Electronic Supplementary Information

For GC analysis, reaction samples were diluted with methanol (1/10 v/v; Aldrich, 99.9+% purity). 1 µl of diluted sample was injected in a Varian 3900 GC apparatus, equipped with a 30 m HP-5 column and a FID detector. Nitrobenzene (Aldrich, 99+% purity) was used as internal standard to calculate the content in free aniline and the distribution of diamines and triamines.

For ^{1}H-NMR analysis samples were exchanged several times with D_2O in order to remove all the labile protons (-NH_2 and HOH signals). Spectra were registered in a 300 MHz Varian Gemini spectrograph using $CDCl_3$ as solvent (Aldrich, 99.9 atom %D). Chemical shift is expressed in δ (ppm) regarding to the tetramethylsilane used as internal standard. Typically, four different groups of compounds can be quantified by this technique (see Figure 1): primary amines (^{1}H-NMR δ: 3.7, s, 2H, Ph-CH_2-Ph), secondary amines (^{1}H-NMR δ: 4.2, s, 2H, Ph-NH-CH_2-Ph), quinazolines (mainly 3-phenyl-3,4-dihydroquinazoline, ^{1}H-NMR δ: 4.8, s, 2H, Ph-CH_2-NR_2), and N-methylated derivatives (basically N-methyl-DADPM, ^{1}H-NMR δ: 2.7-2.6, s, 3H, Ph-NH-CH_3). Fig. 1 shows spectra corresponding to the analysis of the reaction mixtures obtained with ITQ-2 (a) and Beta-2 (b).

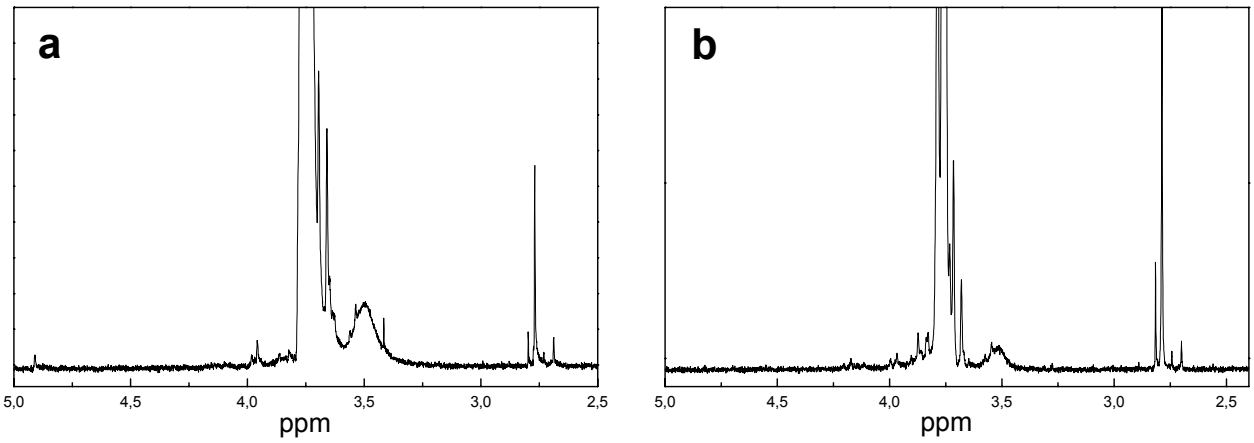

Figure 1 ^{1}H-NMR analysis of reaction samples obtained with ITQ-2 (a) and Beta-2 (b) showing the different groups of products determined. Experimental conditions: A/F=3.0 M (<1% H_2O); T=150 ºC. 20 wt% of catalyst. Reaction time=60 min.. Analytical conditions as in the text.