NASA TECHNICAL MEMORANDUM

NASA TM-76651

N82-14309

DETERMINATION OF A KINETIC REGION IN CATALYTIC OXIDATION OF CARBON MONOXIDE

M. Yu. Sultanov, Kh. A. Sadykhova

Translation of "K voprosu ob opredelenii kineticheskoy oblasti pri kataliticheskom okislenii okisi ugeroda", Azerbaydzhanskiy Khimicheskiy Zhurnal, No. 2, 1980, pp. 3-6.

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION
WASHINGTON D. C. 20546 NOVEMBER 1981

STANDARD TITLE PAGE

1. Report No. NASA TM-76651	2. Government Accession No.	3. Recipient's Catalog No.
4. Title and Subtitle: DETERMINATION OF A KINETIC REGION IN CATALYTIC OXIDATION OF CARBON MONOXIDE		5. Report Date NOVEMBER 1981
		6. Performing Organization Code
7. Author(s) M. Yu. Sultanov, Kh. A. Sadykhova		8. Performing Organization Report No.
		10. Work Unit No.
9. Performing Organization Name and Address SCITRAN Box 5456 Santa Barbara, CA 93108		11. Contract or Grant No. NASw-3542
		13. Type of Report and Period Covered Translation
12. Sponsoring Agency Name and Address National Aeronautics and Space Administration Washington, D.C. 20546		14. Sponsoring Agency Code

15. Supplementary Notes

Translation of "K voprosu ob opredelenii kineticheskoy oblasti pri kataliticheskom okislenii okisi ugeroda", Azerbaydzhanskiy Khimicheskiy Zhurnal, No. 2, 1980, pp. 3-6.

16. Abstract

It has been noted that catalysts with a low content of active substance in a number of cases display higher effectiveness than pure active substances. This project studied the activity of cupric oxide activated with ceric oxide in a broad interval of volumetric (100-50,000 h^{-1}) velocities.

It was determined for practical catalysts used in the diffuse region, dilution of the active substance by an inert diluent can be a method of increasing the effectiveness of the catalyst.

Seven pages, three Figures, four References.

17. Key Words (Selected by Author(s))	18. Distribution Statement

19. Security Classif. (of this report)	20. Security Classif. (of this page)	21. No. of Pages	22. Price
Unclassified	Unclassified	7	

UDC 546.65:62-405.8

DETERMINATION OF THE KINETIC REGION IN CATALYTIC OXIDATION OF CARBON MONOXIDE

M. Yu. Sultanov and Kh. A. Sadykhova*

Catalysts with a low content of active substance in a number of /3** cases display higher effectiveness than pure active substances. This phenomenon is explained by different reasons: effect of the carrier, increase in specific surface of the active substance, etc. The hypothesis has been advanced that the observed effect may be the result of difficulties in heat and mass transfer [1].

We accordingly studied the activity of cupric oxide activated with ceric oxide in a broad interval of volumetric velocities (100-50,000 h^{-1}).

The active substance in all the studied specimens was cupric oxide, activated by 0.1% ceric oxide. The content of cupric oxide in the catalyst that was obtained by granulating the active substance ("pure") was 12 mole/l. The catalysts obtained by granulating the active substance in a mixture with aluminum oxide ("mixed") had a cupric oxide content of 0.05, 0.1, 0.5, 1.0, 1.5 and 2.0 mole/l. The catalysts were roasted at 900°C. The activity was determined on a flow-type unit with carbon monoxide concentration equal to 1%. The carbon monoxide concentration was determined on a flow-type unit with carbon monoxide concentration equal to 1% [sic]. The carbon monoxide concentration was determined on a KhT-2M chromatograph. In certain cases, the activity was characterized by the temperature at which CO was 50% oxidized.

Figure 1 presents the dependence of the catalyst activity on their content of active substance. As is apparent from the figure, with all

*M. A. Azizbekov Azerbaydzhanskiy Institute of Oil and Chemistry

**Numbers in margin indicate pagination in original foreign text.

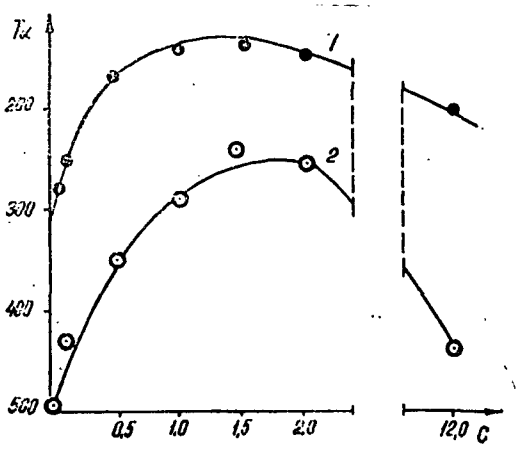

Figure 1. Effect of Concentration of Active Substance on Volumetric Activity of Activated Cupric Oxide
1--at W = 1000; 2--at W = 50,000 h^{-1}

concentrations of the latter to 1.0 mole/l, with an increase in the concentration of active substance in the granules, as should be expected, the effectiveness of the catalysts increases. Then, after a certain constant quantity has been reached, with an increase in the content of active substance, the effectiveness of the catalyst drops. With volumetric velocity of 1000 h^{-1}, the effectiveness of the catalyst drops to a lower degree with an increase in the content of active substance, than with volumetric velocity of 50,000 h^{-1}. This allows us to hypothesize that the decline in effectiveness is to a certain measure the result of the effect of heat and mass transfer.

Two catalysts were selected for further tests, one from those specimens where the effectiveness of the catalyst increases with a rise in the concentration of active substance, and the other for specimens in which it drops.

Figure 2 presents the results of oxidation of carbon monoxide on a mixed catalyst which contains active substance in a quantity of 0.5 mole/l, and on a catalyst made of "pure" active substance. The /4 results are shown on the curve in the form of the dependence of temperature in which 85% of the CO is oxidized, on the volumetric velocity.

As is apparent from the figure, the diluted catalysts in which the quantities of the active surface are much lower than in the pure

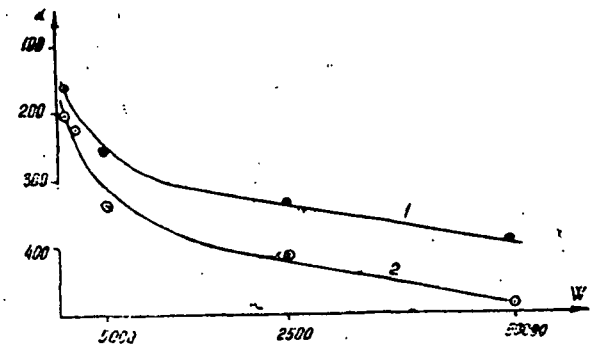

Figure 2. Dependence of Effectiveness of Catalyst on Volumetric Velocity:

1--with concentration of 0.5 mole/l;
2--with pure catalyst.

catalysts, has higher effectiveness. Higher activity in the diluted catalysts may be the result of the effect of the solid diluent, or heat and mass transfer.

In the interval of high volumetric velocities (5,000 - 50,000 h^{-1}) when the effect of heat and mass transfer must be more significant, the difference in the effectiveness of the catalyst is more sharply pronounced. With volumetric velocities of 500 - 1000 h^{-1}, the catalysts are almost equally active.

Study of the catalysts at lower volumetric velocities (100 - 1000 h^{-1}) makes it possible to explain the findings. Figure 3 presents the dependence of $\lg K_{sp}$ on the contact time. Our experiments did not obtain a region of volumetric velocities in whose limits the activity does not depend on the contact time, and it is impossible to be confident that with contact time equal to 36 s ($W = 100\ h^{-1}$), the process occurs in the kinetic region. At the same time, a slight difference in the quantities of the specific reaction velocity constant obtained with contact time of 36 s makes it possible to assume that in this case the process occurs under conditions close to the kinetic region.

As the contact time diminishes, the value of the specific activity /5 of the diluted catalyst increases and with contact time of 4 s reaches a quantity which is almost 10 times greater than the activity of the catalyst with contact time of 36 s. The observed effect qualitatively

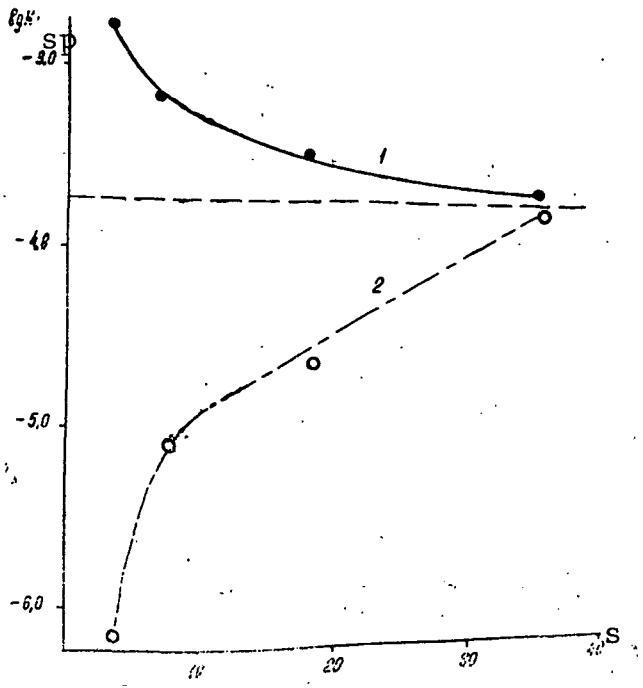

Figure 3. Dependence of lgK_{sp} on the Contact Time

1--with concentration of 0.5 mole/l;
2--with pure catalyst.

coincides with the warming up of the catalyst as a result of difficulties in heat transfer. The reasons for the increase in effectiveness of the diluted catalysts in a diffuse region have been examined previously [1, 3, 4].

Reduction in the contact time results in a decrease in the experimental quantity of specific activity of the "pure" catalyst by more than 2 orders, i.e., with contact time equal to 4 s. No more than 1% of the active surface of the catalyst participates in the reaction. The influence of warming of the active substance particles in homogeneous catalysts does not play a significant role [2]. Thus, decrease in the contact time from 36 to 4 s results in a discrepancy of more than 3 orders between the values of specific activity obtained for a pure active substance and the same active substance diluted by the carrier. It is evident that the different values of specific activity are the result of the influence of heat and mass transfer on the

reaction rate since the influence of the carrier or other factors on the activity of the catalyst in the kinetic region cannot be a function of contact time.

Conclusions

1. During oxidation of CO on activated cupric oxide, the kinetic region of reaction occurrence is implemented with contacts of over 36 s ($W < 100\ h^{-1}$). With higher volumetric velocities, the influence of diffusion may be fairly significant to distort the test data which are obtained experimentally.

2. The effect of heat and mass transfer in the case of granules /6 made of active mass is expressed in a reduction, and in the case of diluted catalysts, an increase in the observed specific ativity.

3. For practical catalysts used in the diffuse region, dilution of the active substance by an inert diluent can be a method of increasing the effectiveness of the catalyst.

Bibliography

1. Sultanov, M. Yu.; Khain, G. I. Uch. zap. AzINEFTEKHIM, series IX, No. 4, 1977, p. 71.

2. Mingle, J. O.; and Smits, J. M. A. J. Ch. E. J., 7, 243, 1961.

3. Sultanov, M. Yu.; and Khain, G. I. Uch. zap. AzINEFTEKHIM, Series IX, No. 4, 1973, p. 77.

4. Hlavacek, V.; and Kubicek, M. Chem. Eng. Sci., 15, 1970, 1537.