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The Effect of Steam on Supported Metal Catalysts

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In order to examine the effect of steam on supported metal catalysts, model supported metal catalysts of Ni, Co, or Fe on alumina have been heated in steam at 700°C. The transmission electron micrographs show that for all these metals, patches of film extend from the crystallites. Prolonged heating results in the disappearance of the patches which probably spread as a contiguous film over the entire surface of the substrate. The degree of spreading is in the order: Co > Ni > Fe. On subsequent heating in H_2, small crystallites were generated, probably via the rupture of the contiguous film. The contraction of the patches of film bridging two or several particles caused the coalescence of the latter. This subsequent heating in H_2 favors redispersion only when the heating time is sufficiently short. Prolonged heating in H_2 leads to the disappearance of the small particles.

INTRODUCTION

Generally speaking, the influence of water on the characteristics of supported metal catalysts is inevitable because it is involved in every stage of their preparation. In addition, many supported metal catalysts are used in reaction atmospheres in which water exists either as a reactant or as a product (for instance, steam reforming or methanation). The presence of water is usually considered to cause deactivation (*1–6*), either because it enhances sintering, or because it promotes phase transformations in the crystallites.

Studies of the effect of water have been carried out with the ceramic materials used as catalysts or as catalyst substrates, such as silica–alumina or alumina. In the case of silica–alumina, Morgan *et al.* (*7*) concluded that the presence of steam accelerates the process of sintering. This process begins with the growth of necks between two adjacent particles and then advances to form large compact particles.

The loss of surface area in the case of alumina is also strongly enhanced by the presence of steam (*6*). Besides sintering, Schaper *et al.* (*8*) reported that the presence of water vapor enhances the transformation of alumina to the α phase.

As concerns the supported metal catalysts, water influences the dispersion of the metal crystallites in various ways, such as: (1) the promoting of chemical transformation in the crystallites; (2) the strengthening or weakening of the interactions between crystallite and support; (3) the carriage of impurities, such as chloride, which contribute to the dissolution of the transition metals.

Few investigators have examined the effect of steam alone on supported metal catalysts. Dowden (*1*) reported that even small amounts of water accelerate the sintering of the crystallites in supported metal catalysts. Bartholomew and Farranto (*9*) observed that by increasing the hydrogen space velocity during the reduction of nickel supported on alumina, the nickel surface area was significantly decreased, probably because of the water which is generated during reduction. Borowiecki *et al.* (*2*) also noted that the rate of sintering is strongly affected by steam (about a threefold increase with respect to the rate of sintering in the absence of steam). On the other hand, as reported by Katsobashvili

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(*4*), the chemical transformation of the metal crystallites may also constitute a major reason for deactivation. Ruckenstein and Chu (*10*) observed redispersion of Pt particles during heating in wet N_2 and Sushumna and Ruckenstein (*11*) reported that the presence of traces of moisture in as-supplied extra-pure hydrogen enhanced the effects caused by the traces of oxygen, i.e., the extension of iron particles on the substrate as well as the formation of iron aluminate.

However, there is no direct evidence on the changes in crystallite morphologies and on the relationship between the morphological changes and the chemical transformation of the crystallites during heating in steam at elevated temperatures. Moreover, most investigators studied the effect of water in a gas mixture instead of steam alone. This does not allow to isolate the effect due to steam from those due to the other components of the mixture. For these reasons, the present authors have examined, by using model catalysts and Transmission Electron Microscopy, the effect of steam at 700°C on the behavior of Ni, Fe, or Co crystallites supported on alumina.

EXPERIMENTAL

Preparation of Model Catalysts

Thin, electron-transparent, nonporous, alumina films, used as supports for model catalysts, were prepared by anodizing high-purity aluminum foils (99.999%, Alfa Products, Inc.) by the technique described elsewhere (*12*). After being heated at 800°C for about 40 h, the alumina films were transformed to the γ phase and were ready for metal deposition.

Deposition of Metal

Nickel or iron were deposited on alumina films by evaporating high-purity metal wires (99.998%, Alfa Products) from a tungsten basket in an Edwards Vacuum Evaporator under a vacuum of better than 2×10^{-6} Torr. Cobalt was deposited on alumina films by evaporating cobalt powder (99.998%, Alfa) in the same vacuum evaporator.

Heating in Steam and Hydrogen

Samples were laid on a quartz boat inside a quartz tube of about 1 in. in diameter and 2 ft. in length, and were heated in a heavily insulated furance. The temperature of the furnace was controlled with an OMEGA Series 920 Solid State Conroller, with a maximum deviation of ±3°C.

Steam was carried by helium (Ultrahigh purity grade supplied by Linde Division, Union Carbide Co.) which was bubbled at the rate of 150 ml/min through distilled water contained in a gas washing bottle. The ratio of steam to helium was maintained at 3 : 1 by immersing the gas washing bottle in an isothermal bath and adjusting the bath temperature to about 95°C. In order to prevent the condensation of steam upstream of the furnace, the connecting line between the furance and isothermal bath was jacketed in a tube and maintained at the bath temperature by circulating the bath fluid through the jacket.

The amount of steam which passed through the furnace could be measured from the condensed water collected downstream of the furnace.

Before heating in steam, the samples were heated in ultrahigh pure hydrogen (Linde Division, Union Carbide Co.) at 500°C for 5 h, and further at 700°C for at least 4 h, in order to reduce as much as possible the metal oxide and the aluminate formed by the interaction of the oxide with alumina. The samples were heated in steam at 700°C in several steps up to 16 h, and finally heated in hydrogen again at 700°C also in several steps for 3 h. Before being taken out for observation, the samples were cooled down to room temperature in a helium atmosphere. The observations were made in the same region of the samples after each heat treatment by using a JEOL 100U TEM.

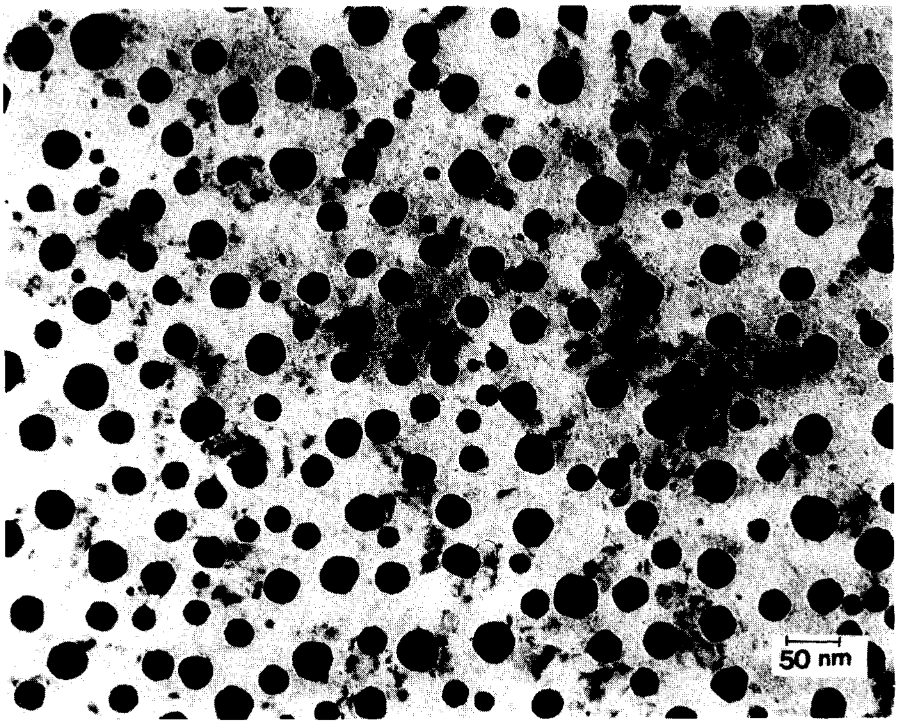

FIG. 1. Sintered particles of Ni/Al_2O_3 after heating in H_2 at 500°C for 5 h and at 700°C for additional 4 h.

RESULTS

The Morphologies of the Crystallites on Heating in Steam

a. Ni/Al_2O_3. An initial Gaussian distribution of well-defined circular-shaped crystallites was obtained after heating in hydrogen at 500°C. Because most of the steam reforming catalysts are used at 700–1000°C, the temperature of 700°C was chosen as the continuous heating temperature. Large particles of 400–500 Å appeared after heating in H_2 at 700°C for 4 hours (Fig. 1). Further heating in H_2 did not produce a significant change. Subsequently, the specimen was heated in steam at 700°C. During heating in steam for only half an hour, "feet" were extended from particles over the surface of the substrate (Fig. 2, particles A). The external diameter of the extended particles is larger than that of the original ones, while the diameter of the core is decreased.

During additional heating, the feet detached from the main bodies of some particles to form rings around the latter. After heating for 2 h, the detached and undetached feet of particles spread out, generating patches of local films surrounding several particles (Fig. 3, regions B and C).

On further heating, the local films begin to disappear, tending to spread over the surface of the substrate. After heating in steam for about 10 h, they disappear completely and a contiguous film undetectable by electron microscopy probably covers the surface of the substrate (Fig. 4). No changes in the size and shape of the large particles were observed during the formation period of the contiguous surface film.

The curves of Fig. 5 show a monotonic decrease of the particle population and of the average particle size during heating in steam. The decrease in the number of particles is probably due to the disappearance

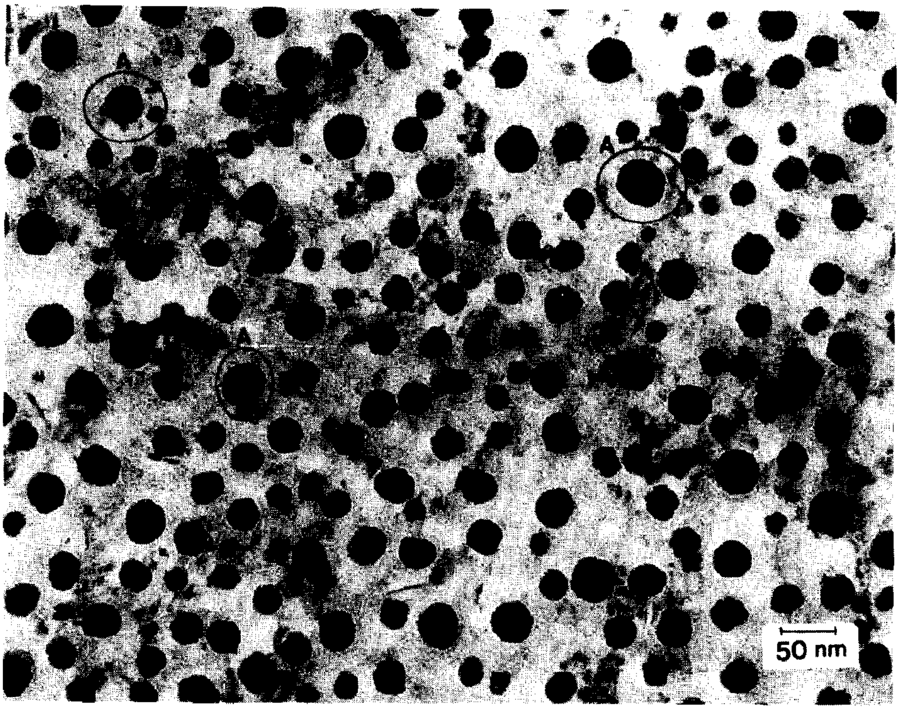

FIG. 2. The appearance of "the feet" at the peripheries of the particles of Ni/Al_2O_3 on heating in H_2O at 700°C for 0.5 h.

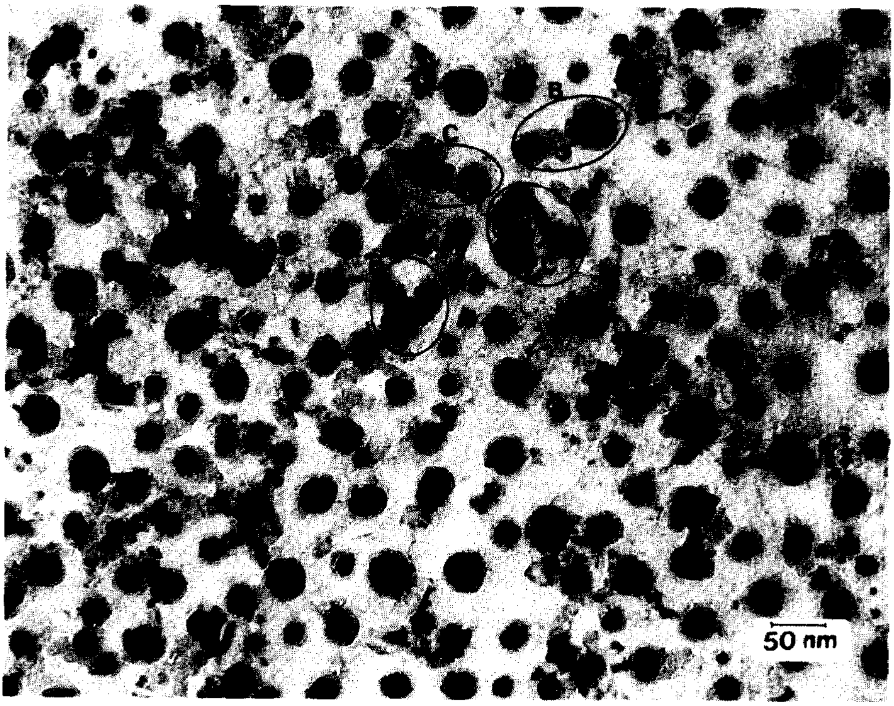

FIG. 3. The formation of patches of film on heating in H_2O at 700°C for 2 h.

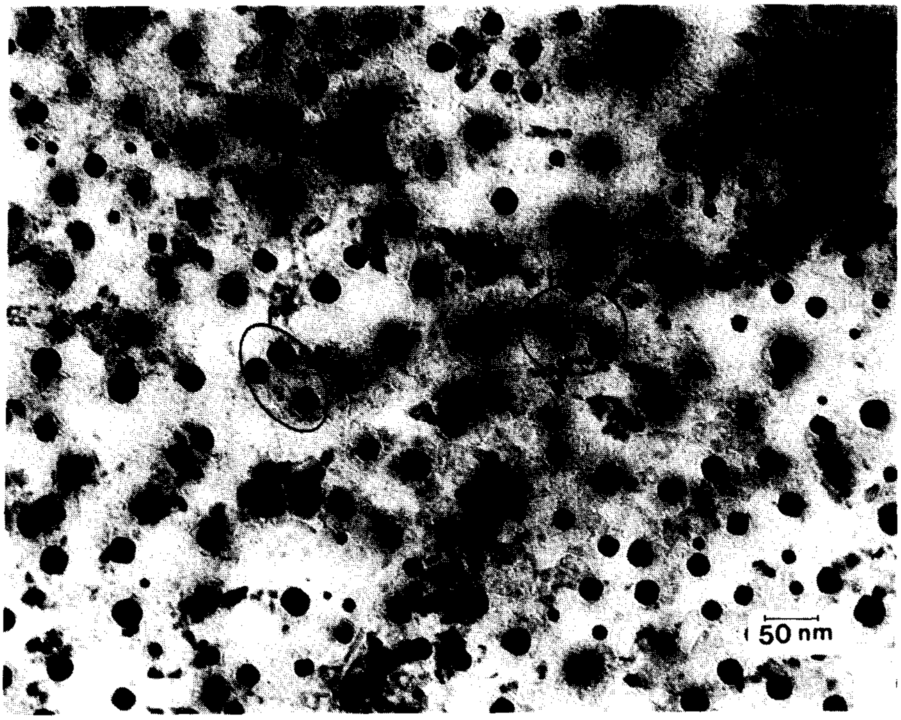

FIG. 4. Disappearance of patches on heating in H_2O at 700°C for a total of 16 h.

by spreading of the smaller particles. The paticle population in the final state is four-fifths of that in the original state, while the average particle size is about two-thirds of the initial one.

b. Co/Al$_2$O$_3$. A longer time was needed for cobalt to form particles during heating in H_2 of the deposited film of Co. The formed particles tend to extend on the surface of the substrate even during heating in H_2 (Fig. 6). On heating the specimen in steam for 20 min, the particles ruptured into a number of smaller units as a result of considerable extension (Fig. 7, particles A, compared with Fig. 6, particles A). Compared to the nickel–alumina system, most of the particles of the cobalt supported on alumina consist of much smaller compact cores and wider films around them. During additional heating, the cores maintained approximately the same size they acquired after 20 min of heating, but the patches of film around them extended further. No further change was observed after heating for 2 hours; by contrast, it took more than 10 h before no further change could be observed with Ni/Al_2O_3.

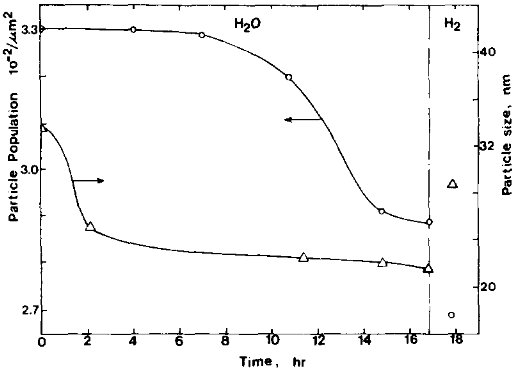

FIG. 5. The monotonic decrease of the average particle size and of the particle population during heating in steam; the rapid change of the particle size and population on subsequent heating in H_2.

c. Fe/Al$_2$O$_3$. It is known that iron is easily oxidized (*11*). Apparently, there is already

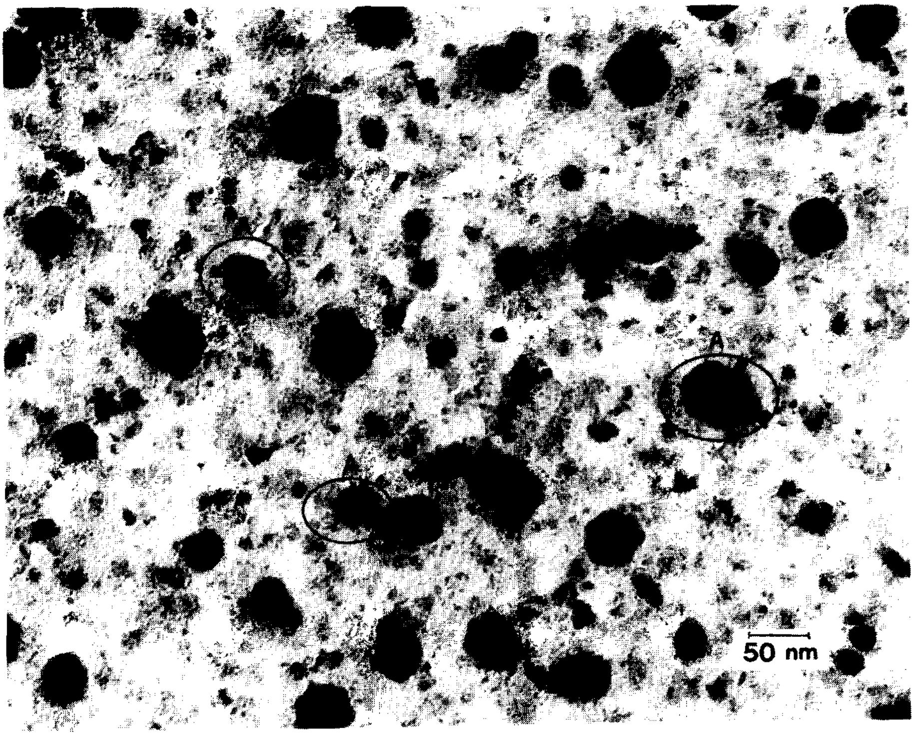

FIG. 6. Sintered particles of Co/Al_2O_3 after heating in H_2 at 500°C for 12 h and at 700°C for additional 6 h.

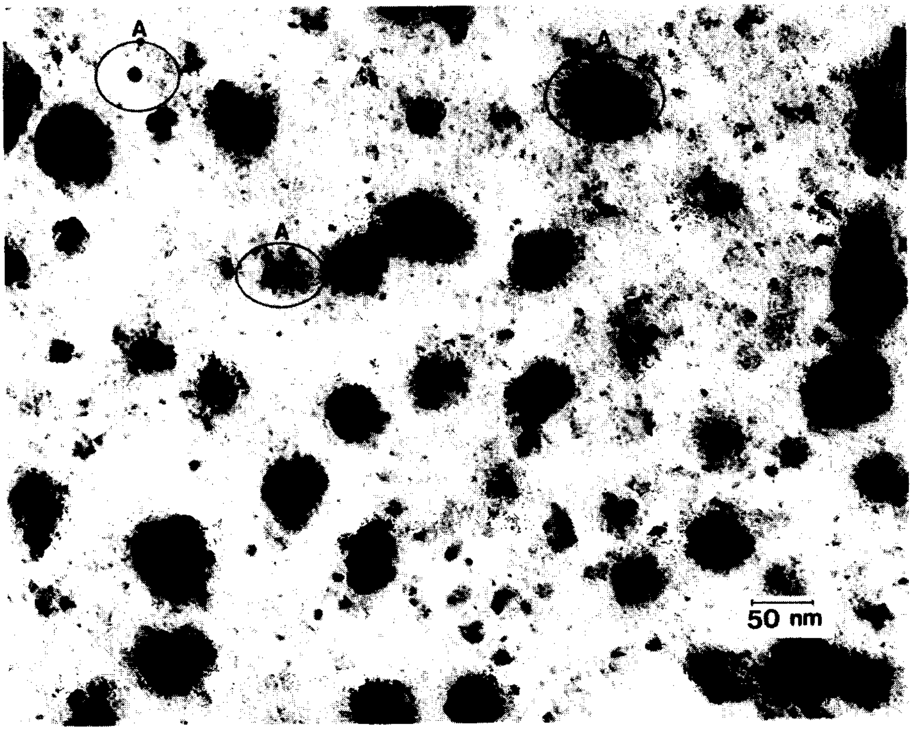

FIG. 7. Extensive extension of particles of Co/Al_2O_3 after heating in H_2O at 700°C for 20 min.

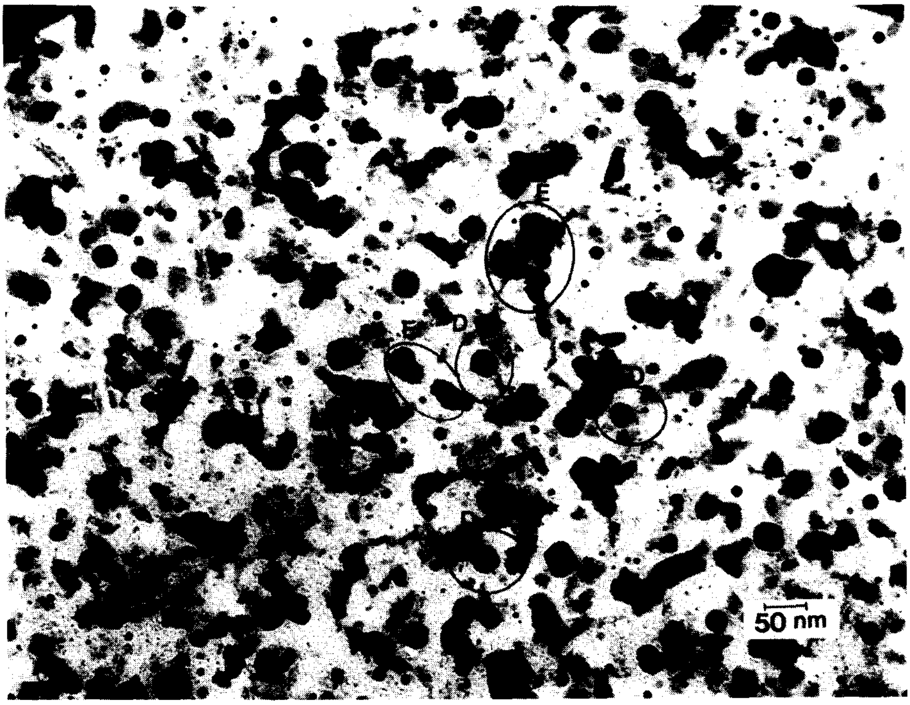

FIG. 8. The formation of patches of film in the neighboring regions of the particles for Fe/Al_2O_3 after heating in H_2 at 500°C for 5 h and at 700°C for additional 5 h.

a film spread over the substrate even before heating in steam. In the present experiment, patches of films could be observed in the neighboring regions of the particle after heating in H_2 for 10 h (Fig. 8, regions E). Accordingly, not much change of the particle size and particle population were observed after subsequent heating in steam even after ten hours of heating. However, the patches of the local films probably spread as a largely undetectable film over the surface of the substrate.

The Morphologies on Subsequent Heating in Hydrogen

a. Ni/Al_2O_3. When the specimen was subsequently heated in hydrogen, the contiguous surface film ruptured and contracted into patches. Several neighboring particles were connected by such patches. The bridged particles might have been pulled to migrate toward each other and to coalesce into a single particle (Fig. 9, regions F, compared with Fig. 4, regions F), which, after further heating, acquired a circular shape (Fig. 10a, regions F).

In addition, some new small crystallites, about 20 Å in size, were formed on the substrate (Fig. 10a, regions H). Some of these small particles disappeared later (Fig. 10b, regions H). A decrease in the population of the large particles because of their sintering by migration and coalescence and an increase in the average particle size was observed (see Fig. 5).

b. Co/Al_2O_3. Figure 11 shows that a large number of cobalt crystallites were formed again on heating in hydrogen for half an hour. Many of the new particles were formed in the neighboring regions of the initial particles that spread during heating in steam. It is therefore likely that the formation of the new particles is a result of the collapse and contraction of the detectable and undetectable surface films that surrounded the remaining compact cores of

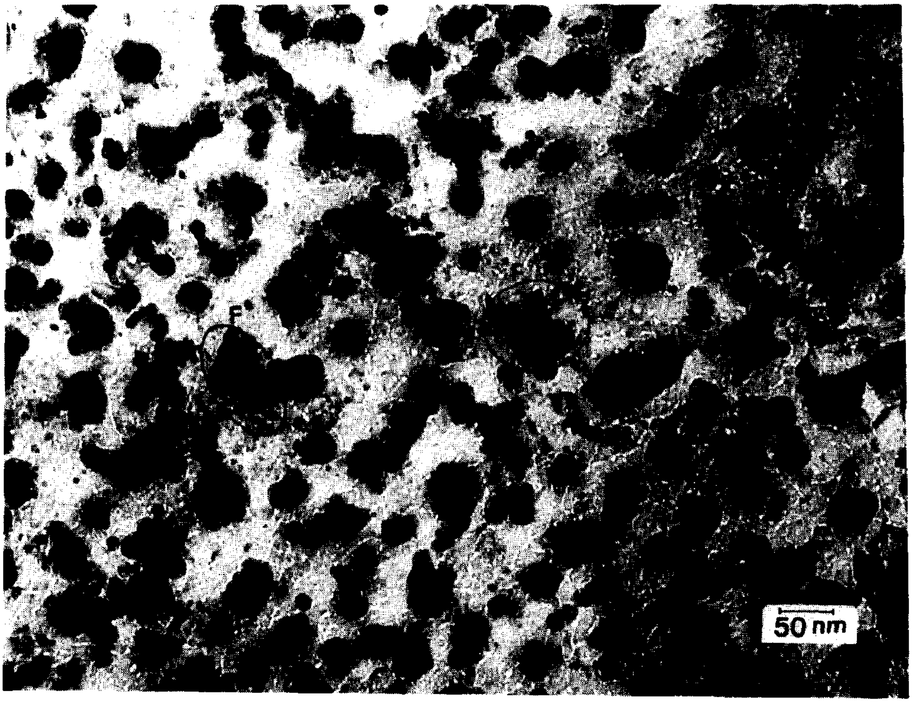

FIG. 9. The rupture and contraction of the surface film for Ni/Al_2O_3 after subsequent heating in H_2 for 1 h (compare with Fig. 4).

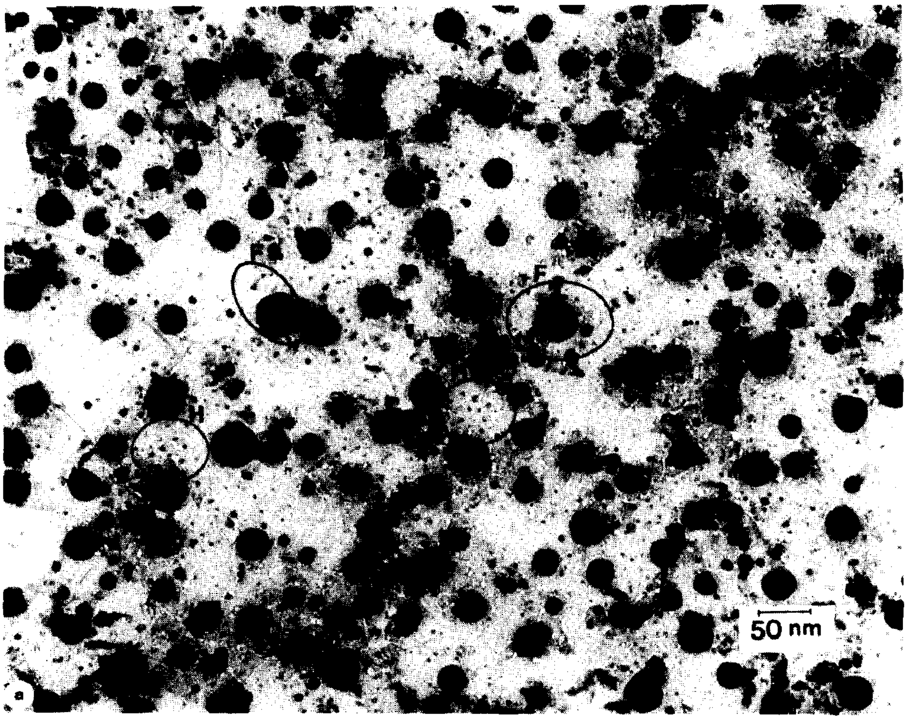

FIG. 10. (a) The complete merging of some film-bridged particles and the appearance of some small particles on heating Ni/Al_2O_3 in H_2 at 700°C for additional 2 h. (b) The disappearance of the small particles on additional heating in H_2 at 700°C for 1 h.

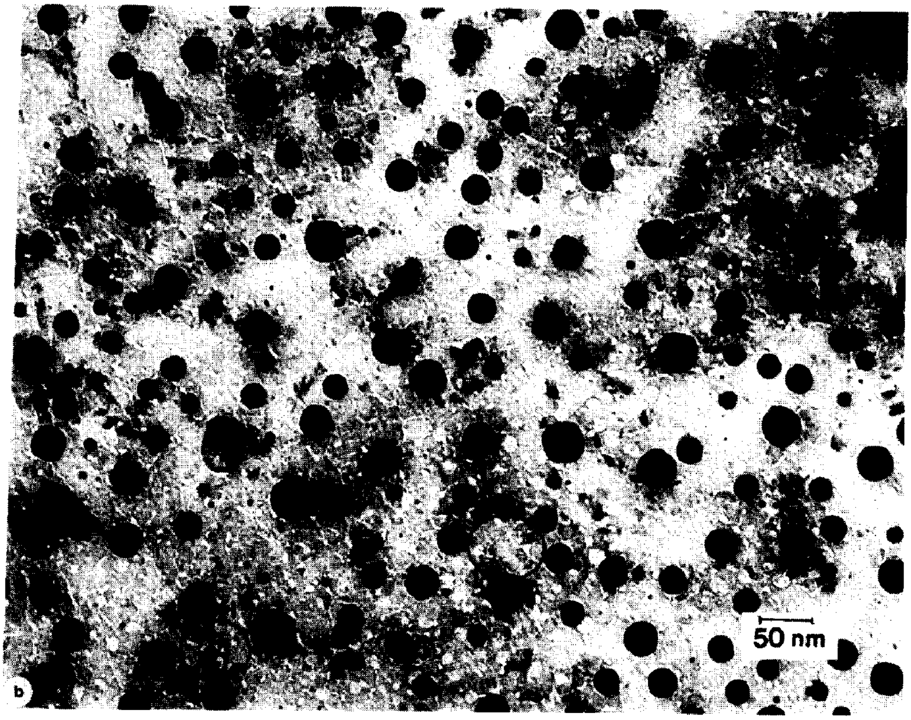

FIG. 10—*Continued.*

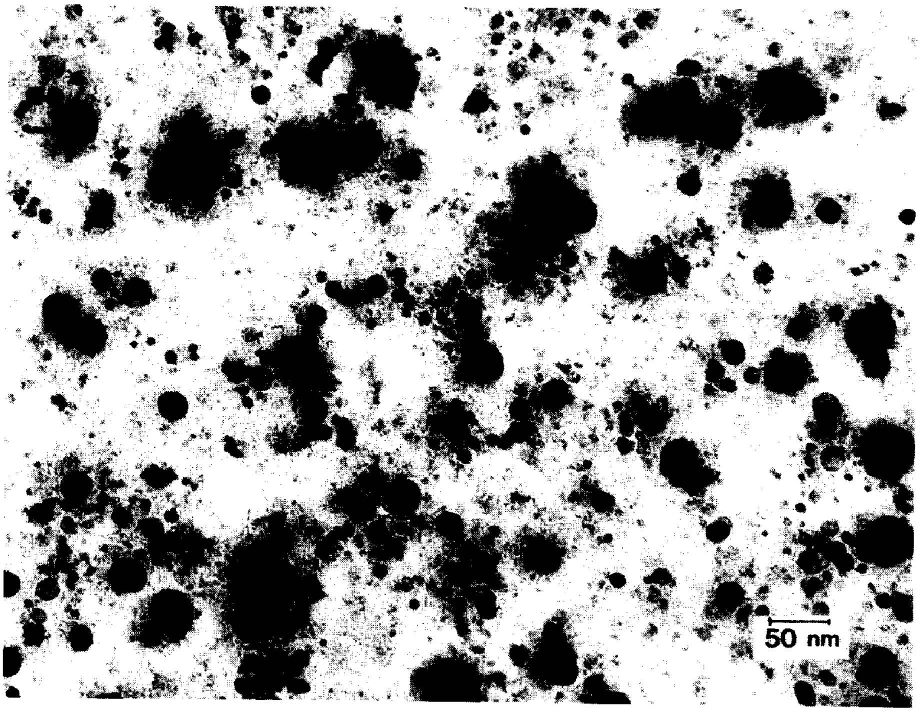

FIG. 11. The redispersion of the particles of Co/Al_2O_3 after subsequent heating in H_2 at 700°C for 0.5 h of the specimen previously treated in steam.

TABLE 1

Comparison of the *d*-Spacings of the Three Strongest Rings in the Diffraction Pattern of Ni/Al_2O_3 after Heating in Steam at 700°C

No.	ASTM card, *d* (Å)			Experimental						
	Al_2O_3	$NiAl_2O_4$	NiO	H_2			H_2O[a]			
				9 h	9.5 h	13 h	5.0 h	8.0 h	12.0 h	14.0 h[b]
1	2.39	2.43	2.41	2.4	2.45	2.44	2.52	2.50	2.53	2.53
2	1.98	2.01	2.09	1.99	2.00	2.02	2.07	2.04	2.07	2.09
3	1.40	1.42	1.48	1.41	1.41	1.43	1.46	1.45	1.47	1.48

[a] The time indicated is the summation of hours of heating in steam.

[b] No further change on heating beyond 14 h.

the particles (*24*). Note that a large number of new particles are smaller than the original ones (compare Fig. 11 with Fig. 6).

c. Fe/Al_2O_3. The appearance of patches of films around the particles (Fig. 12) proves once again indirectly the existence of the surface films which were formed even before the heating in steam and spread out extensively during the subsequent heating in steam. The large particles coalesce to form even larger particles (Figs. 12 and 13, regions K). On the other hand, a larger number of new small crystallites appeared in Fe/Al_2O_3 than in Ni/Al_2O_3. A bimodal distribution of particle sizes is therefore observed after heating for 3 h (Fig. 13).

The Formation and Transformation of Spinel Structure on Heating in Steam

As reported by other investigators (*8*), iron, cobalt as well as nickel react with alumina in the presence of traces of oxygen and form aluminates, which are difficult to reduce in hydrogen.

For nickel on alumina, nickel aluminate could be detected once the crystallites were generated on heating in as-supplied extrapure hydrogen (which contains 1 ppm oxygen). In the same reduction conditions, the lines of the iron aluminate in the diffraction pattern have been even more pronounced than those of nickel aluminate. No cobalt aluminate could be, however, detected until heating in steam. Hence, the readiness of aluminate formation in as-supplied extrapure hydrogen is in the order

$$Fe > Ni > Co.$$

One may note that Ruckenstein and Lee (*13*) detected only Ni (no $NiAl_2O_4$) in almost the same reduction conditions and by using a similar preparation method of the model Ni/Al_2O_3 catalyst. This may be a result of the difference in the heating times employed for the preparation of the alumina substrate. In the present experiment, the heating time was shorter, about 40 h, while in the former it was greater than 72 h. The excess of residual hydroxyls might be responsible for the formation of aluminate.

As indicated in Table 1, the *d*-spacings of the three strongest lines in Ni/Al_2O_3 kept changing during heating in steam. The initial heating led to the formation of aluminate which, during further heating, transformed to a structure intermediate between those of aluminate and nickel oxide.

The changes in the *d*-spacings for Fe/Al_2O_3 is somehow more complicated. The initial stages of heating in steam caused the transformation of $FeAl_2O_4$ to an unknown structure and further heating led to $Al_2Fe_2O_6$. Finally, only Fe_3O_4 (or Fe_2O_3 which has the same lattice parameter) was detected. The changes in the *d*-spacings for Fe/Al_2O_3 are given in Table 2.

No further change in the structure of the cobalt aluminate which was formed during

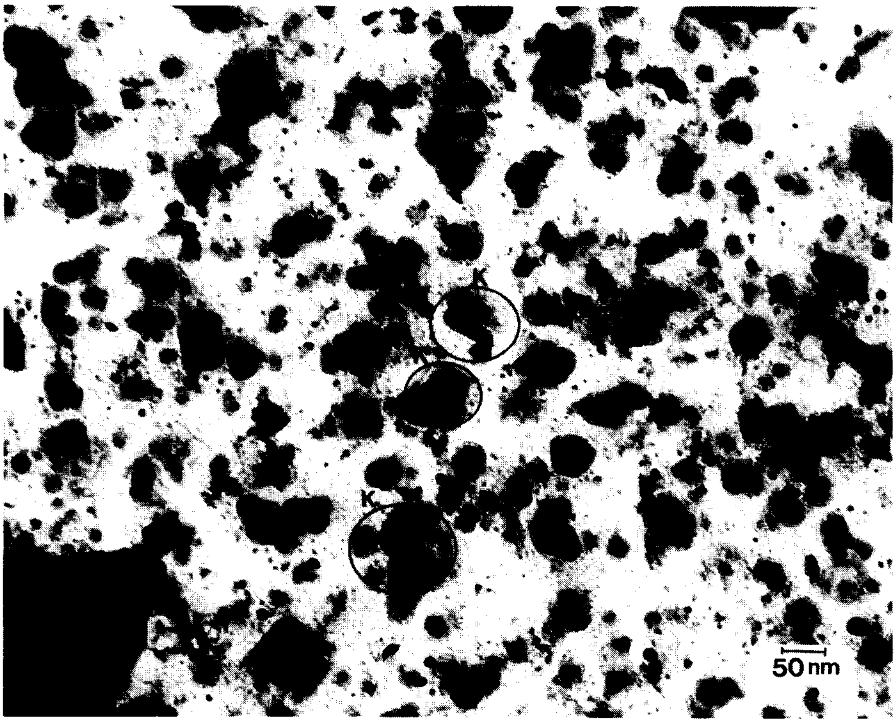

FIG. 12. The reapperance of patches of films around the particles of Fe/Al_2O_3 after subsequent heating in H_2 at 700°C for 1 h of the specimen previously treated in steam.

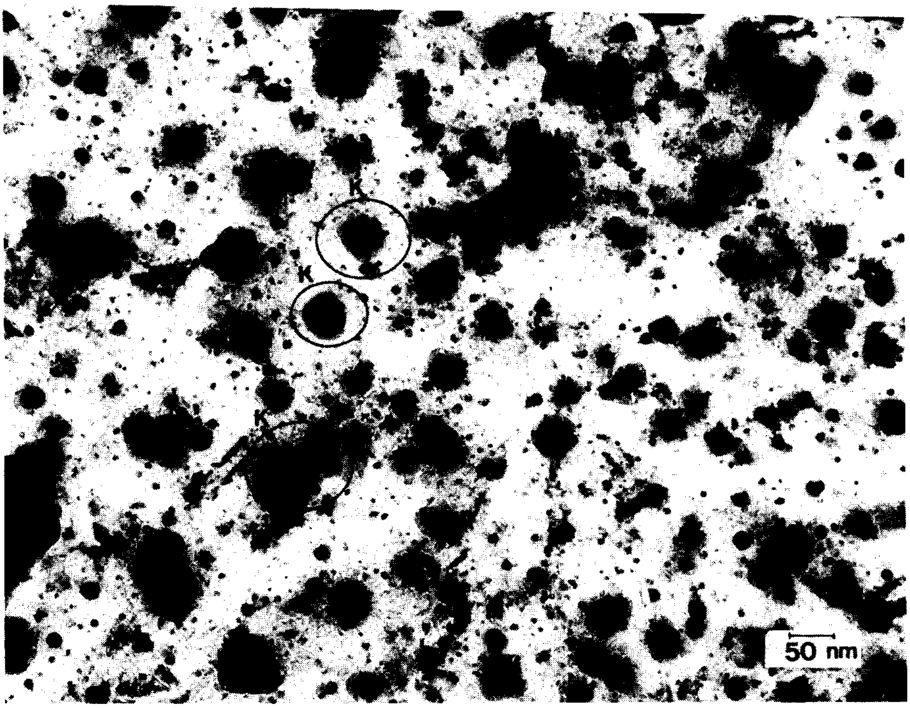

FIG. 13. The complete merging of large particles and the appearance of a large number of small particles in Fe/Al_2O_3 after heating in H_2 at 700°C for additional 2 h.

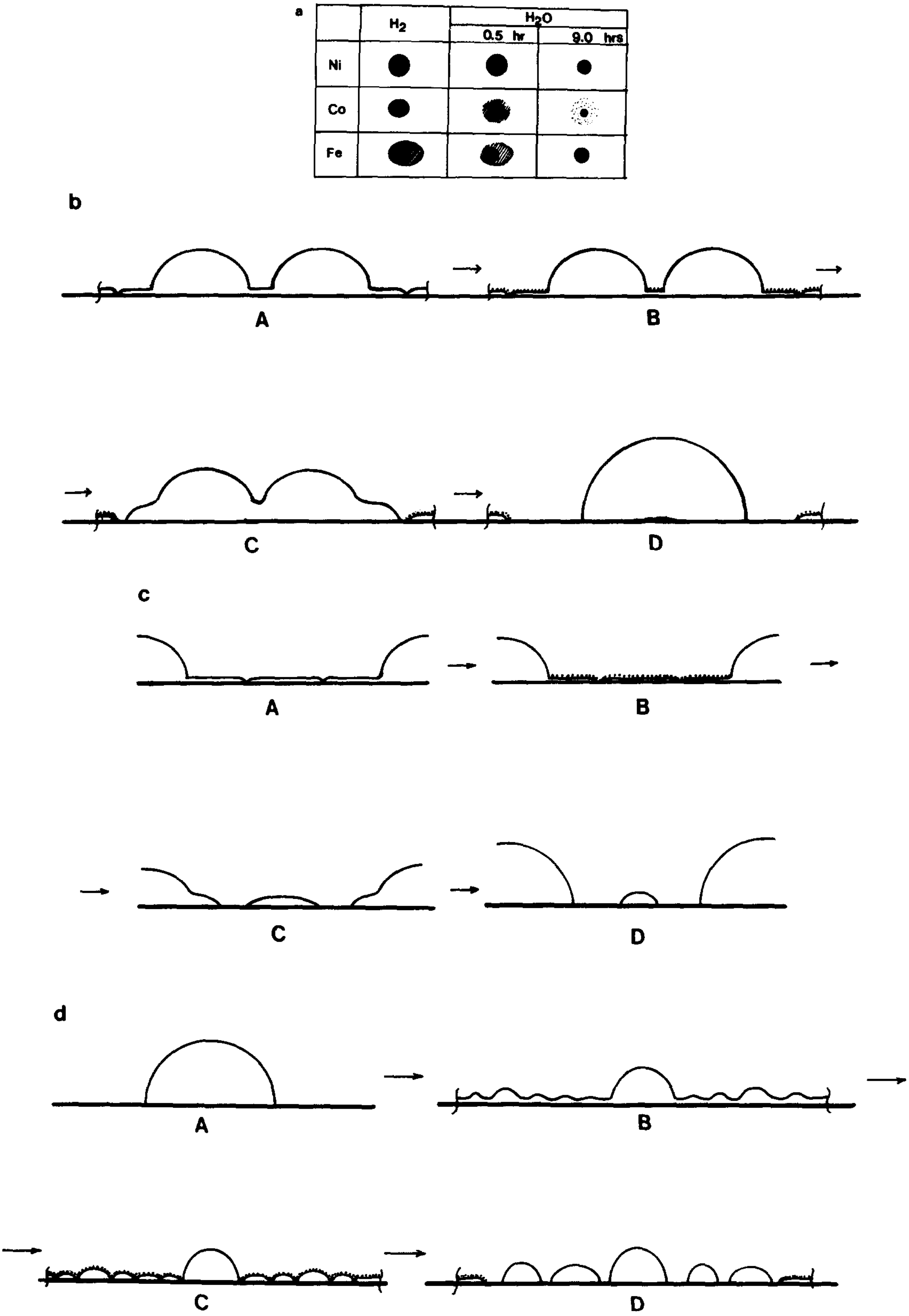
a
H_2
H_2O
0.5 hr
9.0 hrs
Ni
Co
Fe
b
A
B
C
D
c
A
B
C
D
d
A
B
C
D

TABLE 2

Comparison of the *d*-Spacings for Fe/Al_2O_3 after Heating in Steam

No.	ASTM card, *d* (Å)			Experimental							
	$FeAl_2O_4$	$Al_2Fe_2O_6$	Fe_3O_4	H_2			H_2O				
				10 h	10.5 h	12 h	2.0 h	3.5 h	5.0 h	8.0 h	10.0 h[a]
1	2.45	2.63	2.53	2.39	2.42	2.45	2.46	2.49	2.64	2.53	2.53
2	2.02	1.48	1.61	1.97	2.00	2.02	2.02	2.07	1.67	2.08	2.08
3	1.43	1.43	1.48	1.39	1.42	1.43	1.43	1.45	1.48	1.48	1.46

[a] No further change on heating beyond 10 h.

the initial stages of heating in steam was detected during continued heating in steam.

DISCUSSION

Comparison Between Ni, Fe, and Co on Al_2O_3 Heated in Steam

Figure 14a summarizes the morphologies of the particles formed on heating in steam at 700°C for the lengths of times indicated. Clearly, the morphologies are somewhat different for the three metals. In the case of Ni, only parts of the particles extend as feet upon the surface of the substrate, and then the feet spread over the entire surface, the spreading process being relatively slow. In the case of Co, almost entire particles extend very fast over the substrate. In contrast, in the case of Fe, the particles show no change at all because patches of film were already formed during the previous heating in as supplied extra pure H_2. Nevertheless, the experiments with all these metals suggest that a contiguous surface film is formed during heating in steam. As already proposed by Ruckenstein and Chu (*10*), a monolayer (or a multilayer) film appears to coexist with particles on the substrate.

Using Ni on alumina as an example, a possible scenario for the formation of the film could be described as follows.

1. The extension of feet from the particles through their leading edges and formation of thick films around the particles.
2. The coalescence of the thick films and

FIG. 14. (a) A schematic representation of the morphologies of the particles for Ni/Al_2O_3, Co/Al_2O_3, and Fe/Al_2O_3 before and during heating in H_2O. (b) A schematic representation of the coalescence of large particles caused by the rupture and contraction of the surface film for systems such as Ni/Al_2O_3 or Fe/Al_2O_3 after subsequent heating in H_2. (A) The coexistence of large particles and a surface film on heating in steam for a sufficiently long time. (B) Change in surface film on reduction of aluminate on subsequent heating in H_2. (C) The rupture and contraction of the surface film at some special locations where the surface film has cracks. (D) Further contraction of the surface film and complete merging of the two film-bridged particles. (c) A schematic representation of the formation of some small particles in systems such as Ni/Al_2O_3 and Fe/Al_2O_3. (A) The coexistence of a surface film and particles on heating in H_2O. Note that the distance between the two particles is relatively large. (B) The reduction of aluminate on subsequent heating in H_2. (C) The rupture and contraction of the surface film as in Fig. 14b.C (D) Further contraction of a piece of the ruptured surface film which is not connected to any particle and the formation of a small particle. (d) A schematic representation of the redispersion of the particles for Co/Al_2O_3: (A) A sintered particle before heating in H_2O. (B) The extensive extension of the particle and formation of a patch of film of nonuniform thickness around the particle on heating in H_2O. (C) The splitting of the patch of film after the formation of a layer of reduced aluminate on subsequent heating in H_2O. (D) The further contraction of the pieces of the split patch to form several small particles.

formation of patches of films as bridges between neighboring particles.

3. Complete spreading of these local films to form an undetectable monolayer (or multilayer) surface film.

The occurrence of these processes can be mainly attributed to changes in the interfacial free energies of the metal and between the metal and substrate which can lead to enhanced wetting. As noted before (*11*), the chemical interactions at the interface, such as those between the oxidized metal and the oxide substrate, can strongly decrease the interfacial free energy between the two phases. In addition, the metal oxide has a lower interfacial free energy than the metal. The dynamics of the process, which can be treated along the lines already employed for the dynamics of spreading of droplets on a solid (*14, 15*), will be examined elsewhere.

As an oxidizing agent, steam can therefore enhance the wetting and spreading of the metal oxide over the surface of the substrate. In contrast water adsorbed between metal (nonoxidized metal) such as gold and the substrate can decrease the interactions between the metal and substrate (*16*), thus decreasing the wetting of the latter by the former. As a result, the crystallite can have a higher mobility on the surface and enhanced sintering can occur (*17*). The later mechanism is not valid for Fe, Co, and Ni, since in these cases the presence of steam leads to an oxide and further to the formation of metal aluminate (*11, 18–20*). Consequently the chemical interactions leading to an oxide and to aluminate at the metal substrate interface are responsible for the extension of the particles, for the formation of the feet and for the spreading of the patches. In the case of Fe/Al_2O_3, an oxygen partial pressure of 10^{-15} atm is sufficient to oxidize Fe to Fe_2O_3 at 550°C (*21*). Such an oxygen partial pressure or moisture partial pressure is present even in the ultrahigh pure hydrogen. As a result, aluminate forms even in the hydrogen atmosphere (for details see Ref. (*11*)). In contrast to Fe, strong interactions between Co_3O_4 and Al_2O_3 and dramatic decreases in the interfacial free energies for Co/Al_2O_3 occur probably particularly in steam. As a result, the particle shape is likely to undergo a rapid change after heating in steam. Accordingly, the degree of spreading of the particles during heating in steam decreases in the order:

$$\mathrm{Co} > \mathrm{Ni} > \mathrm{Fe}.$$

Note that this order is reversed compared to that of the ease of aluminate formation in hydrogen mentioned before. It is known that the zero-valent metal (*22*) constitutes the active phase which is responsible for catalyzing the steam reforming reaction. However, on heating in steam, the crystallites change to a bottom layer of aluminate and a cap of metal oxide. In addition, whenever oxide molecules spread on the surface of the substrate they combine with alumina to form aluminate. The cap of the particles may in turn be partly or entirely covered by a skin of aluminate if the aluminate can spread over oxide on further heating. Although the spreading of particles increases the exposed area of the crystallites, the active surface area is however decreased because aluminate is inactive in the steam reforming reaction. Consequently, the deactivation of the catalysts that occurs on heating in steam can be largely a result of phase transformation rather than sintering. Borowiecki (*2*) observed that, even though there is no major change in the mean diameter of the crystallites measured by the method of X-ray line broadening, the active metal surface, measured by chemisorption, is decreased. The formation of aluminate can explain this effect.

Our observations also indicate that, after sufficiently long heating in steam, the aluminate transforms to a metal oxide in the case of Fe/Al_2O_3 and to a structure intermediate between nickel aluminate and nickel oxide in the case of Ni/Al_2O_3. The transformation of aluminate is possibly due to the formation of an inverted spinel structure $(Ni_{1-x}Al_y)$ $[Ni_xAl_{2-y}O_4]$ (where the parentheses and brackets stand for tetrahedral and octahedral sites, respectively). Hoster *et al.* (*23*) observed such a transformation to a partial inverted spinel structure when studying the steam reforming Ni catalysts by X-ray photoelectron spectroscopy.

The Competition of Redispersion with Sintering on Subsequent Heating in H_2

One interesting phenomenon, which was observed after subsequent heating in H_2, is the occurrence of redispersion of particles followed by coalescence.

Let us examine the processes occurring during heating in H_2. The first step of this process is the reduction of the surface film which is very likely composed of aluminate. The film will rupture at those locations where it contains cracks (see Ref. (*24*) for a detailed mechanism) and the resulting patches contract because of the enhanced surface free energy of metals compared to oxides. If particles coexist with a contiguous undetectable film which covers the substrate, such as in the cases of Fe/Al_2O_3 and Ni/Al_2O_3, then contraction of the patches which connect the particles causes the bulk movement of particles toward each other (Fig. 9, regions F). The particles "communicate" either by the transport of atoms via the film which bridges them or via the above mentioned contraction of patches. The result in both cases is their sintering. This sequence is schematically represented in Fig. 14b. Small crystallites are formed by the fracture of the undetectable surface film as indicated in Fig. 14c. These small crystallites are more mobile so that they may disappear soon due to their capture by other particles. If almost the whole particle extends over the substrate, such as in the case of Co/Al_2O_3, the subsequent heating in H_2 splits the film into several pieces which further contract to small particles as schematically shown in Fig. 14d. Thus, redispersion can dominate during initial heating. Comparing Figs. 6 and 11, one can see that the average particle size of Co after heating in H_2 is indeed smaller than the original one and that the particle population is increased. Consequently, the active metal area is increased in comparison to that before heating in steam. Sintering will, however, occur on further heating in H_2.

CONCLUSION

The presence of steam at high temperature causes the spreading of crystallites in the systems for which the interactions between crystallites and support are strong. Among Ni, Fe, Co, Co is the most sensitive to steam and Fe is the least sensitive.

Although steam appears to be able to redisperse the sintered particles of catalysts at high temperature, it is not always suitable for the regeneration of the sintered catalysts because:

(a) Steam will always decrease the surface area of porous support at high temperatures.

(b) Crystallites may transform from the active phase to an inactive phase, for instance, to metal aluminate.

(c) Traces of impurities in steam, such as HCl, can dissolve the transition metals (Pt, Pd, Ni).

However, on subsequent heating in H_2, either redispersion followed by sintering, or simultaneous redispersion and sintering followed by sintering occur, depending upon the degree of previous spreading in H_2O. If the whole particle has spread and the temperature was relatively low, redispersion is dominant during the initial heating. If, however, only a part of the particle has spread, a bimodal particle size distribution is obtained.

Consequently, heating in steam followed by sufficiently low temperature hydrogen treatment may regenerate some sintered catalysts provided the particles spread during heating in steam.

The film fracture during hydrogen treatment may also occur even without a previous steam treatment, since an oxide or aluminate film can form during the preparation of the catalyst.

Furthermore, if the reduction agent is mixed with steam, both sintering and loss of active phase can be the cause of catalyst deactivation.

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