Electronic Supplementary Information

Fig. S1. Photograph of the pilot-scale reactor set-up developed for performing operando UV-Vis spectroscopy on catalyst bodies under realistic reaction conditions.

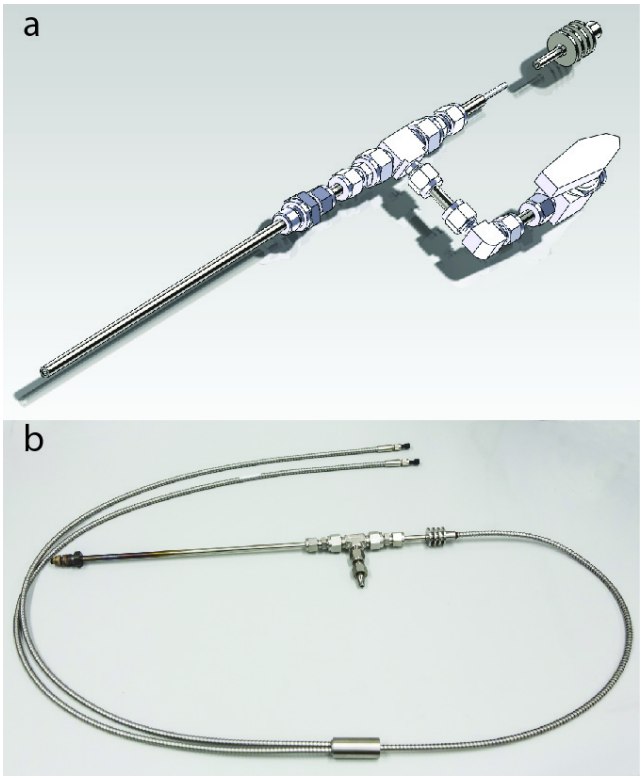

Fig. S2. Scheme of the custom designed probe, which was used to gather UV-Vis spectra in the pilot-scale reactor set-up (a) and related picture (b).

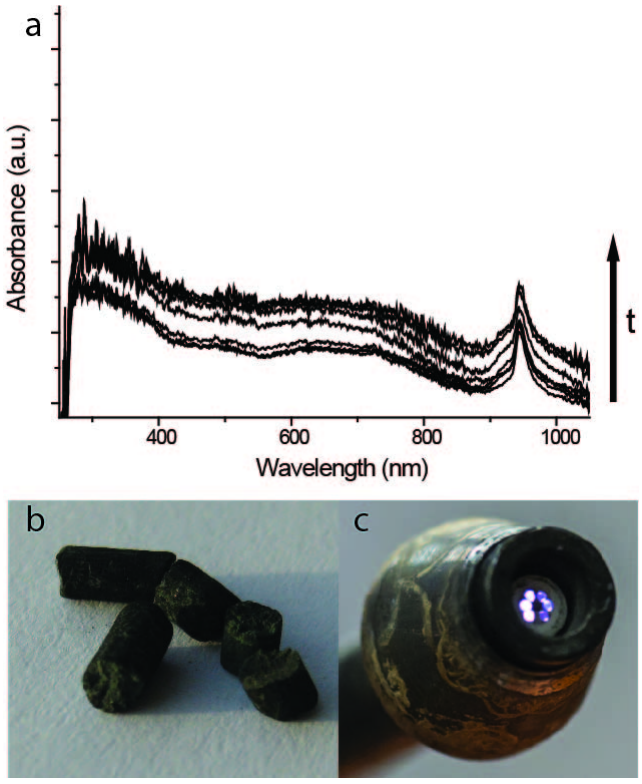

Fig. S3. (a) Operando UV-Vis spectra of a CrO_x/Al_2O_3 catalyst obtained during the propane dehydrogenation reaction with the top probe as described in the article. The experiment is halted after 1 h to study the effect of the nitrogen flow along the probe's length. The spectra are comparable with those shown in Fig. 3a. (b) After the reactor is shut down, the probe is removed and the extrudates measured are taken out of the reactor. These extrudates, shown in the picture, are darker than the pristine Cr/Al_2O_3 bodies, which have a very distinct bright green colour. These particles show a similar level of darkening as other extrudates at that height of the reactor. (c) At the same time the probe's tip remains clear of coke as visualized in the picture.

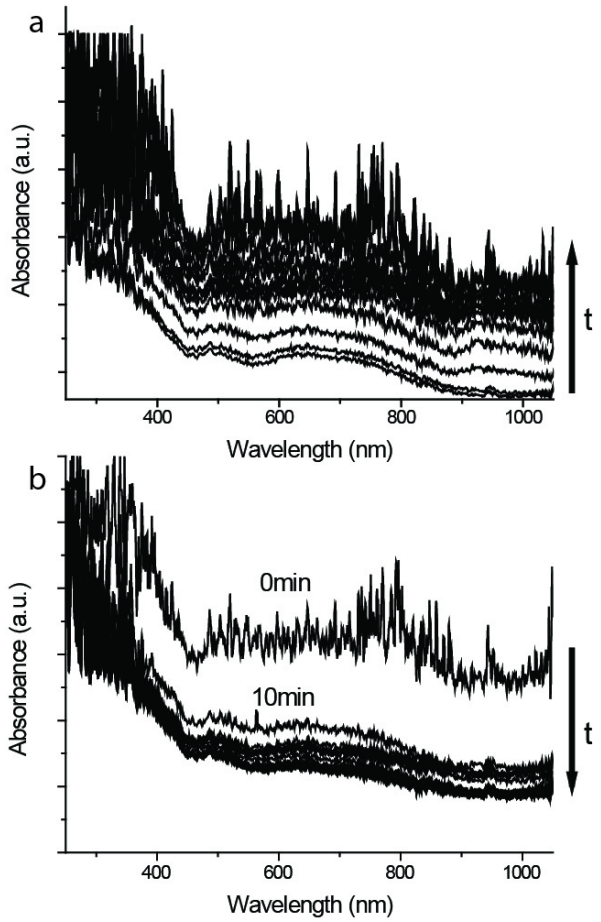

Fig. S4. Operando UV-Vis spectra obtained by the top probe measuring (a) the dehydrogenation process and (b) the oxidation/regeneration process as a function of time on stream. Spectra are collected at a 10 min interval. These spectra, as well as those displayed in Figure 3ab, have been smoothed. Each data point is the average of the original point and four neighbours. The points are about 0.6 nm removed from each other.

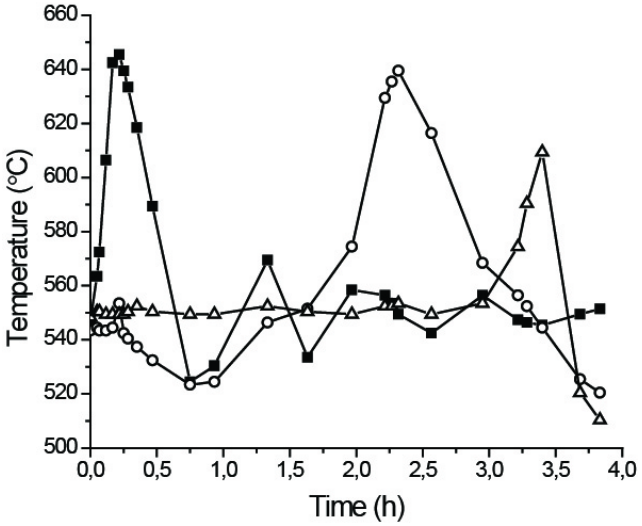

Fig. S5. Temperature read out of the top (■), middle (○) and bottom (Δ) thermocouples inside the pilot-scale reactor set-up during the oxidation/regeneration step. The oxidation provides extra heat to the system, which can be measured. This effect is visible at different times for the different thermocouples, first for the top, followed by the middle and bottom thermocouples. This is another clear indication that the combustion of coke takes place as a front along the reactor.