml chloroform], stir until dissolved
(6) [(5) + 25 g sodium sulfate anhydrous], stir, left for 30 min, remove solid by filtration, evaporate about 80 ml of chloroform using rotovap
(7) [(5) + 250 ml diethyl ether], mix, recover solid by filtration, wash with diethyl ether, dry at ambient temperature for 12 h
(8) [solid (6)[b] + 36 g water + 30 g Dowex®], stir for 2 h, remove Dowex® by filtration
(9) [solution (7) + 0.062 g aluminum hydroxide], stir until dissolved[c]
(10) [solution (8) + 2.79 g germanium oxide], stir until dissolved[c]
(11) [(9) + 3.15 g Cabosil], stir for 30 minutes[d]

Crystallization
Vessel: Teflon-lined stainless steel autoclave
Temperature: 170° C
Time: 20 days

Agitation: 60 rpm

Product Recovery

(1) Dilute reaction mixture with water
(2) Filter and wash with water
(3) Dry at ambient temperature or at 90°C
(4) Yield: 3.5 g

Product Characterization

XRD: UTL; competing phase: STF (when Al / (Si + Ge +Al) > 0.015 or the pH > 12.0 in initial gel)
Elemental analysis: 84 SiO_2 : 15 GeO_2 : 0.5 Al_2O_3[e]
Crystal size and habit: thin platelet-like crystals

Reference

[1] M. V. Shamzhy, O. V. Shvets, M. V. Opanasenko, P. S. Yaremov, L. G. Sarkisyan, P. Chlubná, A. Zukal, V. R. Marthala, M. Hartmann, J. Čejka, J. Chem. Mater. 22 (2012) 15793
[2] M. Shamzhy, O. V. Shvets, M. V. Opanasenko, D. Procházková, P. Nachtigall, J. Čejka, Adv. Porous Mater. 1 (2013) 103

Notes

a. The satisfactory mixing of two phases at > 1000 rpm.
b. 90 % yield of SDA
c. Clear solution
d. pH of final gel is 12.0. The required amount of 1M SDAOH solution or 5 M HCl was added to the above mixture under stirring to adjust the pH of the gel.
e. According to Ref. [1] and [2] the chemical composition of the product depends on the pH of the final gel.