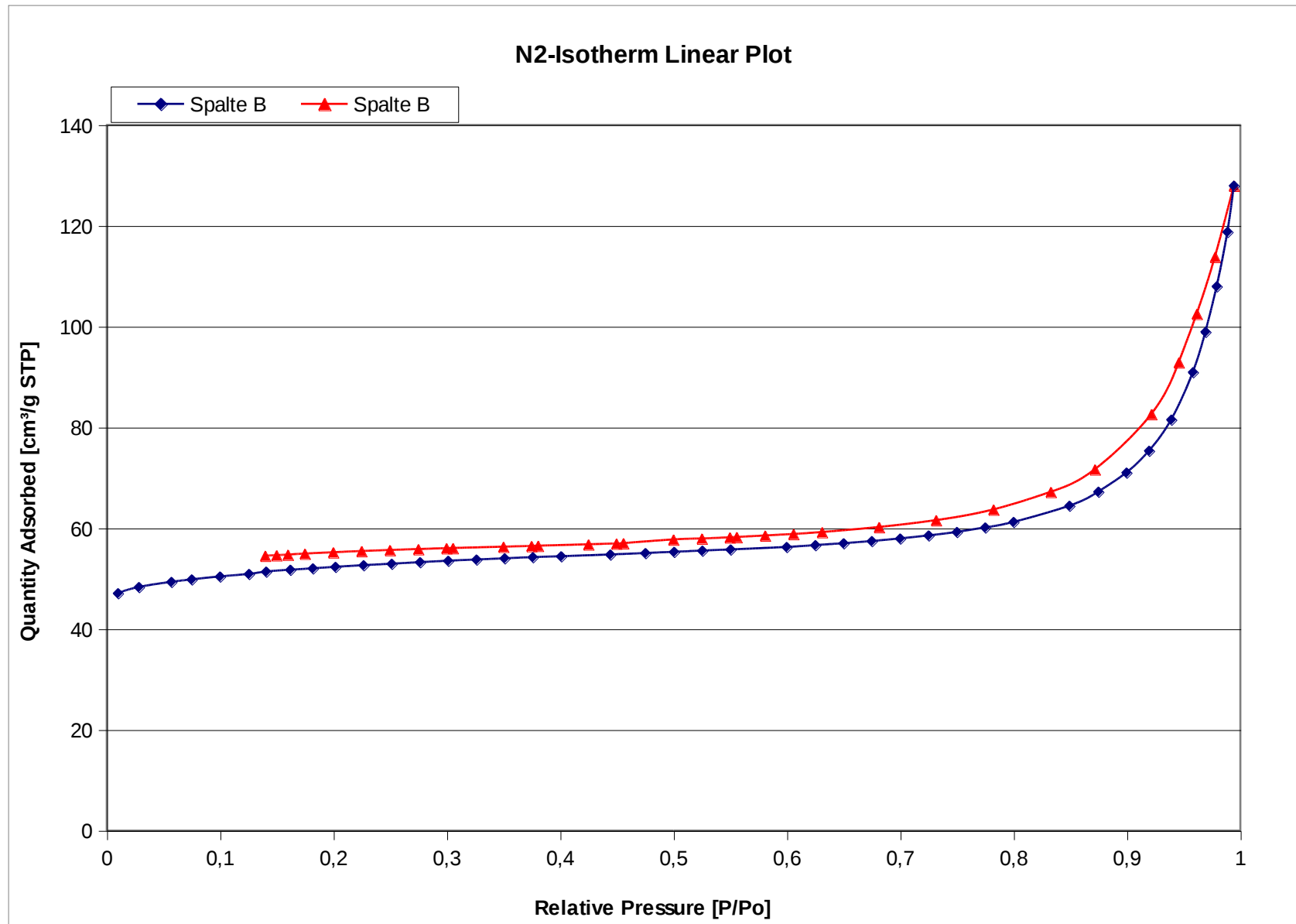
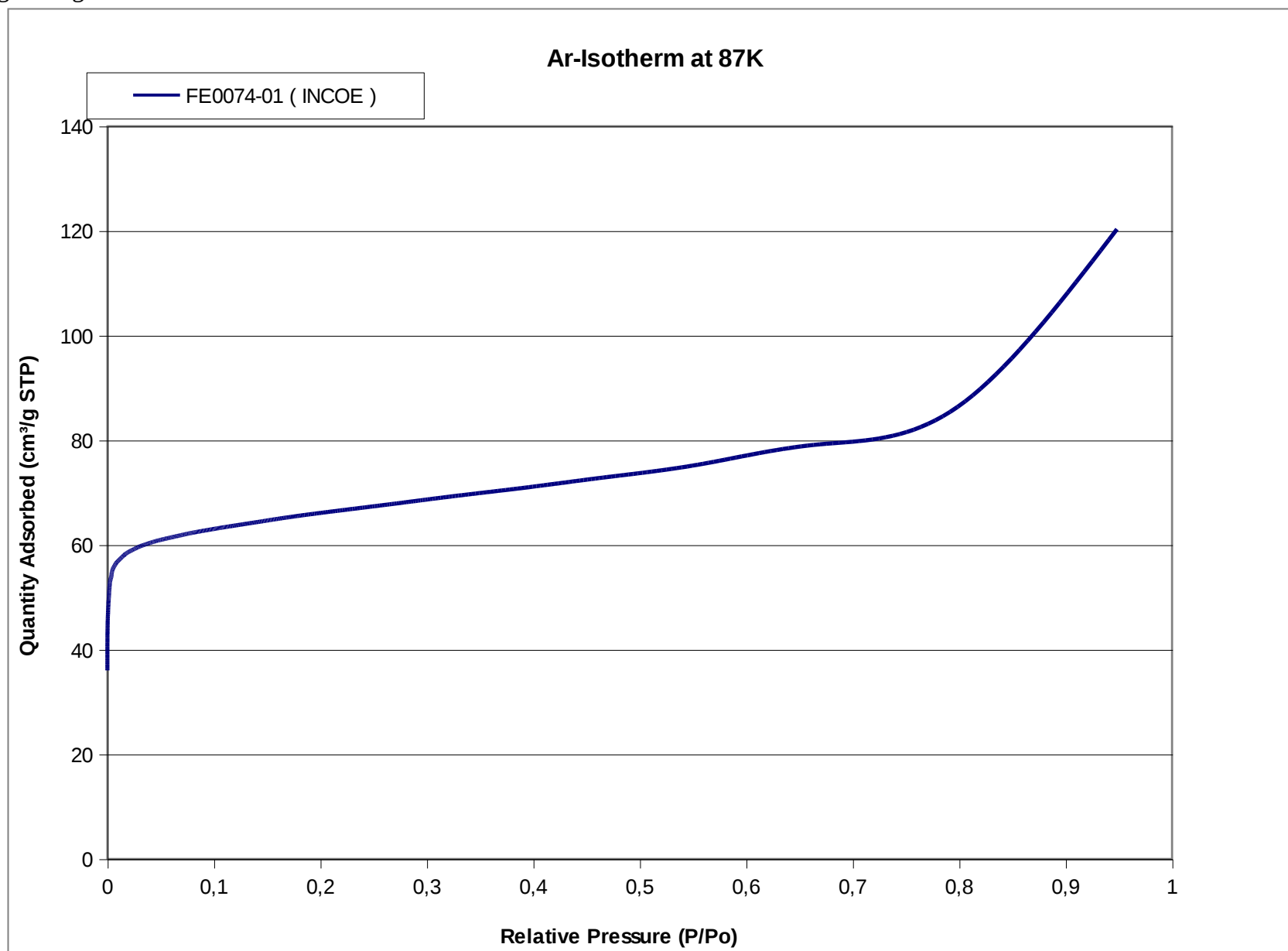


Supplementary Information

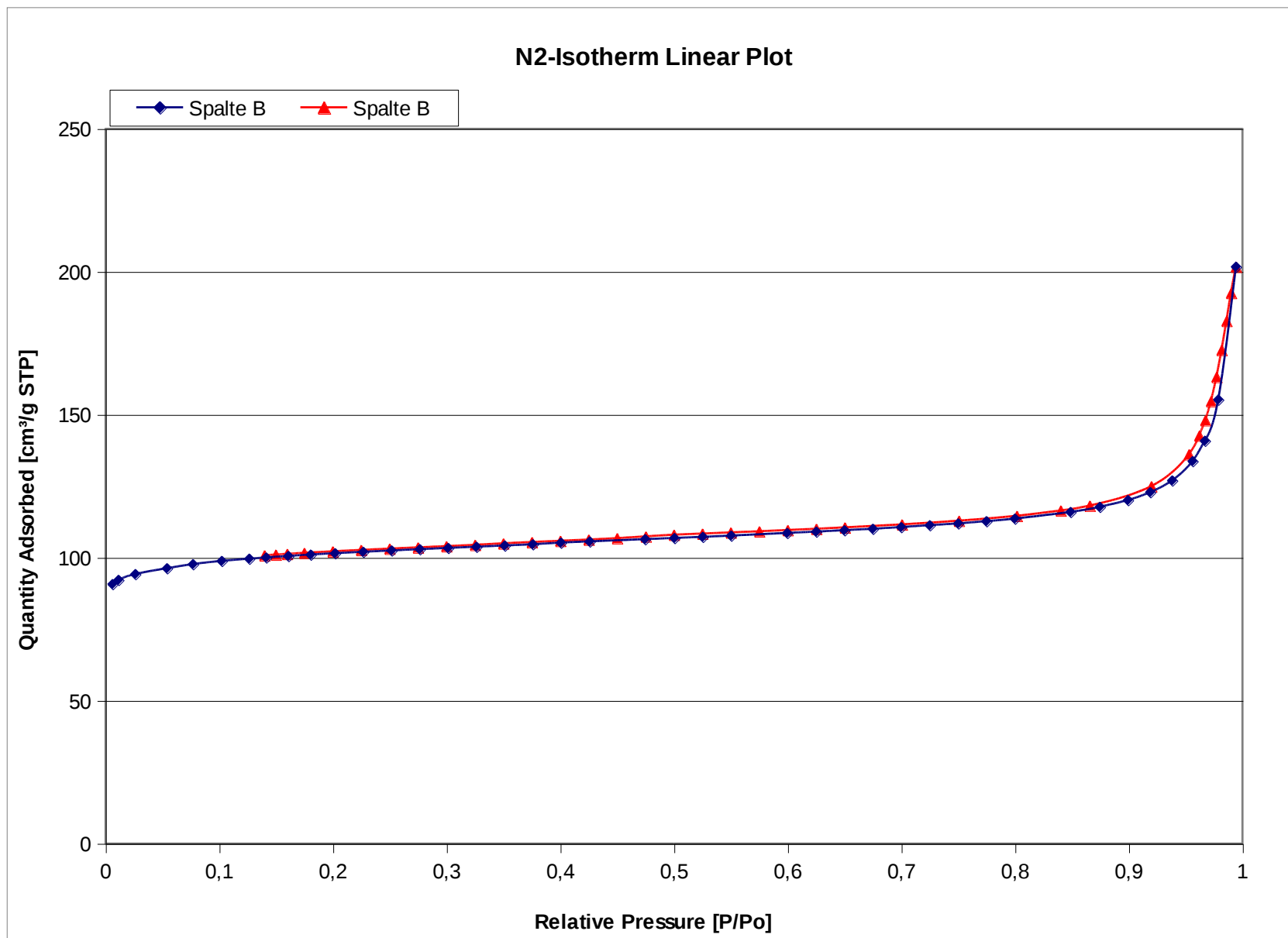
SFig. 1: Nitrogen isotherm of COE-3 in the as made form.



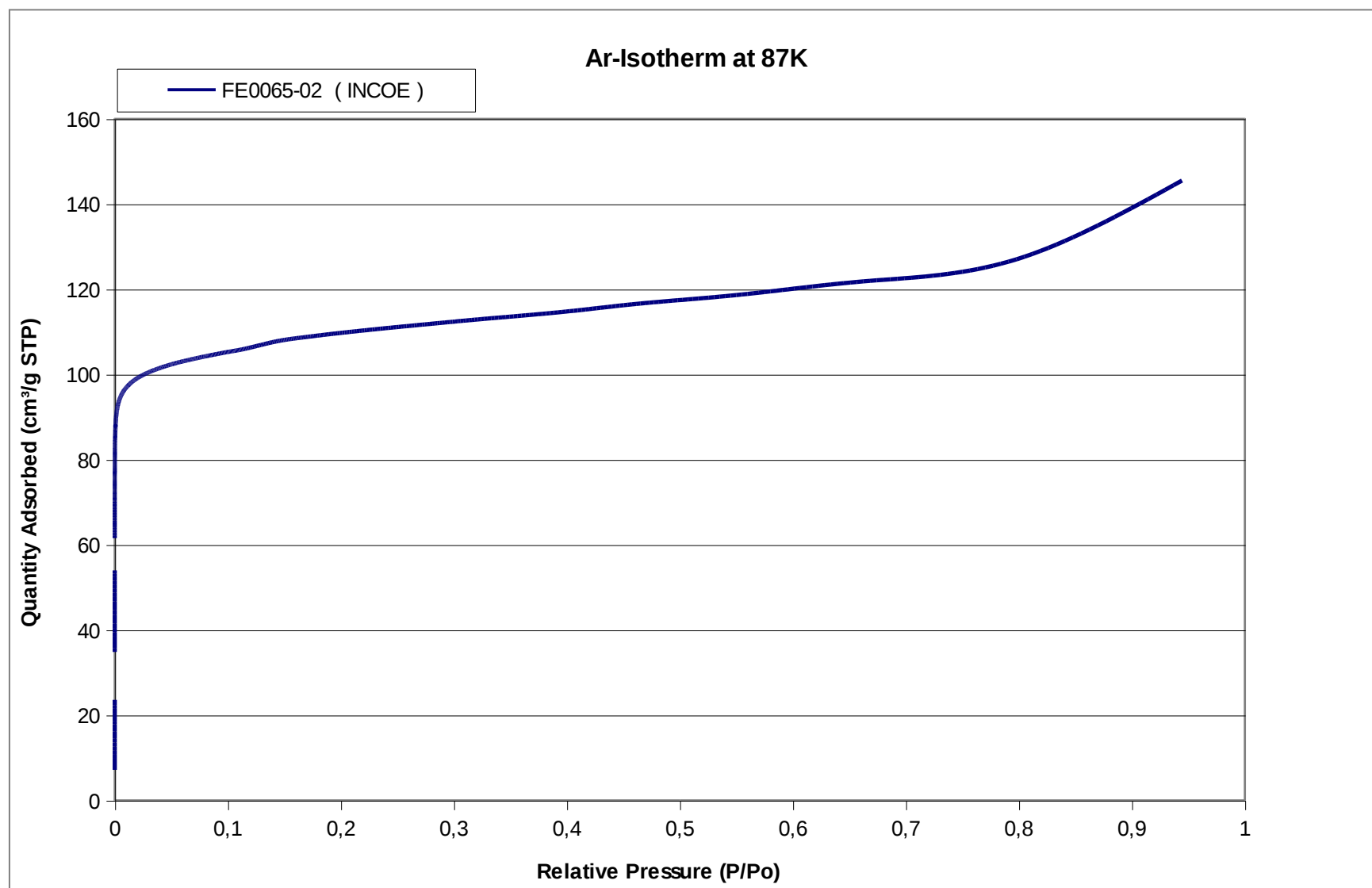
SFig. 2: Argon isotherm of COE-3 in the as made form.



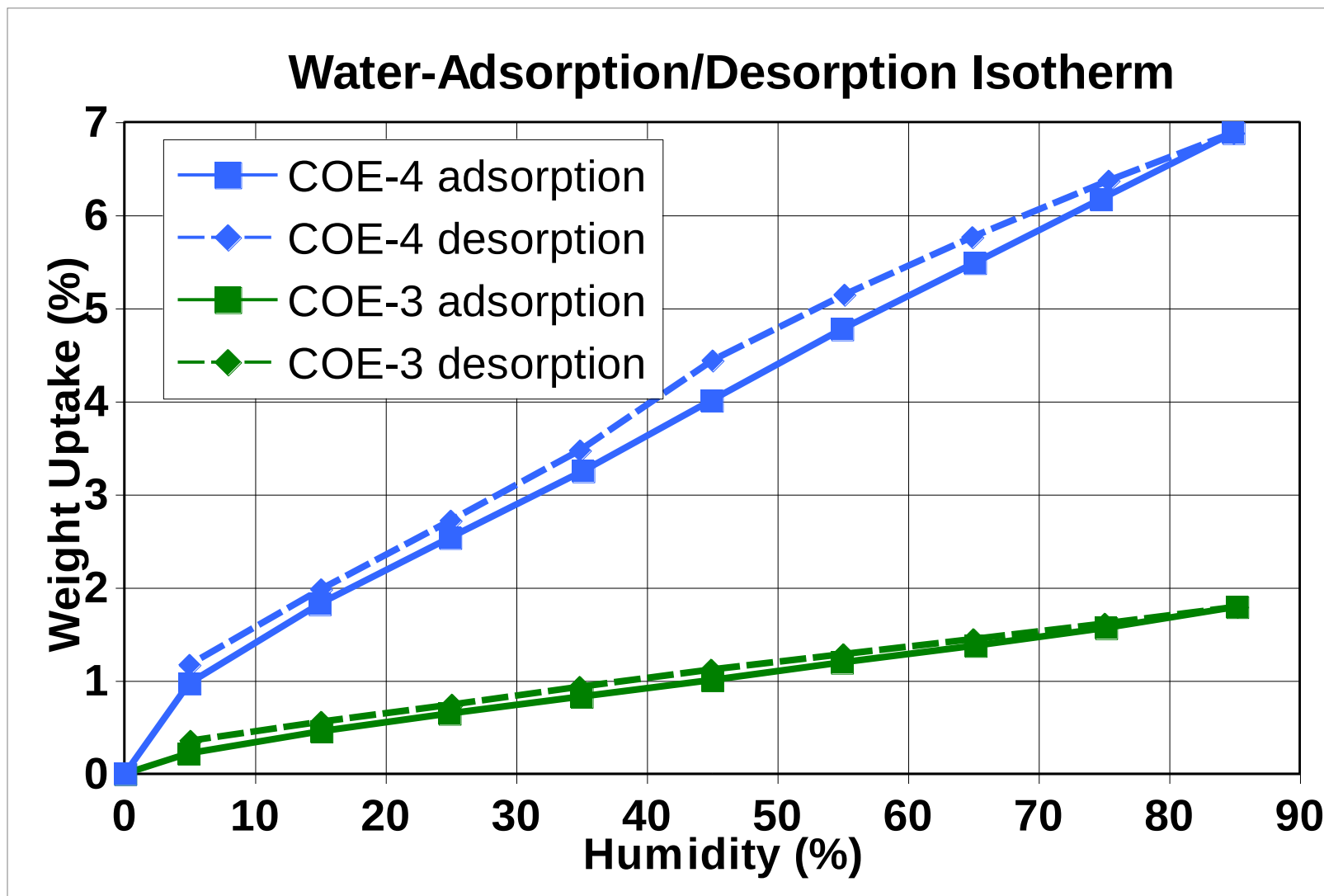
SFig. 3: Nitrogen isotherm of COE-4, the calcined material after interlayer expansion.



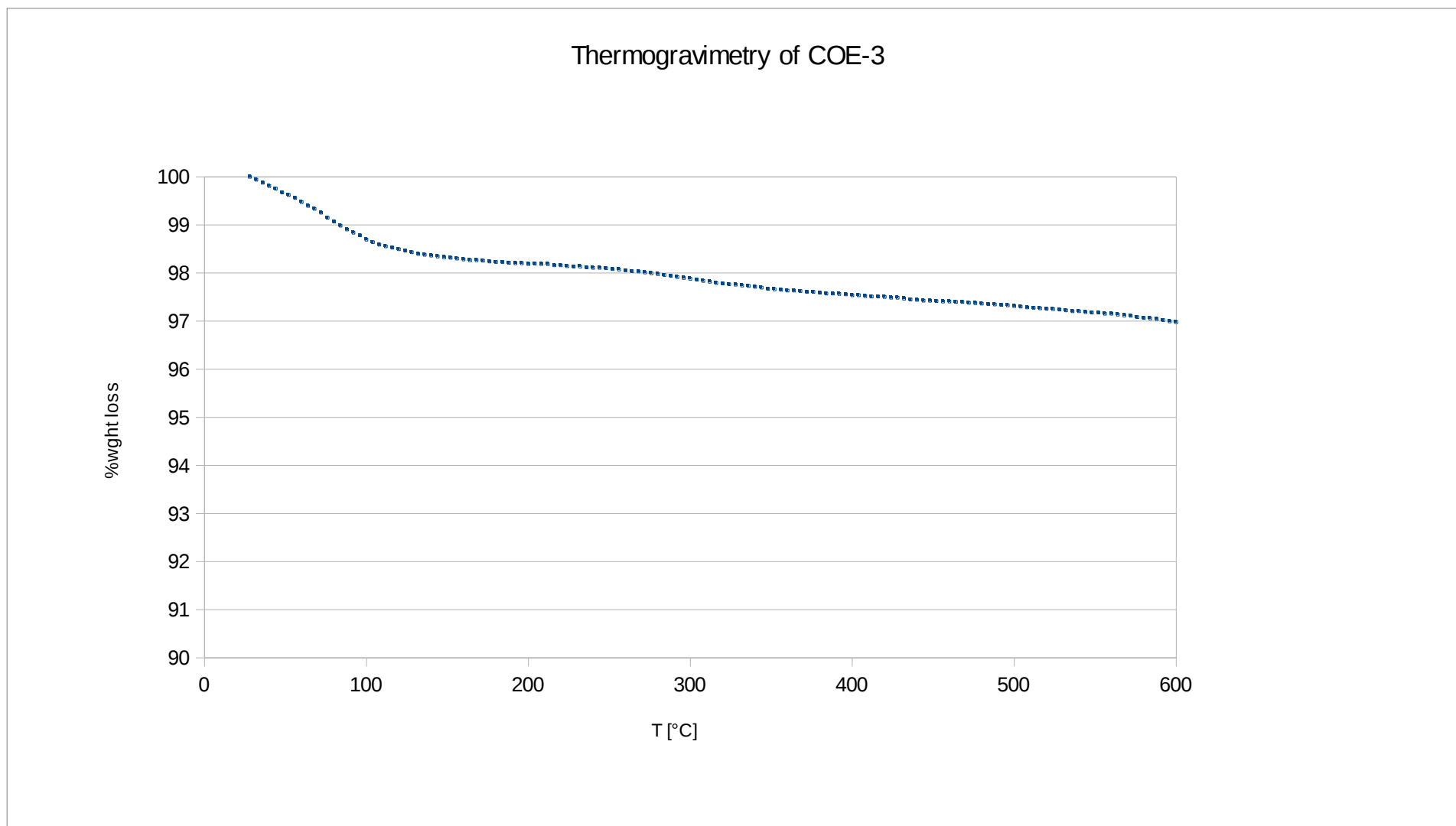
Sfig. 4: Argon isotherm of COE-4, the calcined material after interlayer expansion.



Sfig. 5: Water adsorption-desorption isotherm of COE-3 and COE-4 material showing the distinct hydrophobic and hydrophilic properties respectively.



SFig. 6: Thermogravimetry of COE-3, the interlayer expanded product of the silylation reaction.



SFig. 7: ^1H MAS NMR spectrum of COE-4. Asterisk denotes the background signal of the NMR probehead. The peak at 1.7 ppm is ascribed to the isolated single silanol groups. The peak at 2.3 ppm is assigned to the single silanol groups with hydrogen-bond effect. Additionally, the dominant signal at 3.3 ppm should be attributed to the dual silanols at the interlayers of COE-4. There is no signal in the low field up to 20 ppm, which means no hydrogen bond similar to the layered precursor in COE-4.

