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In Situ Reactivation of Passivated Ni-on-Alumina Catalysts for Toluene Hydrogenation in a Dielectric Field

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In recent years quite some research has been carried out in microwave-assisted catalysis and in microwave-assisted catalyst synthesis. Presently we have tested the use of dielectric fields for *in situ* reactivation of passivated Ni-on-alumina using dielectric fields. As a model catalytic reaction, hydrogenation of toluene was employed. The reaction itself was studied using both dielectric and conventional heating, thereby assessing the impact of dielectric heating on catalyst structure and activity. Further, we have studied the application of a dielectric field to regenerate Ni-on-alumina deactivated by coke formation. It is shown that *in situ* activation of a passivated catalyst in a dielectric field is indeed possible, but *in situ* decoking is not likely to be feasible, because for moderate electric field strengths the temperatures attained by coke particles are insufficient to realize carbon methanation. © 2002 Elsevier Science (USA)

Key Words: **dielectric heating; catalyst reactivation; toluene hydrogenation; *in situ* decoking; Ni-on-alumina; microwaves.**

1. INTRODUCTION

Supported base and noble catalysts are often prepared using impregnation, deposition precipitation, or coprecipitation. Subsequently, the material is dried, calcined, reduced, and sometimes passivated. The passivated material can be reactivated in a reducing atmosphere. In recent years, various attempts have been made to replace conventional heating by dielectric heating in one or more synthesis steps.

In the preparation of porous support materials it has been observed that the crystallization of molecular sieves, zeolites, and hydrotalcites (1) proceeds (much) faster under dielectric heating. The underlying mechanism has as yet been resolved satisfactorily, however. As water efficiently absorbs microwaves, dielectric drying of solids has received much attention (2–5). Compared with conventional heating, a high drying rate is observed and the resulting product may show a lower and more homogeneously distributed residual moisture content. Liang *et al.* (6) showed that dielectric drying of a Mn/γ-Al_2O_3 catalyst, prepared by wet impregnation, is faster than conventional drying. Bond *et al.* (7), studying the dielectric drying of Ni/Al_2O_3 catalysts prepared by impregnation, demonstrated that also a more uniform metal distribution and a mechanically stronger catalyst pellet may be obtained. This result can be rationalized in view of the self-controlled nature of dielectric drying: the part of the pellet with the highest moisture content absorbs microwaves most efficiently. This effect is also known as moisture leveling, which is the reason for the more uniform metal ion distribution inside catalyst pellets dried in a dielectric field as opposed to those dried conventionally. Finally it was demonstrated that a small moisture gradient between the surface of the pellet may lead to reduced mechanical tension during drying and, thus, to higher mechanical integrity of the resulting catalyst pellet.

The high heating rates achievable by (bulk) dielectric heating lead to short processing times at elevated temperature and soaking of the catalyst at high temperatures is thus prevented. This may reduce the degree of metal sintering during calcination. Using V_2O_5-on-SiO_2 and Co_3O_4-on-carbon respectively, Liu *et al.* (8) and Kim and Jeon (9) showed that calcination in a dielectric field is faster than conventional heating but in addition may produce a catalyst with higher V_2O_5 and Co_3O_4 dispersion. In contrast, Sai Prasad *et al.* (10), using Pd-on-Al_2O_3 and Pd-on-SiO_2, observed that Pd particle size increases when calcination is carried out in a dielectric field. These authors speculate that the interaction between the palladium precursor (palladium nitrate) and the support is weakened during dielectric heating. In a study of the reduction of alumina-supported Pt catalysts in a dielectric field, Ringler *et al.* (11) show that the metal particles are slightly larger after dielectric heating than after reduction using conventional heating.

A range of authors have investigated the impact of dielectric heating on reaction rates. Roussy *et al.* examined this effect for alumina-supported Pt for isomerization of 2-methylpentane (12, 13), showing that selectivity toward

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the isomer increases due to exposure of the catalyst to a dielectric field (either during pretreatment or under reaction conditions). Roussy *et al.* speculate that dielectric heating induces a permanent surface modification of the Pt particles or a permanent modification of the Pt–alumina interface. The existence of a temperature difference between the Pt particles and the underlying support was assessed by Perry and co-workers (14, 15), on the basis of a quantitative study for alumina-supported Pd and Pt catalysts. In a study of the impact of dielectric heating on the gas-phase decomposition of H_2S using MoS/γ-Al_2O_3 catalysts, the observed rate enhancement was attributed to formation of hot spots (16). Additionally, phase changes in MoS and γ-Al_2O_3 were observed. Experimentally observed high reaction rates in the partial oxidation of methane over ZrO_2- or La_2O_3-supported Ni or Co in a dielectric field were also attributed to hot-spot formation, although changes in the surface composition of the catalysts were noted (17). Oligomerization of methane or oxidative coupling of methane in a dielectric field has been tried using several catalysts (18, 19). In all cases, increased selectivity toward C_2^+ hydrocarbons at lower temperatures than under conventional heating are reported.

Whereas the reaction rate and selectivity of a variety of solid catalysts are reported to be influenced by the use of dielectric fields, it is likely that nonthermal effects arise only from the impact of such fields on the structure of a catalyst. Selective heating of metal particles cannot be excluded from theoretical arguments, but quantitative studies shed serious doubt on this effect. It is nonetheless clear that dielectric fields may be used favorably in the synthesis of catalysts, as during drying, calcination, and reduction, and that potentially favorable structural modification may be induced during reaction. The impact of dielectric heating on the reduction of supported catalysts has barely been addressed.

In this work the influence of dielectric heating on the reactivation of passivated Ni-on-alumina catalyst was investigated. Ni-on-alumina is widely used in industry, as in syngas production, steam reforming, fat hardening, hydrogenation of aromatics, and isomerization of hydrocarbons (20). It is known that the Ni dispersion of Ni-on-alumina catalysts is affected by reduction conditions, such as temperature, hydrogen space velocity, and moisture content (21, 22). One may therefore speculate that efficient removal of water formed during reduction may lead ultimately to a modified catalyst.

Hydrogenation of toluene was chosen as a suitable model reaction to test the activity of these catalysts. We have studied this reaction using both dielectric and conventional heating, thereby assessing any changes in catalyst structure and activity. Finally, the feasibility of *in situ* removal of carbonaceous deposits, formed during reaction, using dielectric fields is addressed.

TABLE 1

Characterization of Sample Ni-5124

Parameter	Value	Analysis method
Ni content	58 w/w%	Inductively coupled plasma—atomic emission spectroscopy
S_{BET}	110 $m^2\ g^{-1}$	BET/N_2 adsorption
Pore volume	0.136 $cm^3\ g^{-1}$	BET/N_2 adsorption
Pore width	4.8 nm	Mercury porosimetry
He density	3.5 $g\ cm^{-3}$	Helium density
Ni particle diameter	11 nm	H_2 chemisorption and O_2 uptake

2. EXPERIMENTAL

2.1. *Catalyst Preparation and Characterization*

A commercial 58 wt% Ni/γ-Al_2O_3 catalyst (Ni-5124) from Harshaw Chemie B.V. was used. The as-received (reduced and passivated) catalyst was crushed and sieved to a particle size of $\leq$200 μm. This catalyst is referred to as sample Ni-5124. Characterization data are given in Table 1.

2.2. *Reactivation of the Passivated Catalyst*

To study the feasibility for *in situ* reactivation of passivated Ni catalysts by dielectric heating, reactivation was carried out by either conventional or dielectric heating.

2.2.1. Conventional heating. About 7 g of sample Ni-5124 was reduced in a H_2/He flow (v/v 2/1, GHSV $= 2 \times 10^2\ h^{-1}$) at 400°C for 2 h. During reduction, the temperature was initially raised to 150°C at 5°C min^{-1}, maintained at this level for 1 h, and subsequently raised to 400°C at a heating rate of 1°C min^{-1}. At this temperature level the catalyst was flushed with He (GHSV $= 2 \times 10^2\ h^{-1}$) for 1 h and subsequently cooled to room temperature under a He flow. When required, the sample was transferred to and from the microwave unit under a He flow, thus preventing reoxidation.

2.2.2. Dielectric heating. Sample Ni-5124 was first reactivated in a conventional furnace as described above. However, after being cooled to room temperature, the sample was passivated in a stream of O_2/He (0.5 vol% O_2, GHSV $= 2 \times 10^2\ h^{-1}$) for 2 h. The sample was subsequently transferred to the microwave unit under a He flow.

Reduction in the microwave unit was subsequently carried out using a H_2/He flow (v/v 2/1, GHSV $= 2 \times 10^2\ h^{-1}$) at 400°C for 2 h. The reduction temperature was first raised to 150°C at a heating rate of 5°C min^{-1} and maintained for 1 h. It was further raised to 240°C at 1°C min^{-1}. After reduction the sample was kept at 240°C in a He flow (GHSV $= 2 \times 10^2\ h^{-1}$) for 4 h and cooled to room temperature under a He flow.

2.3. Toluene Hydrogenation Experiments

The same sample was used throughout all the experiments to eliminate possible deviations in the results due to differences in catalyst packing. Toluene was obtained from by Acros (analytical grade) and used without further purification. Hydrogen (Praxair; 99.999% pure) and helium (Praxair, 1 vol% Ar) were further purified using an oxygen adsorbens and 3-Å molecular sieves. Experiments were carried out in a toluene/hydrogen/helium flow (vol% 8/19/73, GHSV $= 6 \times 10^2$ h^{-1}) at ambient pressure. Toluene was introduced using a Milton–Roy solvent delivery system (CM 4000). The reaction mixture was heated to 120°C before entering the reactor. The exit gas stream was passed to a gas chromatograph through heated lines. Methane (Praxair, 99.995% pure) was introduced into the exit gas stream as an internal standard. The activity and selectivity of the catalyst were assessed at 125°C, 175°C, and finally again at 125°C. Gas composition analysis was performed using a Hewlett–Packard gas chromatograph (5710A) equipped with a DB-wax capillary column (Alltech; 30 m; $d_{\text{in}} = 0.25$ mm; film $= 0.5$ μm; operating at a fixed temperature of 90°C) and a flame ionization detector.

2.4. Temperature-Programmed Reduction (TPR)

2.4.1. Analysis of sample Ni-5124. A conventional TPR setup was used. The sample is placed in a quartz tubular reactor, containing approximately 25 mg of sample, weighed to 0.1 mg accuracy. A H_2/Ar mixture (v/v 2/1) is used (SV $= 1 \times 10^2$ kg $\cdot$ s m^{-3}) and heating rates are 5 K s^{-1}.

2.4.2. TPR analysis after toluene hydrogenation. Following toluene hydrogenation, the catalyst is cooled to room temperature in a He flow (GHSV $= 2 \times 10^2$ h^{-1}) and subsequently transferred to a conventional furnace under a He flow. Reduction is carried out in a H_2/He mixture (v/v 2/1, GHSV $= 2 \times 10^2$ h^{-1}). The sample is heated to 450°C at 1°C min^{-1}. The exit gas stream was analyzed using a Balzers mass spectrometer (Prisma QMS 2000).

2.5. Dielectric Heating

The dielectric heating system (Fig. 1) consists of a microwave source (2450 MHz, 1 kW), a circulator, a three-stub tuner section, a monomode microwave cavity TE_{10}, and a water load. Reflection of the microwaves is minimized using stub tuners. The waveguide is formed by a copper rectangular channel with the dimensions 7.2 × 3.6 cm (width × height). The microwave setup is operated in traveling wave mode, with any microwave radiation passing through the sample cavity being absorbed by a water load.

A quartz tubular reactor (i.d. = 18 mm), designed to accommodate an optical fiber, is positioned perpendicular to the direction of propagation. Throughout all experiments the volume of sample was kept constant (10 cm^3).

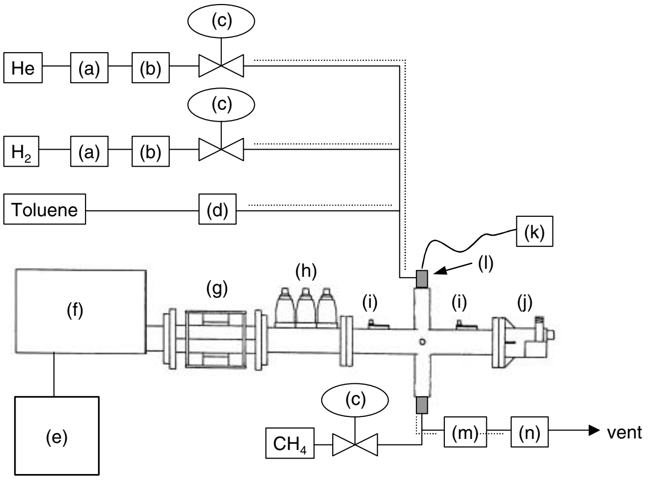

FIG. 1. Schematic drawing of 2.45-GHz microwave setup. Dashed lines denote heated tubing. (a) Oxygen sorbent, (b) Molsieve 3 Å, (c) mass flow controller, (d) toluene delivery system (Milton-Roy; CM 4000), (e) generator (Muegge, MW-GIR 2 M), (f) microwave source (Muegge, MW), (g) circulator (Philips), (h) stub tuners (Muegge), (i) power sensor (Rhode & Schwarz), (j) water load, (k) optical fiber (Luxtron, Accufiber—OFT straight end lightpipe), (l) quartz reactor tube, (m) gas chromatograph (HP 5710A), (n) quadrupole mass spectrometer (Balzers, Prisma QMS 2000).

The temperature of the sample during dielectric heating is controlled through a temperature control loop coupled to the microwave power and the temperature of the sample bed. During experiments in a conventional furnace, the temperature of the sample bed is monitored simultaneously with a thermocouple and an optical fiber, thereby allowing calibration of the optical fiber.

3. RESULTS AND DISCUSSION

3.1. Reactivation of Passivated Ni Catalyst

To assess the feasibility of *in situ* reactivation of passivated Ni catalysts by dielectric heating, these samples were reactivated either in a conventional furnace or in a dielectric field. During dielectric heating, it was noted that the absorbed microwave power does not vary with temperature, suggesting an approximately constant dielectric loss factor. However, using our dielectric heating setup, the dielectric loss factor for the passivated Ni catalyst is not sufficiently high to heat the sample to temperatures in excess of 240°C (Fig. 2). To investigate whether reactivation at 240°C is sufficient for complete reactivation, TPR data were obtained. In TPR of the passivated catalysts a main peak is observed at 200°C, reflecting the reduction of the passivation layer of oxygen (Fig. 3) (24). This layer consists of approximately two monolayers of oxygen (25). In TPR, reactivation is essentially complete at 240°C, with reactivation at 400°C leading to only a slightly higher degree of reduction. This

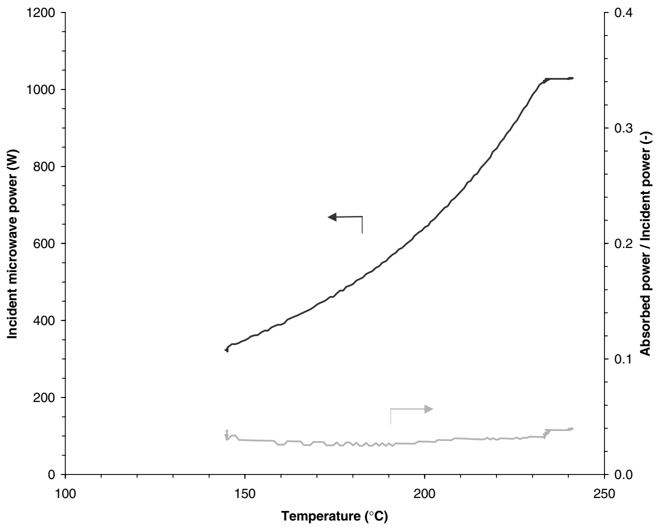

FIG. 2. Microwave absorption during reactivation of a passivated 58 wt% Ni/Al_2O_3 catalyst in a dielectric field. Heating at 1°C min^{-1} under a H_2/He flow (v/v 2/1, GHSV $= 2 \times 10^2$ h^{-1}). Incident microwave power (left axis) and absorbed microwave power (right axis) versus temperature.

suggests that reactivation of a passivated catalyst using the dielectric heating setup may well result in a near-complete reactivation.

This was further validated in activity tests. After reactivation by either technique described above, the catalysts were tested for their activity in toluene hydrogenation. Activity data for conventional heating at 125°C are presented in Fig. 4. The initial toluene conversion at 125°C does not depend on the reactivation method employed. No partially hydrogenated products were observed and thus selectivity toward methylcyclohexane is 100% for the catalysts, irrespective of the reactivation method (conventional or by dielectric heating).

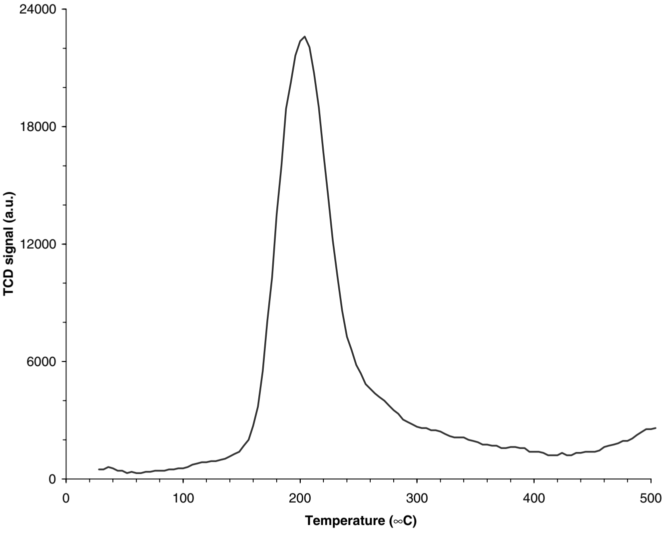

FIG. 3. TPR pattern of a passivated 58 wt% Ni/Al_2O_3 catalyst. See Experimental for details.

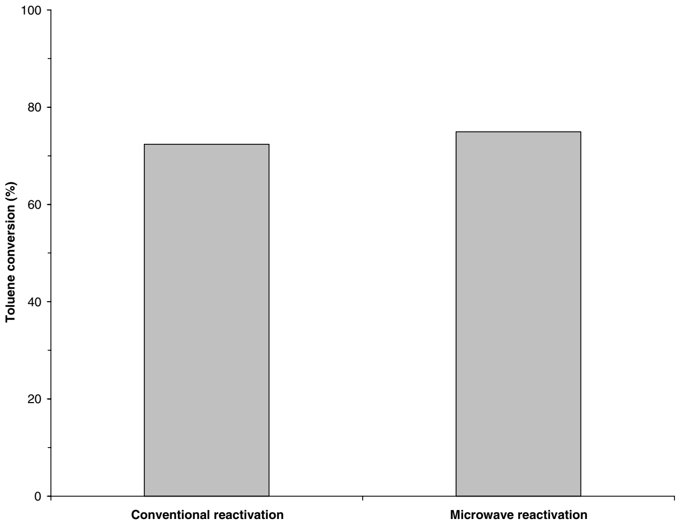

FIG. 4. Toluene conversion during toluene hydrogenation at 125°C using conventional heating for a reactivated 58 wt% Ni/Al_2O_3 catalyst. See Experimental for details.

3.2. *Effect of Heating Mode during Reaction on Catalyst Performance*

After reactivation by conventional heating the alumina-supported nickel catalyst was tested in toluene hydrogenation experiments using both conventional and dielectric heating (Fig. 5). The initial conversion of toluene at 125°C for dielectric heating is comparable to that observed under conventional heating, i.e., approximately 70%. The selectivity toward methylcyclohexane is 100% for both cases studied as no partially hydrogenated products were observed.

After the reaction temperature was maintained at 125°C for 12 h, it was raised to 175°C. This results in a drop in toluene conversion to 50%. For a hydrogen/toluene molar ratio of 2.3 as used in our study the equilibrium conversion of toluene at 125°C is around 90% (26). As the system is thus operated far from equilibrium, the lower conversion at a higher temperature is not attributable to equilibrium limitations. Toluene hydrogenation is thought to proceed through a reaction scheme involving (26): (1) associative adsorption of toluene and dissociative adsorption of hydrogen, (2) the subsequent surface reaction of the adsorbed species forming methyl cyclohexane, and (3) desorption of methyl cyclohexane from the catalyst surface. Lindfors *et al.* (26) studied the Ni/Al_2O_3-catalyzed hydrogenation of toluene, assuming that quasi-equilibrium conditions apply to step 1, the surface reaction (2) is rate-limiting, and desorption of methyl cyclohexane (3) is fast and irreversible. For these conditions, Lindfors *et al.* showed

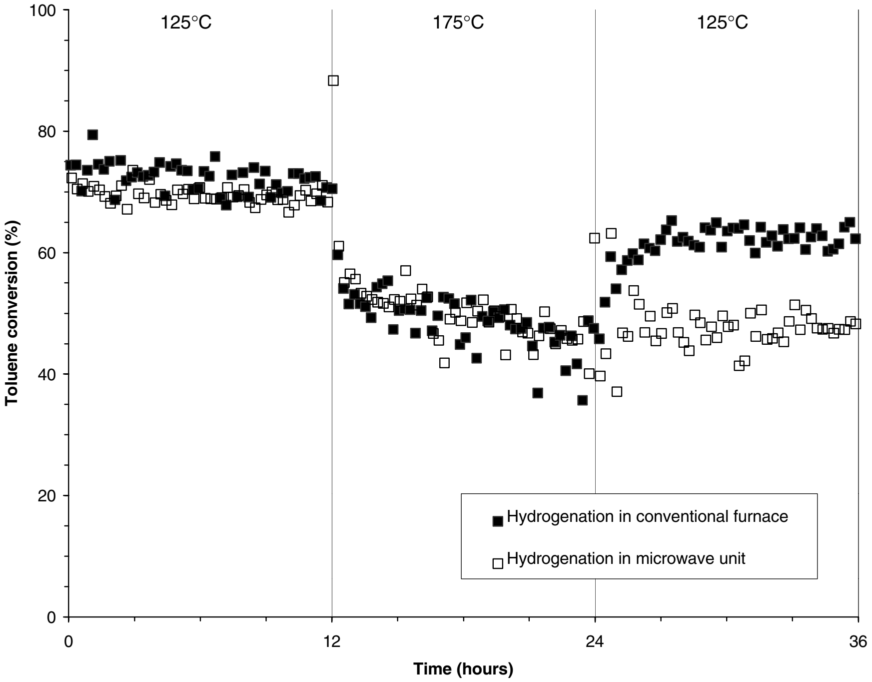

FIG. 5. Toluene conversion versus time-on-stream during toluene hydrogenation for a reactivated 58 wt% Ni/Al_2O_3 catalyst, using conventional heating. Toluene hydrogenation during conventional heating (■). Toluene hydrogenation during dielectric heating (□).

that the increase in the surface reaction rate for higher temperatures is overcompensated by the lower hydrogen adsorption equilibrium constant, the result being a lower overall reaction rate. This agrees with the presently observed lower reaction rate for a higher temperature.

Following a short period at 125°C, the catalyst is exposed to an extended time-on-stream (12–24 h) at 175°C. Toluene conversion at that temperature gradually decreases with time. After 24 h the temperature is restored to the initial temperature, but conversion falls short of the initial level. Clearly, some form of deactivation takes place. The final toluene conversions for the toluene hydrogenation reaction at 125°C are approximately 60 and 50% for conventional heating and dielectric heating, respectively. This suggests that for dielectric heating either structural modifications of the catalyst take place or deactivation is stronger.

In contrast to the other samples tested in this study, the passivated sample reactivated in the dielectric field and subjected to toluene hydrogenation in the microwave setup shows an increase in toluene conversion when the temperature is increased from 125 to 175°C (Fig. 6). Concurrently, some partially hydrogenated products are formed, which is not observed for the other samples. After 24 h the temperature is lowered to the initial temperature, 125°C, and the conversion decreases to around 40%.

Clearly the catalyst deactivates to a certain degree during operation at 175°C, as was also the case with the other samples tested in this study. The conversion of toluene at 175°C is comparable to the initial conversion. This was not observed with the passivated catalyst reactivated under conventional heating and reacting in a dielectric field. Reactivation under dielectric heating apparently induces structural changes in the catalyst, which influence both activity and selectivity. The exact nature of these changes is the subject of further work.

For the initial toluene hydrogenation activity at 125°C it hardly matters whether the passivated catalyst is reactivated by conventional or by dielectric heating (see Table 2): the initial toluene conversion for the hydrogenation in a dielectric field deviates only slightly from that observed for conventional heating.

For toluene hydrogenation in a dielectric field, the initial toluene conversion is higher when the passivated catalyst is reactivated conventionally (Table 2). Also, during toluene hydrogenation in a dielectric field the sample reactivated in a conventional furnace demonstrates enhanced microwave absorption as compared with the sample reactivated by dielectric heating (Fig. 7). This indicates that the degree of

TABLE 2

Initial Toluene Conversion at 125°C

Reactivation of passivated catalyst in	Toluene conversion (%) under	
	Conventional heating	Dielectric heating
Conventional furnace	72	69
Microwave unit	75	63

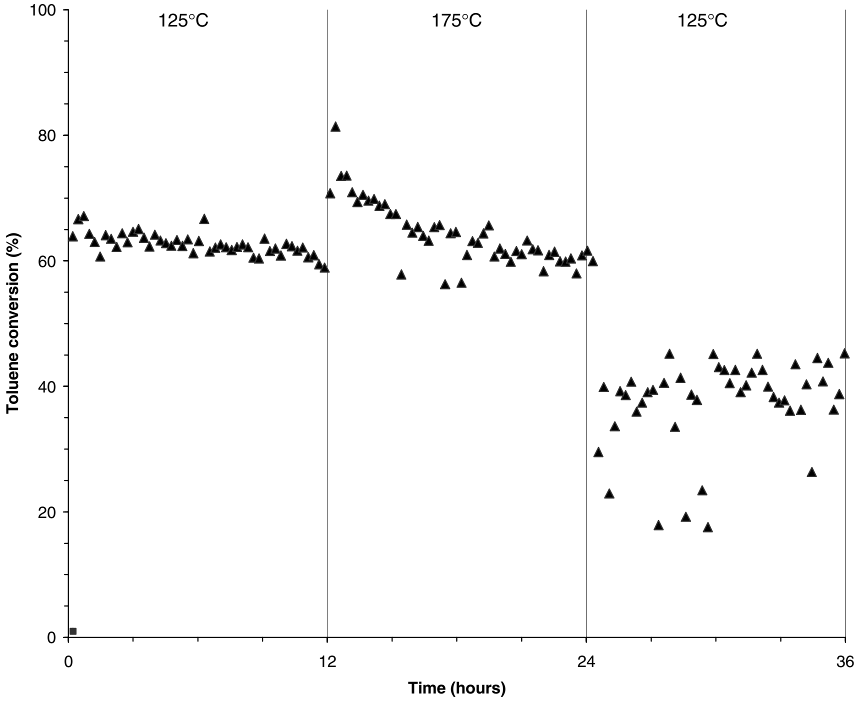

FIG. 6. Product analysis versus time-on-stream for toluene hydrogenation for a reactivated 58 wt% Ni/Al_2O_3 catalyst. Catalyst reactivation and toluene hydrogenation were performed under dielectric heating.

reduction after microwave reactivation is somewhat lower than that obtained using conventional reactivation. This is confirmed from an analysis of the dielectric properties. The microwave absorption is related to the dielectric constant of the material. Following reactivation in a microwave setup this dielectric constant is smaller than after reactivation in a conventional furnace. This also suggests that the degree of reduction of the catalyst after reactivation under

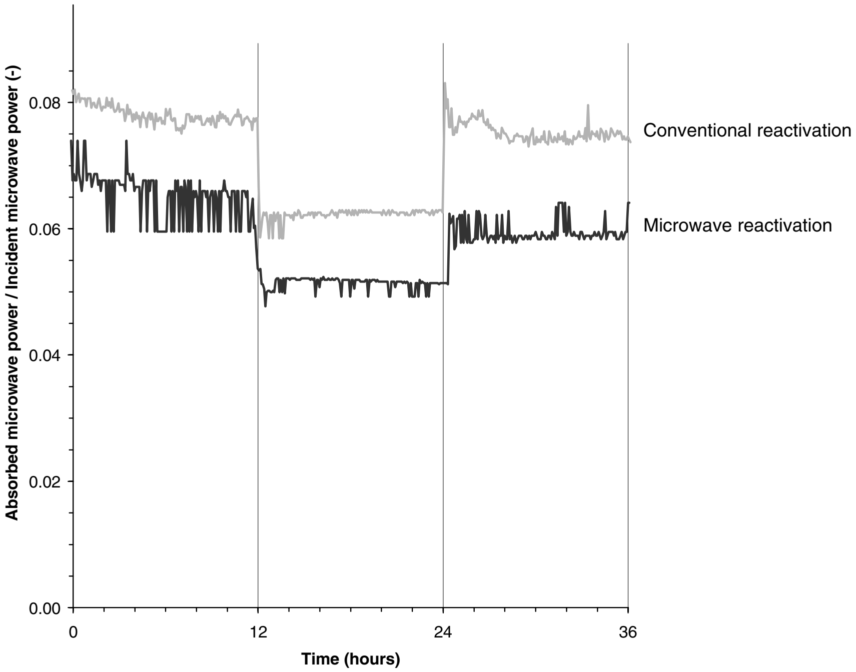

FIG. 7. Absorbed microwave power versus time-on-stream for toluene hydrogenation for a reactivated 58 wt% Ni/Al_2O_3 catalyst under dielectric heating.

dielectric heating is lower than after reactivation by conventional heating.

3.3. Catalyst Deactivation and in Situ Reactivation

To investigate the nature of the catalyst deactivation observed during toluene hydrogenation at higher temperatures, the catalysts obtained after reaction were subjected to TPR (Fig. 8). This analysis revealed three bands: one at 160°C, a shoulder at 250°C, and a broadband from 250 to 400°C. TPR bands at 160 and 250°C probably arose from desorption of CH_4 adsorbed on the surface of the catalyst (27). It is suggested that CH_4 is formed during coke buildup on the catalyst. The band at higher temperature is attributed to methanation of coke on the catalyst by H_2. During toluene hydrogenation, coke builds up on the acidic sites of the Al_2O_3 surface over benzyl intermediates (28), which react further to produce a variety of coke precursors. Catalysts used in toluene hydrogenation under conventional heating contain a comparable amount of coke, irrespective of the reactivation method. Beyond 400°C formation of CH_4 ceases, indicating the complete regeneration of the catalyst.

The surface area of the bands in Fig. 8 is proportional to the amount of coke present in the catalysts. Catalysts operated under dielectric heating during toluene hydrogenation clearly contain more coke than catalysts operated under conventional heating, in agreement with the observed stronger deactivation of these samples. As the peak maximum of the broadband shifts toward higher temperature when the toluene hydrogenation reaction is performed under dielectric heating, it may be concluded that coke deposited under these conditions is less reactive in methanation, and can be removed only at higher temperatures.

When conventional heating is used during toluene hydrogenation the amount of coke formed and the location of the peak temperature observed in TPR (around 300°C) are independent of the heating method employed during catalyst reactivation. Therefore, reactivating the passivated catalyst in a dielectric field prior to toluene hydrogenation does not influence the deactivation process during the subsequent reaction.

To study whether the catalyst regains its initial activity in toluene hydrogenation after the temperature is lowered from 175 to 125°C, the catalyst was kept on stream for several hours more (Fig. 9). It was observed that the initial activity of the catalyst is not regained in any of the four experiments. After deactivation at 175°C and lowering the temperature to 125°C, the activity and selectivity of the catalyst remain constant, regardless of the heating method employed (microwave or conventional).

From TPR analysis after toluene hydrogenation it was observed that all samples contain coke. The catalysts subjected to dielectric heating under toluene hydrogenation conditions contained more coke than the samples subjected to conventional heating. Clearly dielectric heating does not lead to *in situ* methanation of carbon.

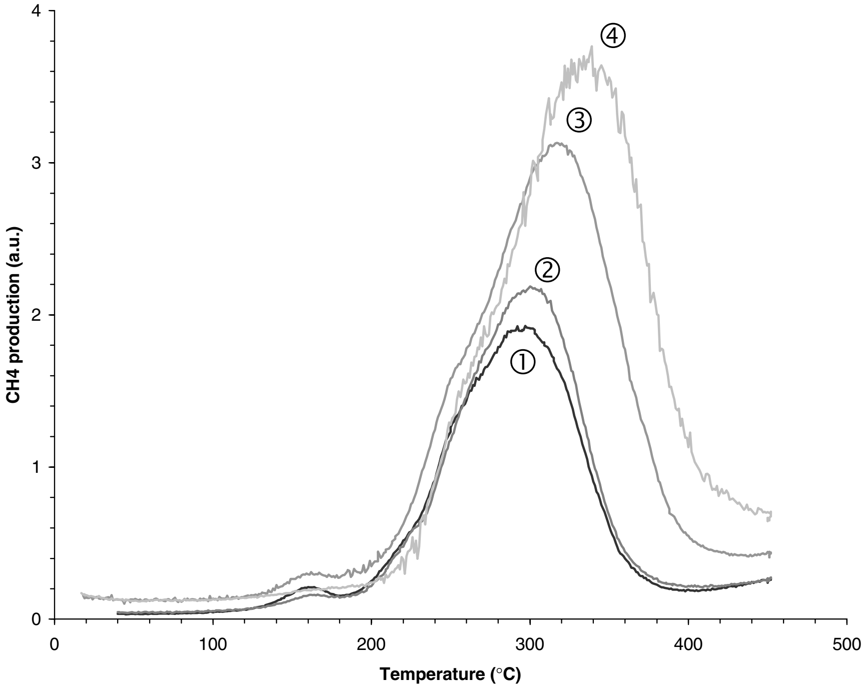

FIG. 8. Methane formation during TPR of catalysts following use in toluene hydrogenation. Heating in "reactivation" and "toluene hydrogenation": (1) conventional–conventional, (2) microwave–conventional, (3) conventional–microwave, (4) microwave–microwave.

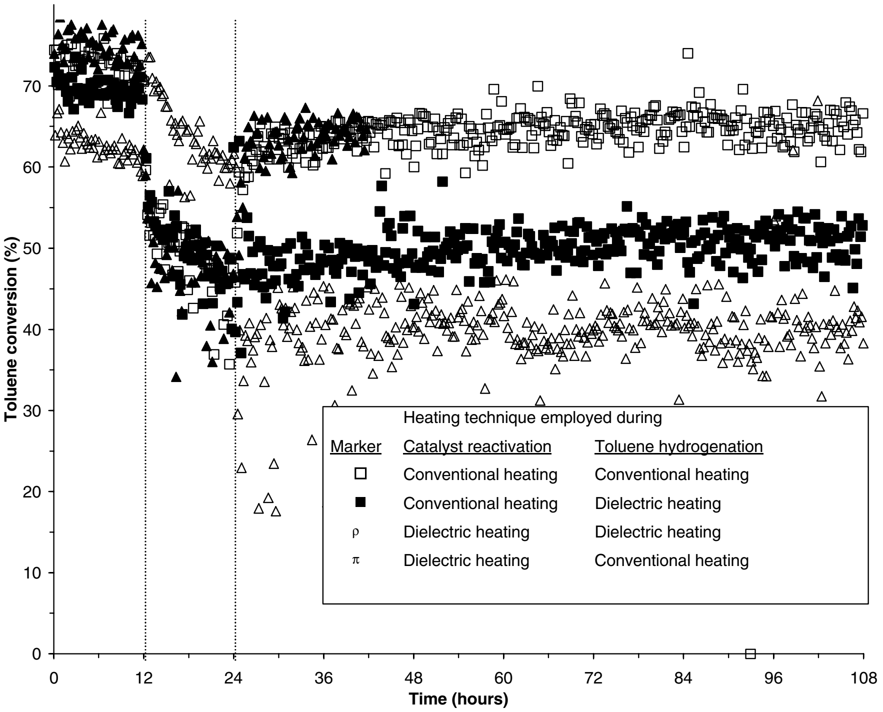

FIG. 9. Toluene conversion versus time-on-stream for prolonged toluene hydrogenation experiments over a 58 wt% Ni/Al_2O_3 catalyst.

4. CONCLUSIONS

The influence of dielectric heating on the reactivation of passivated Ni/Al_2O_3 and the subsequent performance in the hydrogenation of toluene were investigated. Both reactivation of the passivated catalyst and toluene hydrogenation experiments were carried out by conventional and dielectric heating.

The degree of reduction of the catalyst after reactivation in a dielectric field is lower than for conventional heating, as a result of the lower reactivation temperature attainable in a dielectric field. However, this is not reflected in the activity of the catalyst in toluene hydrogenation. When this reaction is carried out in a dielectric field, the initial toluene conversion is slightly lower than that obtained in conventional heating. This may be due to enhanced coke formation, even at temperatures as low as 125°C (see below).

A higher reaction temperature results in a lower toluene conversion, due to the fact that the reaction is limited by hydrogen sorption. Surprisingly, the sample reactivated in a dielectric field and subjected to microwaves also during reaction shows a higher toluene conversion when the temperature is raised. Concurrently, some partially hydrogenated products are formed, whereas no such products were observed for the other catalyst samples. This strongly suggests that dielectric heating induces structural changes in the catalyst. One may speculate that these may in general be the predominant factor explaining the impact of dielectric heating on reactivity of catalysts.

Toluene hydrogenation at high temperature leads to partial deactivation of the Ni-on-alumina catalyst by coke formation. Once the catalyst has been deactivated, *in situ* reactivation of the catalyst by selective heating and subsequent methanation of carbonaceous deposits in a dielectric field does not appear feasible at bulk catalyst temperatures of 125°C, despite the high dielectric loss factor of carbon. In contrast, larger amounts of coke are observed for catalysts operated in a dielectric field. This coke can in principle be removed by methanation under hydrogen, but complete regeneration of the catalyst is feasible only at temperatures as high as 400°C.

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