Supplementary Section

Isotherms
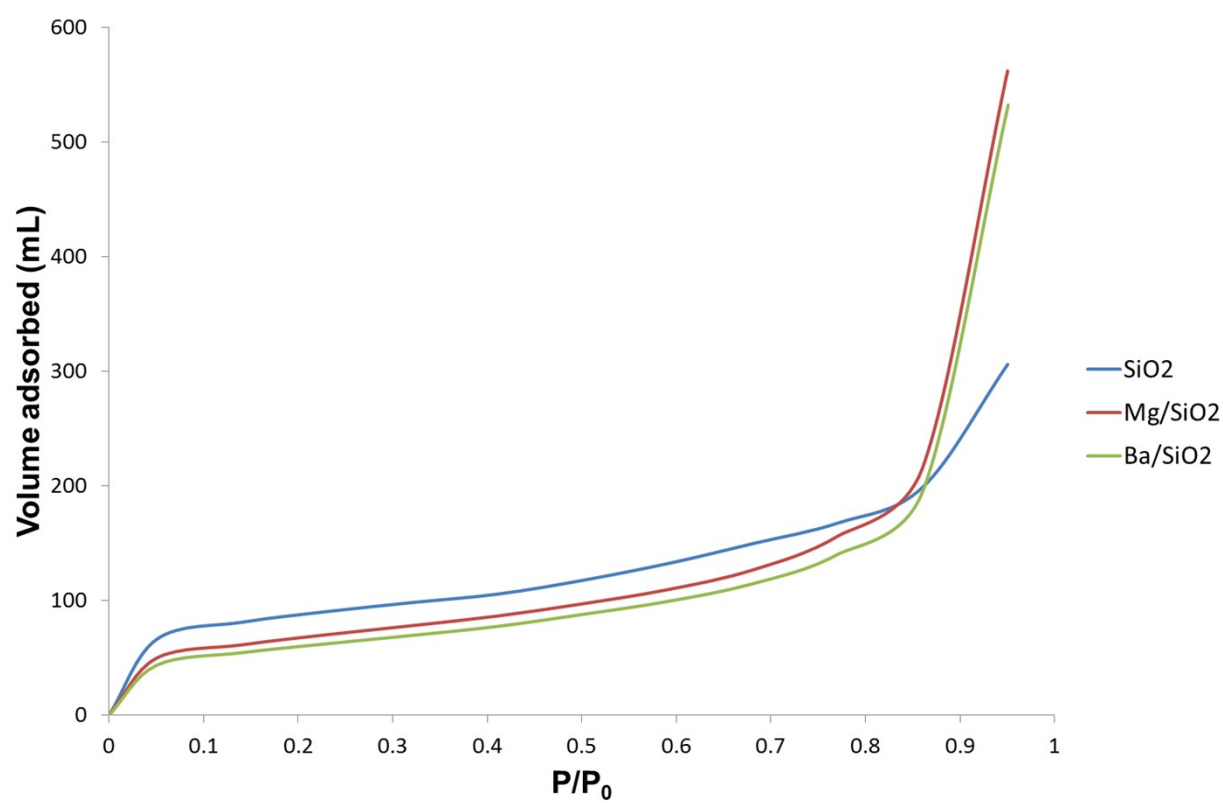

Fig S1: Isotherms of SiO$_2$ catalysts.
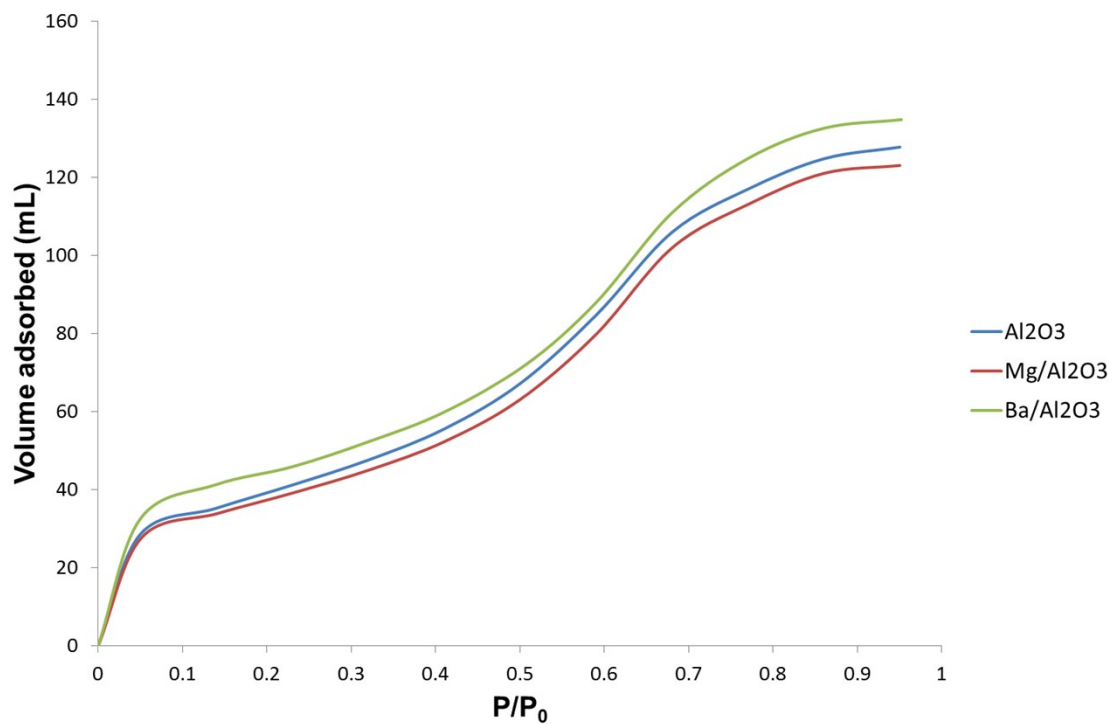
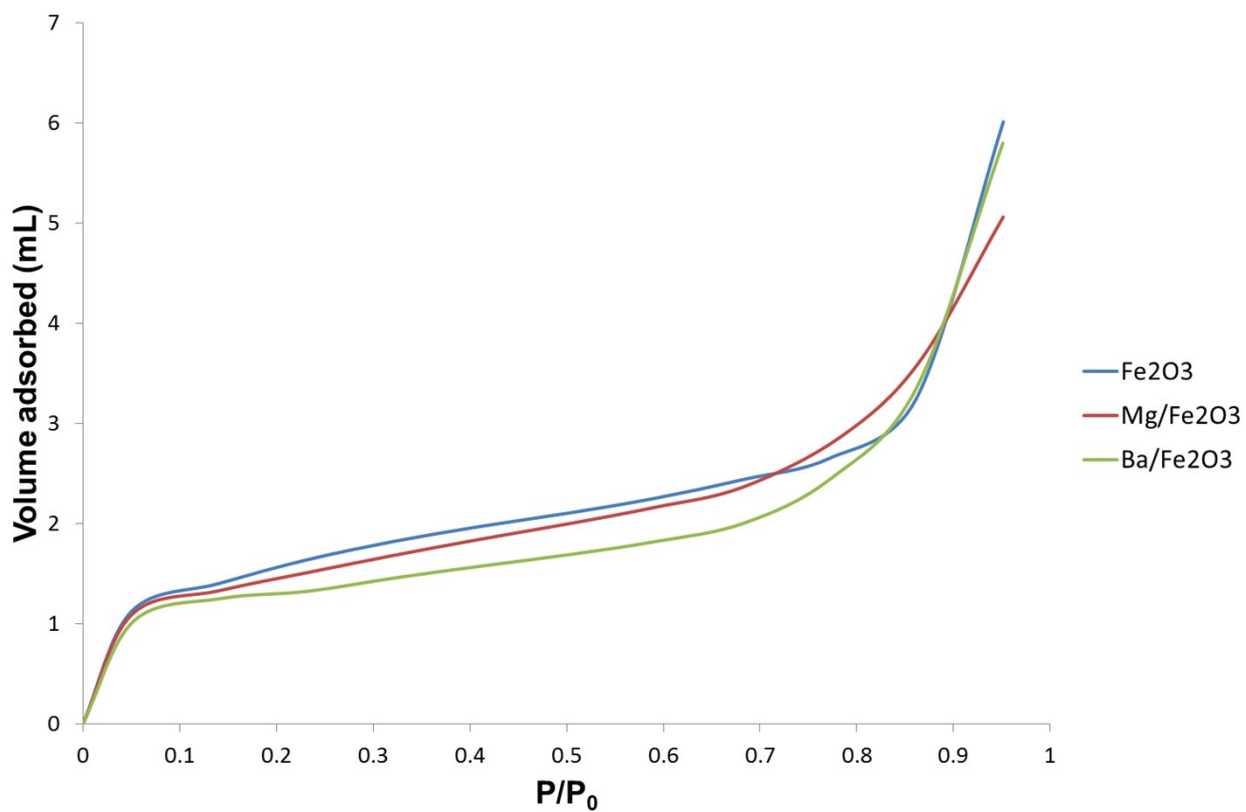**Fig S2: Isotherms of Al$_2$O$_3$ catalysts.**

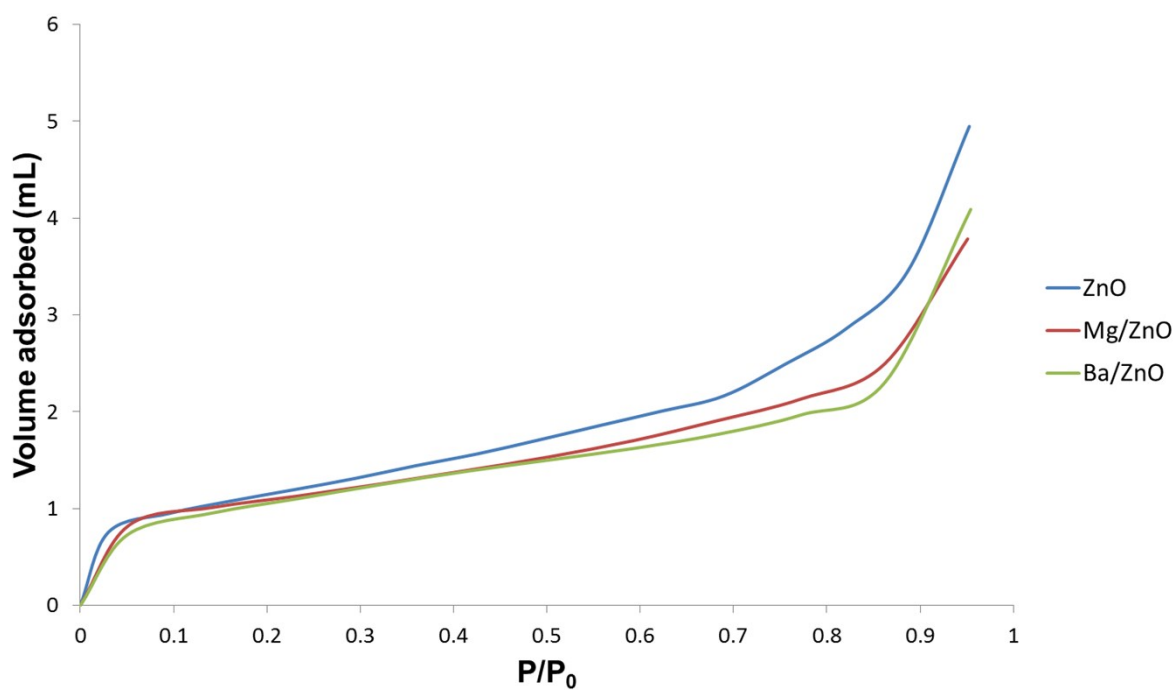**Fig S3: Isotherms of Fe$_2$O$_3$ catalysts.**

Fig S4: Isotherms of ZnO catalysts.
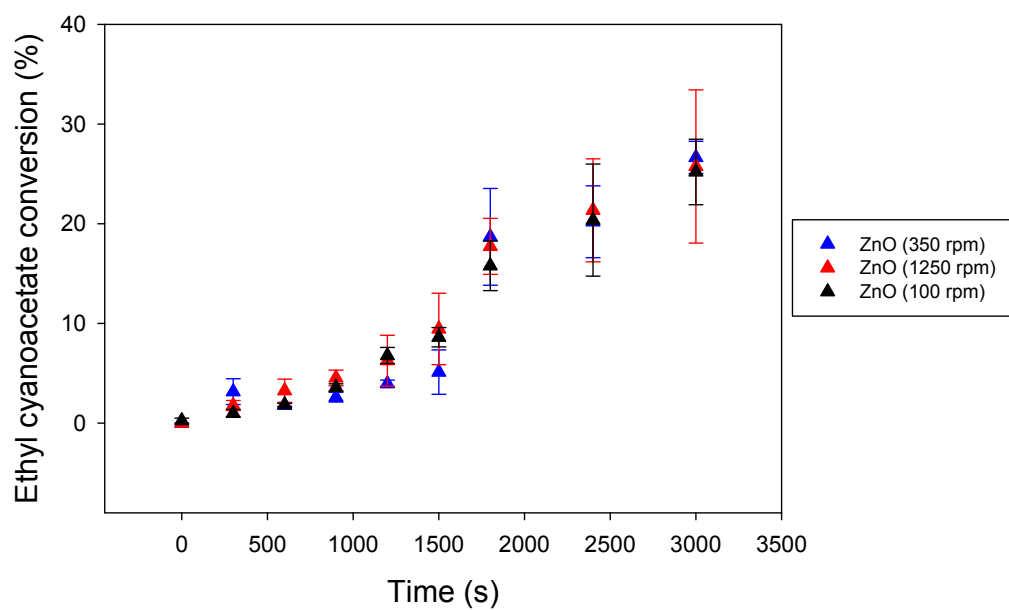
Mass transfer test

Fig S5: A plot of EC conversion against time, comparing the stirrer speed used in the reaction (for ZnO).

Tab. S1: Reaction data showing rate constants for ZnO catalysts used in the Knoevenagel condensation reaction with varying magnetic stirrer speed.

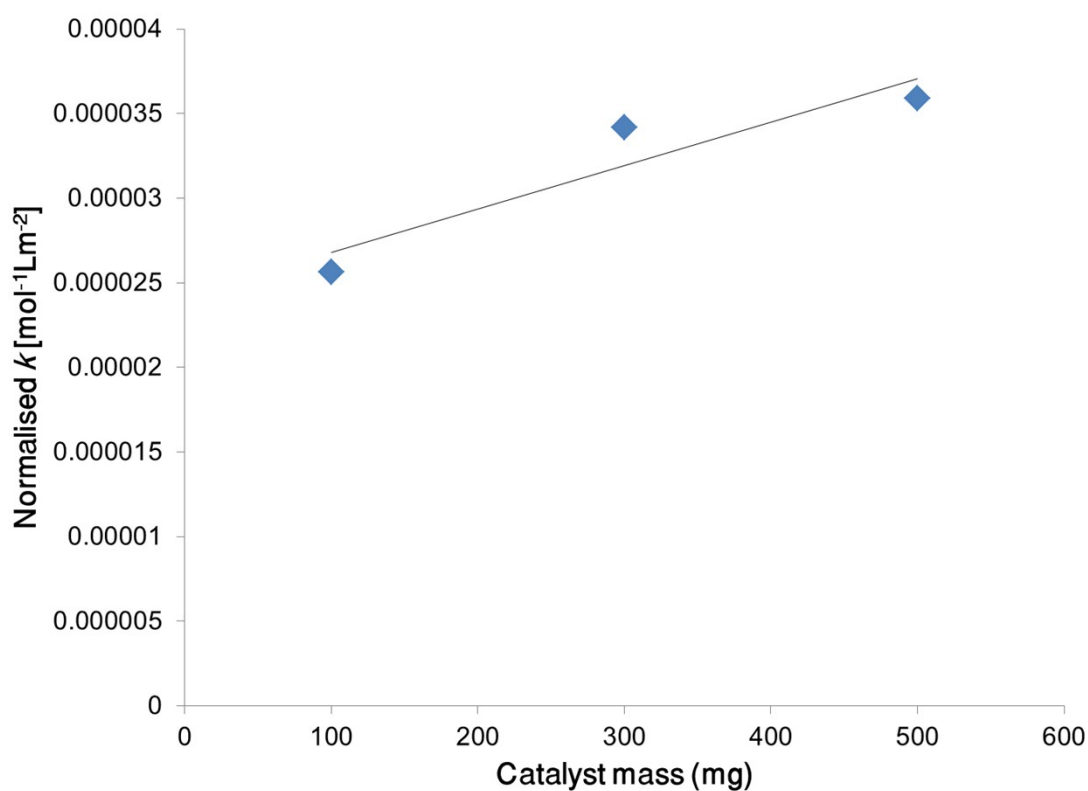| Magnetic stirrer speed (rpm)	Normalised Rate constant k $(mol^{-1}L.s^{-1}m^{-2})$	Normalised Initial Rate $(mol.L^{-1}.s^{-1}m^{-2})$
100	3.4×10^{-5}	2.9×10^{-5}
350	3.4×10^{-5}	2.9×10^{-5}
1250	3.6×10^{-5}	3.0×10^{-5}

Fig S6 Variation of rate constant with catalyst mass

Discussion on transport effect
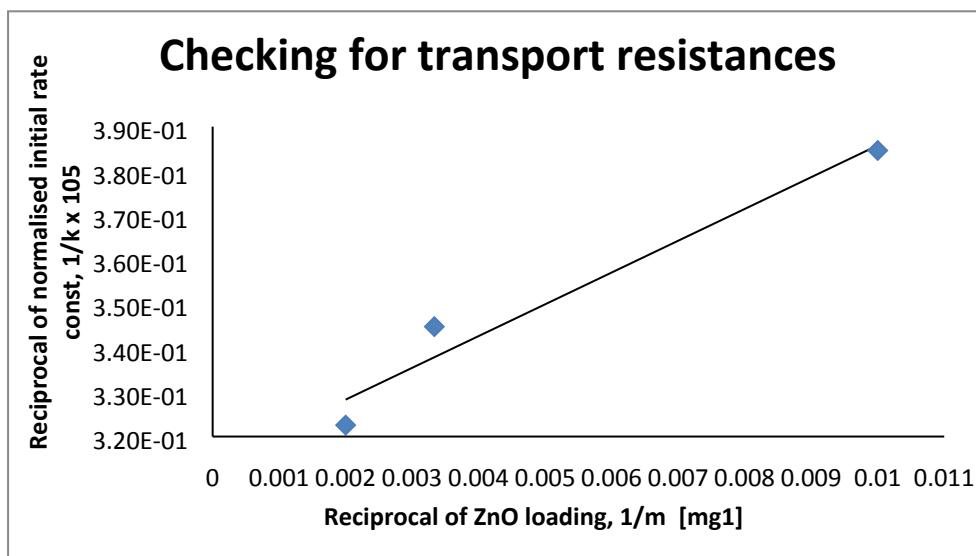

Checking for transport resistances

Fig S7 Variation of reciprocal of normalized initial rate vs. reciprocal of catalyst loading

Tab. S2: Reaction data showing normalized rate constants for ZnO catalysts used in the Knoevenagel condensation reaction, reciprocal of catalyst mass and reciprocal of rate constant

ZnO loading, m	Normalised rate const k	1/m	1/k
100	7.60E-07	0.01	1.32E+01
300	9.90E-07	0.003333333	1.01E+01
500	1.10E-06	0.002	9.09E+00

The data on Tab S2 may also be used to determine the relative magnitude of the transport resistances in the stirred reactor. A plot of the reciprocal of the normalised initial rate constant, 1/k, against the reciprocal of the catalyst loading, 1/m, shown in Fig. S7 supports the earlier assertion that the agitation speed of 350 rpm was sufficient ensure strong liquid phase mixing since the y-intercept of the linear plot, the liquid interfacial resistance (cf. SEQ 1), is very small.

$$\frac{1}{k} = r_{liq} + \frac{1}{m}(r_{ext} + r_{int})$$

(SEQ1)

where, r_{liq} is the interfacial resistance between the benzaldehyde and ethyl cyanoacetate liquid droplets during liquid phase mixing to form a homogeneous reactant liquid phase, r_{ext} is the external surface resistance between the solid and liquid phase while r_{int} is the diffusion resistance inside the catalyst pore. It is readily seen that the ratio of the slope (r_{ext} + r_{int}) to the

intercept, r_{liq} is 22.86 (>>1) suggesting that liquid interfacial resistance was far smaller than the combined external and internal catalyst resistances. Given that the latter resistances are themselves small (Thiele modulus, $\varphi_{exp} = 8.12 \times 10^{-4}$ and hence, effectiveness factor, η would be essentially 1.0 regardless of the reaction rate order), we concluded that there were no transport intrusions in the kinetic data collected.
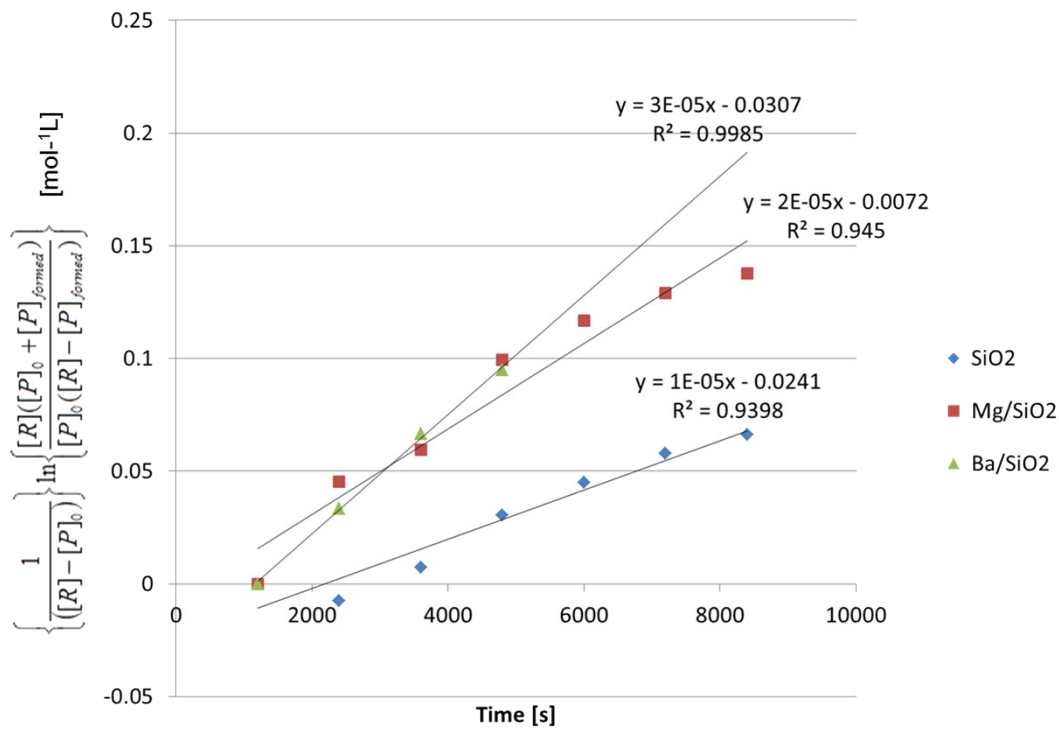
Kinetic modelling

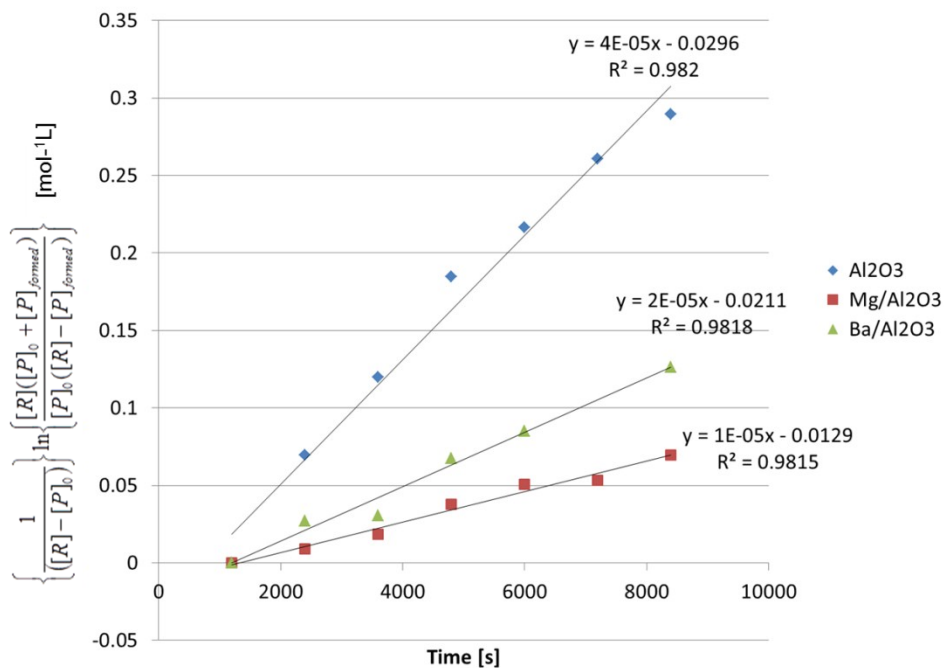**Fig S8: Fitted reactor data for SiO$_2$ based catalysts**

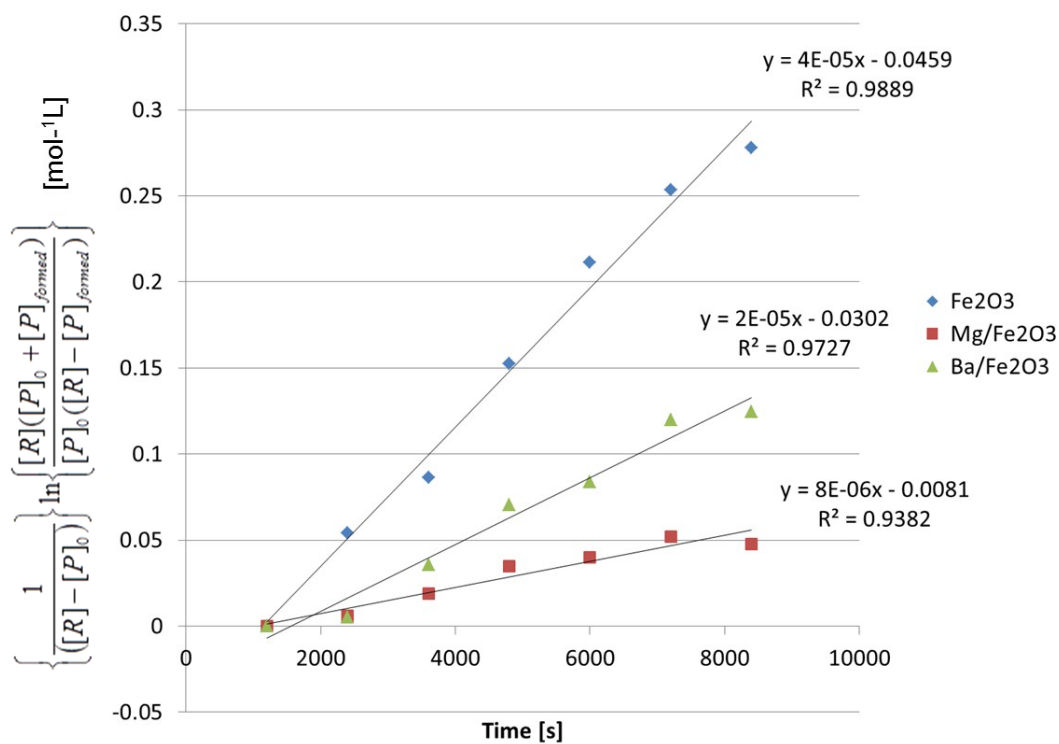**Fig S9: Fitted reactor data for Al$_2$O$_3$ based catalysts**
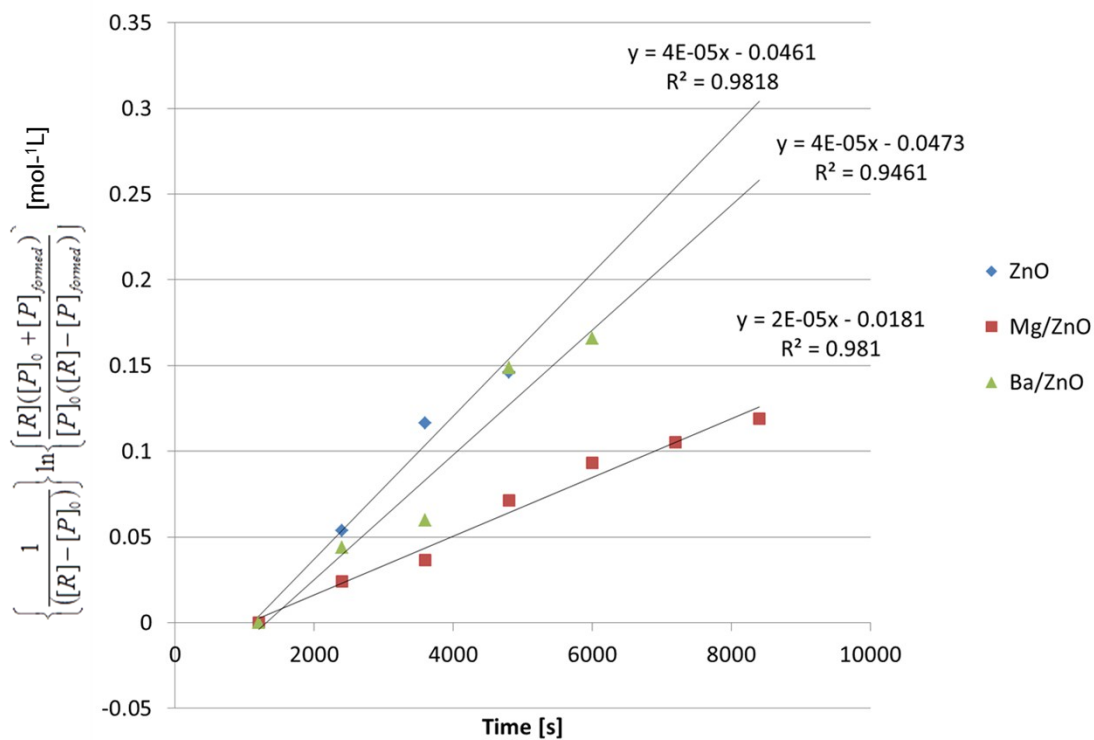

Fig S10: Fitted reactor data for Fe$_2$O$_3$ based catalysts

Fig S11: Fitted reactor data for ZnO based catalysts