Supplementary informations:

Catalytic coupling reaction:

Haloarene (1 mmol), aryl boronic acid (1.5 mmol), potassium carbonate (3 mmol), Pd(0) catalyst (0.03 g), 10 ml of 1,4-dioxane-water(5:1) as solvent were charged in a round bottom flask. Reaction was carried out at 80 ^{o}C for 10 h. After completion of the reaction, the reaction mixture was filtered and extracted with diethyl ether. The final product was analyzed by Gas chromatography.

XRD:

The XRD pattern of LDH-NO_3 (Figure S-1) shows a basal reflection at 8.62 Å (2θ =10.2), which is similar to that reported in the literature [1]. In case of SDS intercalated LDH the position of (003) peak shifted to lower 2θ =3.3 angle corresponds to the and give access to the interlayer distance d_{003} ~ 26.9 Å. Organic moiety (TPED) intercalated LDH the position of (003) peak shifted to low 2θ =3.6 angle corresponding to the interlayer distance d_{003} ~ 24.9 Å. LDH-Pd^{o} is showing four distinct reflection patterns for palladium along with the peaks for LDH. The most intense peak around 40^{o} (indexed as the {111} plane) of Pd showing the lattice spacing 0.22 nm. Another three well distinguish peaks at {200}, {220} and {311} appeared at 46^{o}, 67^{o} and 82^{o} respectively with low intensities. The retention of characteristic peak (110) around $2\theta = 60^{o}$ indicated that the structure of the layer has been retained [2] even after multi step synthesis.

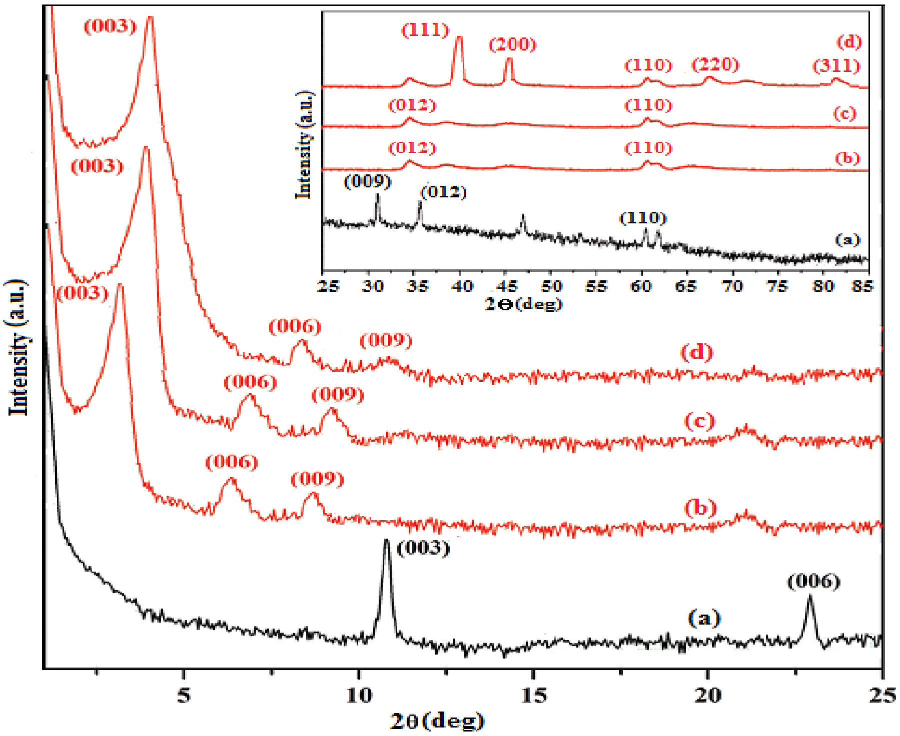


Figure S-1. XRD patterns of (a) LDH-NO_3 (b) LDH/SDS (c) LDH/TPED (d) LDH-Pd^o

FTIR Studies

FTIR spectra of LDH, LDH/SDS and LDH/TPED are shown in Figure S-2. In case of LDH bands around 445 and 675 cm^{-1} are due to Al-O and Zn-O lattice vibrations. Sharp band around 1384 cm^{-1} is assigned to stretching vibration of interlayer NO_3^-. Interlayer water also give rise to medium intensity absorption close to 1620 cm^{-1} δ(H2O). Broad bands in between 3300 and 3600 cm^{-1} (as a result of H-bonding) is due to the ν(OH) modes of the hydroxyl groups (both from brucite like layers and from water molecules). The characteristic bands are detected for SO_4^- group in LDH/SDS. Bands around 1047 and 1202 cm^{-1} are due to asymmetric stretching of C-O and SO_4^- bonds of surfactant. Typical C-H stretching modes for antisymmetric stretching of CH_3 and CH_2 at 2962 and 2927 cm^{-1} and symmetric stretching of CH_3 and CH_2 at 2873 and 2852 cm^{-1} respectively are observed. For TPED intercalated LDH, the intensities of the characteristic bands due to SDS molecules are generally weakened as compared with that of LDH/SDS. This is a clear indication of replacement of SDS molecules by TPED moiety and stabilized by Br^- anions. In the spectra of LDH/ TPED, the bands for amine group of TPED molecule overlap with O-H stretching vibration of LDH to produce a merged spectrum in the range of 3300 to 3500 cm^{-1}. All the above results provide an indication of the presence of TPED in the LDH host.

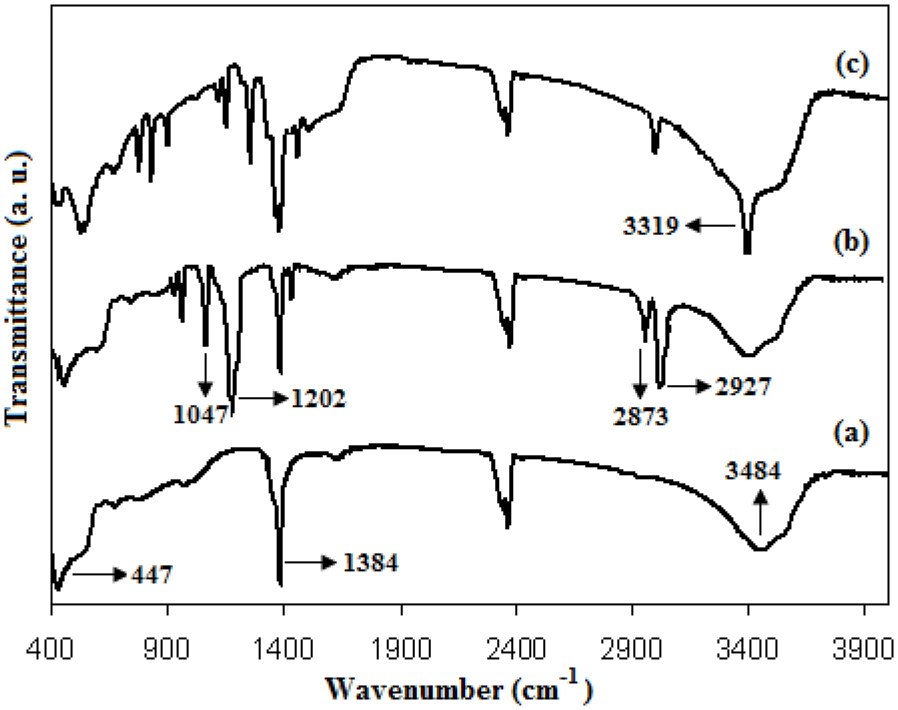
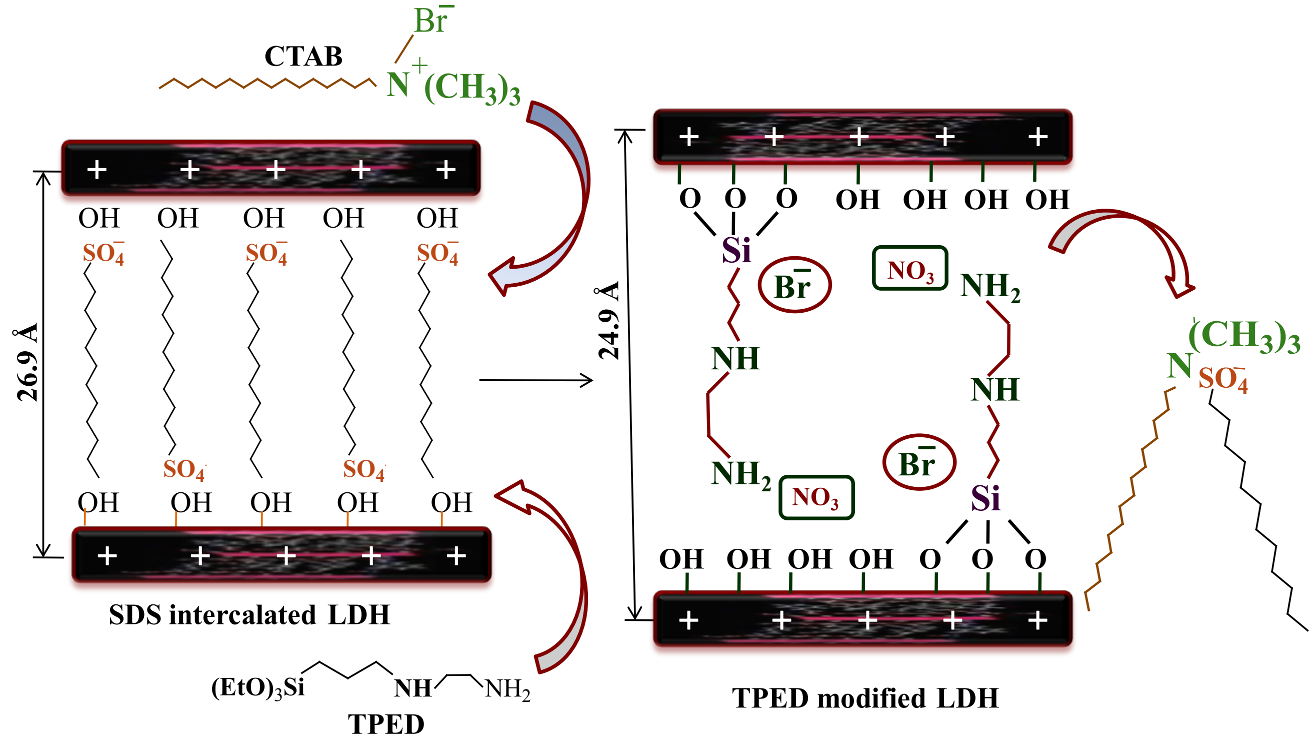


Figure S-2. FT-IR spectra of (a) LDH (b) LDH/SDS (c) LDH/TPED.

Mechanism:

The key procedure is the formation of a salt between the pre intercalated anionic surfactant (Sodium dodecyl sulfate, SDS) and cationic surfactant (cetyl trimethyl ammonium bromide, CTAB) added afterwards. The salt so formed is hydrophobic in nature, insoluble in aqueous medium but soluble in organic solvents (like methylene dichloride). Once the hydrophobic salts (SDS-CTAB) are formed, they are simultaneously replaced by organic moiety N-[3-(Trimethoxysilyl)-propyl] ethylenediamine (TPED) through the migration of these salts to the organic phase and subsequently a covalent attachment of the TPED to the interlayer surface of LDH is formed via M-O-Si bond (M = Zn and Al). The bromide anion seems to be remaining in the interlayer to neutralize the LDH host. The whole mechanistic pathway is illustrated in the above Scheme *S-1*.



Scheme *S-1*: Mechanism responsible for the formation of TPED modified LDH

Recycled Test:

In an effort to confirm the heterogeneity nature of the catalyst, separate experiments were conducted under identical conditions. Initially chlorobenzene and boronic acid were taken as ingredients. The results indicated that catalytic activity remains almost same after four cycles (Table *S-1*). All the other substrates showed similar types of results.

Table S-1. Recyclability test upto four cycles

No. of cycles	Yield (%)
Fresh	88
1	88
2	87
3	87
4	86

Reaction conditions:Chlorobenzene (1mmol), boronic acid (1.5 mmol), potassium carbonate (3 mmol), Pd(0) catalyst (0.03 g), 1,4-dioxane-water(5:1)=10 ml, temp=80 °C, time=10 h.

References

[1] C. Li, L. Wang, D. G. Evans, X. Duan, Ind. Eng. Chem. Res., 48 (2009) 2162-2171.

[2]G. R. Williams, D. O'Hare, J. Mater. Chem. 16 (2006) 3065.