# Supporting Information

# Highly mesoporous organic aerogels derived from soy and tannin

Gisele Amaral-Labat,[a] Liudmila Grishechko,[a,b] Andrzej Szczurek,[a] Vanessa Fierro,[a] Antonio Pizzi,[c] Boris Kuznetsov[b] and Alain Celzard*[a]

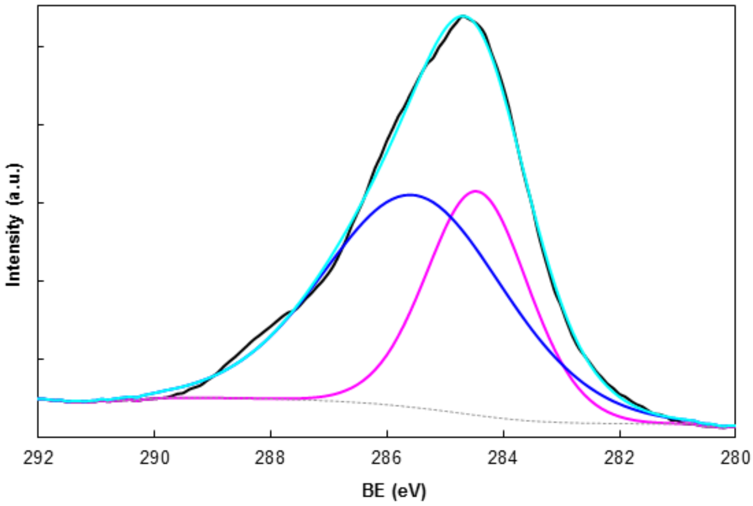
[a] *Institut Jean Lamour - UMR Université de Lorraine – CNRS 7198, ENSTIB, 27 rue Philippe Séguin, BP 1041, 88051 Épinal cedex 9, France. Fax: (+33) 3 2929 6138; Tel: (+33) 3 2929 6114; E-mail: Alain.Celzard@enstib.uhp-nancy.fr*

[b] *Institute of Chemistry and Chemical Technology, SB RAS, 42 K. Marx Street, Krasnoyarsk 660049, Russia*

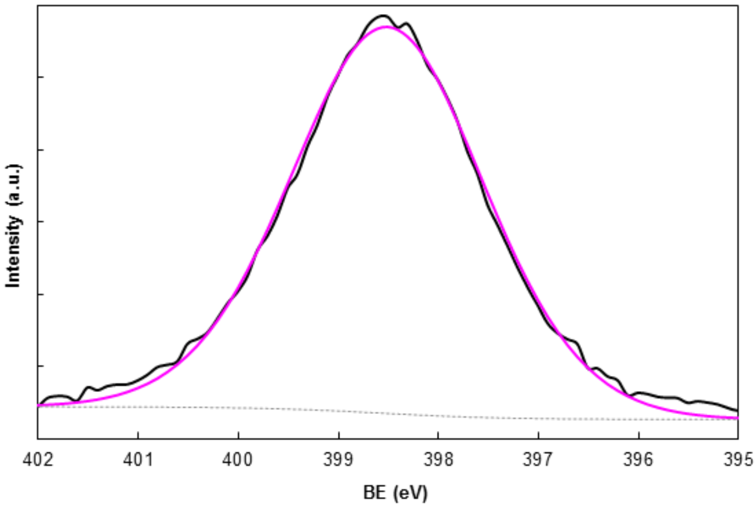
[c] *LERMAB - ENSTIB, 27 rue Philippe Séguin, BP 1041, 88051 Épinal cedex 9, France*

*Fax: (+33) 3 2929 6138; E-mail: Alain.Celzard@enstib.uhp-nancy.fr*

*To whom correspondence should be addressed.



(a)



(b)

**Fig. S1** XPS spectra of ASTF6 organic aerogel sample, and corresponding deconvolution of the fit (colour online). (a) C1s binding energy (BE) region; (b) N1s binding energy (BE) region.

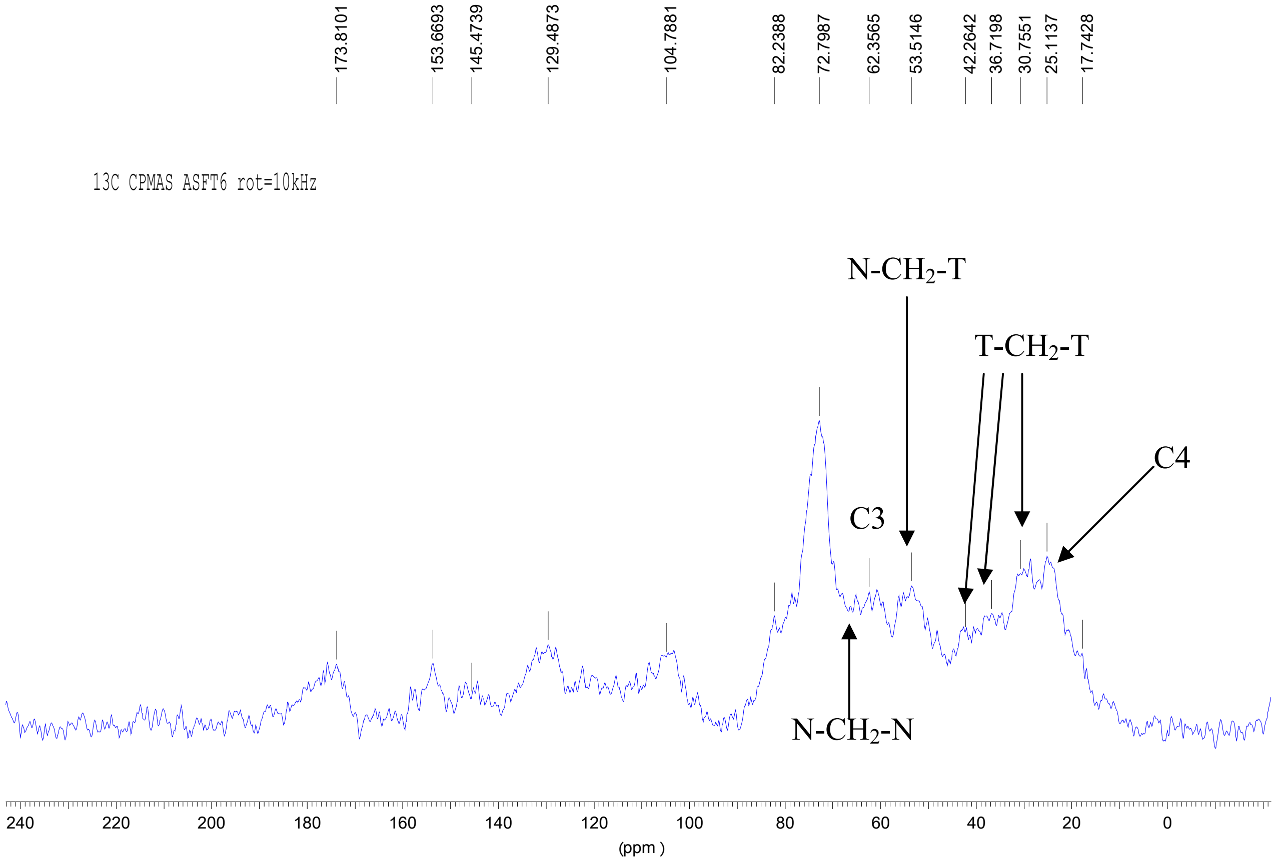


**Fig. S2** Solid-state CP-MAS $^{13}$C-NMR spectrum of ASTF6 organic aerogel sample.

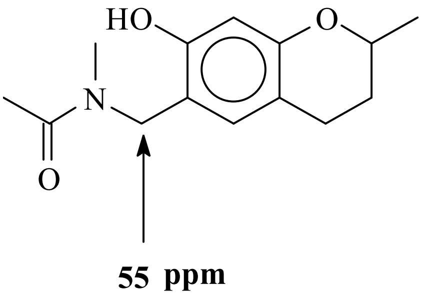
The above NMR spectrum may be interpreted as follows. Arrows added to the spectrum indicate which is which.

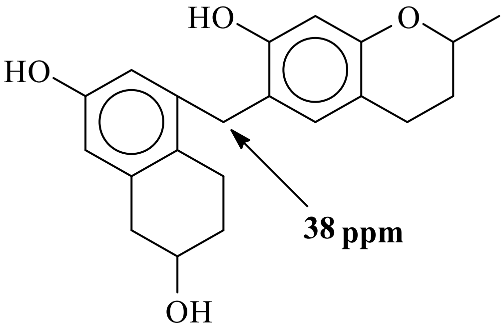
First of all, the peaks of the tannin unaltered are at 155-156 ppm (C5, C7, C9, see atom numbering in Fig. 1), at 145-146 ppm (C3', C4'), at 130 ppm (C1'), at 105 ppm (the interflavonoid C4-C6 bond), at 81-82 ppm (C2) and at 24-25 ppm (C4). These are absolutely normal. The soya carbohydrate peak is the biggest one at 70-75 ppm.

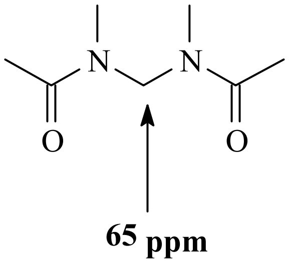
Now, the following indicates that the tannin and the protein of soy flour have well reacted:

1. The free reactive sites peaks of the flavonoid (C6 and C8) at 95-98 ppm are practically non-existent, they have disappeared, indication that something, either the formaldehyde or the methylol group on the preformylated soy have completely or almost completely reacted with it.

2. Confirmation of this comes from the -CH$_2$- signals region. Indeed, in this region there is the C4 of the tannin, but also the methylene bridges between tannin and tannin (thus, T-CH$_2$-T), between amido-group of soy protein and amido-group of soy protein (thus Soy-CH$_2$-Soy), and finally the methylene bridges linking the soy amido-groups to the tannin, thus T-CH$_2$-Soy. Those links of interest have been pictured below.







It is evident from the spectra, given that the $N\text{-}CH_2\text{-}N$ peak is rather small, that the greatest number of bridges occurs between tannin and tannin and especially between premethylolated soy with tannin. This is logical for two reasons: (1) the soy has been prereacted with the formaldehyde so we are forcing the more reactive species, the tannin, to react with the methylol groups created on the soy by formylation; (2) methylene bridges between amido-groups are easily cleaved to recreate the following equilibrium:

$$N\text{-}CH_2\text{-}N \rightleftarrows N\text{-}CH_2OH + \text{-}NH\text{-}$$

As they dissociate to form either the methylol group or even free formaldehyde, they will react with the most reactive species around, recreating $Soy\text{-}CH_2\text{-}Tannin$ bridges as well as $Tannin\text{-}CH_2\text{-}Tannin$ bridges if the above equilibrium had regressed even to free HCHO.