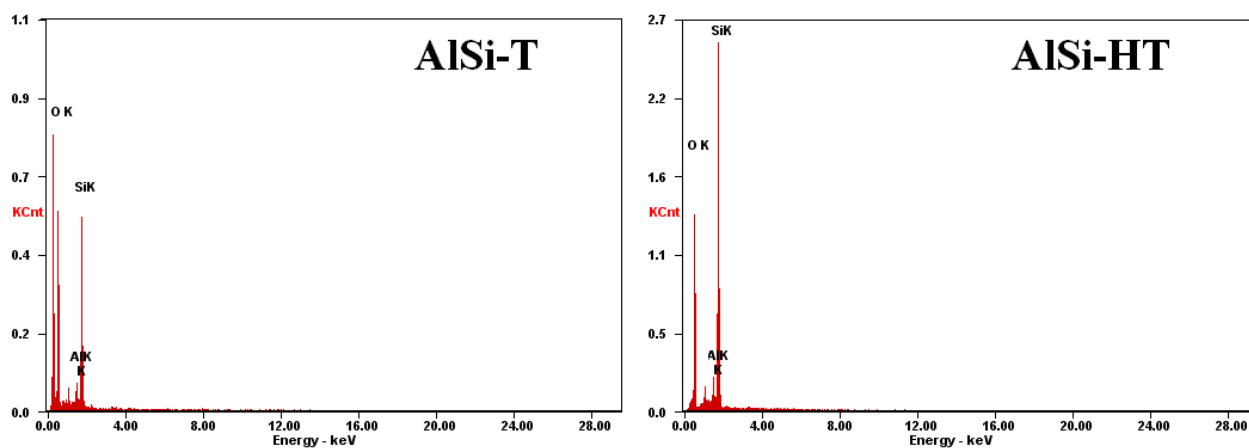# Facile synthesis of mesoporous aluminosilicate nano particles for selective production of N-Benzylidenaniline in solvent-free reaction of aniline with benzyl alcohol[†]
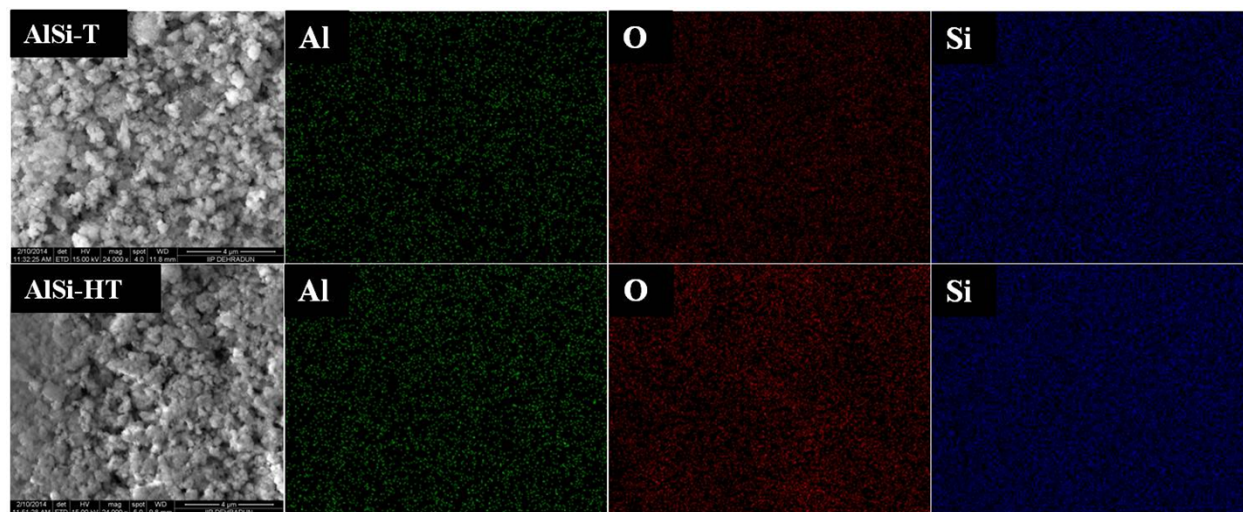
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**Supporting Figure 1:** EDX spectra of the AlSi-Tand AlSi-HT



**Supporing Figure 2:** SEM images and EDX elemental mapping of the AlSi-T and AlSi-HT samples.

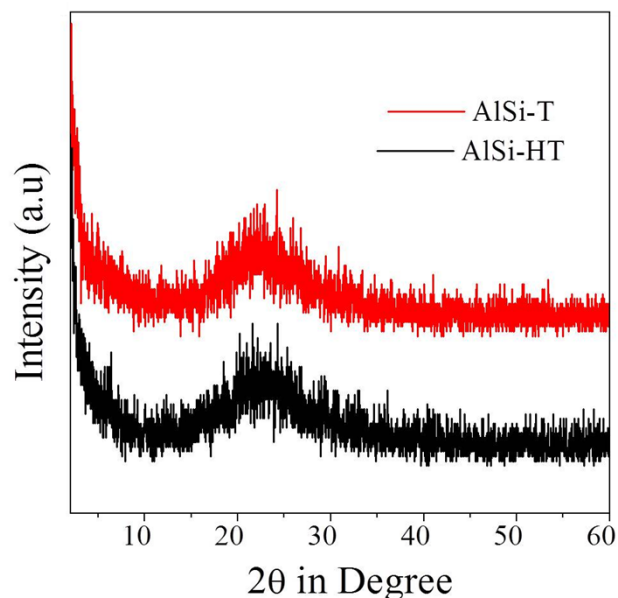**Supporting figure 3:** Wide angle XRD spectra of the AlSi-T and AlSi-HT



**Table S1:** Performance comparison of AlSi-T and AlSi-HT catalysts (with those reported in literature) towards N-alkylation of aniline for selective production of imine

| Support | Precious metal load | Time (h) | Aniline : Benzyl Alcohol (mole:mole) | Aniline Conversion (%) | N-benzylidenaniline (imine) Selectivity (%) | Ref. |
|---|---|---|---|---|---|---|
| [a]TiO$_2$ | (0.5%)Pt | 12 | 1:1000 | 54 | 88 | S1 |
| [a]TiO$_2$ | (1.0%) Pt | 12 | 1:1000 | 42 | 86 | S1 |
| [a]Pt@TiO$_2$ | (0.3%)Pt | 12 | 1:1000 | 99 | 98 | S1 |
| [b]Al$_2$O$_3$-Ga$_2$O$_3$ | Ag | 26 | 1:2 | 89 | 3 | S2 |
| AlSi-T | ……….. | 4 | 1:5 | 40 | 100 | Present study |
| AlSi-HT | ……….. | 4 | 1:5 | 42 | 100 | Present study |
| [a]N2, 1 atm;  temperature, 25°C; photoirradiation time, 12 h; Catalyst, 0.005g. [b] NaH,  toluene, 26 h, temperature,110∘C;  Ar; 3% mole catalyst. [c]Temperature, 100˚C; Reaction time, 4h; Pressure ,atmospheric; Catalyst, 0.25g. | | | | | | |

S1.  Y. Shiraishi, M. Ikeda, D. Tsukamoto, S. Tanaka and T. Hirai, *Chem. Commun.,* 2011, **47**, 4811

S2.  I. Geukens, F.  Vermoortele, M.  Meledina, S Turner, G.  V.  Tendeloo, D.  E. D. Vos, Applied Catalysis A: General , 2014, 469, 373.

**Table S2:** Reusability studies of AlSi-T catalyst in N-Alkylation of aniline

| Reaction cycles | Reaction time (h) | Conversion (%) | N-benzylidenaniline (imine) Selectivity (%) |
|---|---|---|---|
| 1 | 4 | 40 | 100 |
| 2 | 4 | 38 | 100 |
| 3 | 4 | 35 | 100 |

**Table S3:** Reusability studies of AlSi-HT catalyst in N-Alkylation of aniline

| Reaction cycles | Reaction time (h) | Conversion (%) | N-benzylidenaniline (imine) Selectivity (%) |
|---|---|---|---|
| 1 | 4 | 45 | 100 |
| 2 | 4 | 44 | 100 |
| 3 | 4 | 42 | 100 |