

# Nano-scale Sensor Networks for Chemical Catalysis

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**Abstract**—Following the success of conventional macro-scale wireless sensor networks, researchers are now investigating the viability of nano-scale sensor networks (NSNs), which are formed by establishing communication between devices made from nanomaterials. Due to unique properties of nanomaterials, it is envisaged that such NSNs can support completely new types of applications beyond what could be achieved with conventional sensor networks.

In this paper, we propose and investigate a novel application of NSNs in the field of chemical catalysis. More specifically, our goal is to explore the use of NSN to improve product selectivity of Fischer-Tropsch (FT) reaction, a major class of chemical reactions for converting natural gas to liquid fuel. Given that nano-scale wireless communication is at very early stages of development, we investigate FT selectivity as a function of communication reliability of the underlying NSN using molecule-level simulations.

Our study reveals that packet loss probability ( $p$ ) of the NSN is an important factor for improving FT selectivity, but the rate of improvement in selectivity is not uniform in the entire range of  $p$ . We can quickly increase selectivity up to a certain point by decreasing  $p$ , but further improvement in selectivity requires an exponential reduction in  $p$ . This finding suggests that although chemical catalysis may benefit from early adoption of NSNs, communication reliability will be a major factor in taking the full-advantage of such nano-networks.

## I. INTRODUCTION

A recent insightful article by Akyildiz and Jornet [1] has shown how the phenomenal progress in nanotechnology has brought us closer to realizing nano-scale sensor networks (NSNs). Technically, a NSN is a network of nano-scale devices capable of some basic computing, sensing, actuation, and communication tasks. Although nano devices are not yet available commercially, there are significant relevant developments in recent years that point to a future when such devices could be produced in bulk. For example, a miniature *hydrogen sensor* was reported in [2]. Yonzon et al. [3] surveys many other types of nanosensors that can be used for chemical and biological sensing. Similarly, progress has been recorded in chemical and biological nanoactuators that can be used to accomplish some basic tasks at molecular level [1], [4].

Finally, work has begun exploring communication possibilities at nano-scale, which would ultimately connect these nanosensors to form a NSN. The seminal paper by Akyildiz et al. [5] revealed that conceptually it is possible to achieve communication at nano-scale either using electromagnetic or

some form of molecular-based transceivers. This has sparked a flurry of new research activities in the recent years to understand the unique properties of nanomaterials that could be utilised for communication between nano devices [1], [6], [7] [8].

Because a NSN can work at atomic levels, they can be used for totally new kind of nanotechnology applications which cannot be realized with conventional sensor networks. Some of current interesting proposed applications for NSNs are in biomedical (health monitoring and drug delivery), environmental (plant monitoring and defeating insect plague), industrial (ultra-sensitive touch interfaces), and military (biological and chemical defense) domains [1]. In all these applications, distributed communication between nanosensors are envisaged to accomplish the application goal.

In this paper, we propose and investigate a novel application of NSNs in the field of chemical catalysis. More specifically, our goal is to explore the potential of NSN in improving product selectivity of chemical catalysis and as an example Fischer-Tropsch (FT) reaction [9], a major process for converting natural gas to liquid hydrocarbons, is examined. While FT reaction produces highly useful hydrocarbons in the form of liquid fuels including methanol, ethanol, diesel and gasoline, it also produces low-grade hydrocarbons such as methane. FT selectivity refers to the ratio of highly useful hydrocarbons to the total product output.

Improving selectivity of FT reaction is an active area of research worldwide due to its high economic and environmental significance. Traditional research focused on improving selectivity by controlling the global reactor variables, such as temperature and pressure or by developing more efficient catalysts [10]. The recent trend in improving catalysis performance is to apply nanotechnology in producing new nanostructured materials and harnessing the new unique properties of these nanoparticles for more efficient catalysis [11], [12]. However, these works mainly focus on building novel nano materials for catalysis, but do not consider communication between nano materials as a source of further potential improvement in selectivity. Therefore, the proposed application of NSN is novel and can be considered complementary to the ongoing research in *nano catalysis*.

The remaining of this paper is structured as follows. Section II reviews the basics of FT catalysis. The proposed application of NSN to FT selectivity control is presented in Section III. In Section IV, we model the effect of NSN communication reliability on FT reactions, followed by chemical simulations in Section V. Results are discussed in section VI. We conclude the paper in Section VII.

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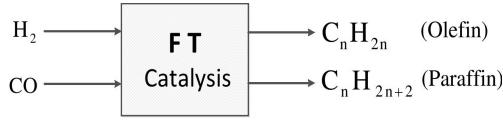


Fig. 1. Input and output of Fischer-Tropsch Catalysis.

## II. FISCHER-TROPSCH CATALYSIS

### A. Inputs, Outputs, and Selectivity

FT is a complex process involving many intermediate chemical reactions or steps, but we can depict the overall function using the ‘black-box’ of Figure 1. There are two inputs,  $H_2$  (Hydrogen) and  $CO$  (carbon monoxide; note that  $H_2$  and  $CO$  are obtained from gasification of natural gas.), which are fed to a FT reactor with a predetermined ratio. After several intermediate reactions, we obtain two main types of output products, whose chemical formulae are  $C_nH_{2n}$  (olefin) and  $C_nH_{2n+2}$  (paraffin).

Olefins are unsaturated organic compounds which are used as ‘building block’ chemicals for making other petrochemicals and polymers used in a wide variety of industrial and consumer markets [13], [14]. In contrast, paraffins are saturated hydrocarbons that cannot be used for further product development and therefore have low commercial value. Naturally, the performance of FT refers to the olefin-to-paraffin ratio or the fraction of total hydrocarbon output that is produced as olefin, which is referred to as the *selectivity* of FT.

There are many factors that affect the selectivity of FT. It is now well known that the process conditions, such as temperature and pressure, as well as catalyst composition influence the selectivity [15], [16]. How to improve FT selectivity by controlling these factors is a hot topic of research in chemical catalysis [10], [11]. It is important to note that the proposed NSN aims to improve FT selectivity in a completely different way. A NSN deployed on the catalyst surface would be able to monitor all the reactions and intermediate steps. As such, it would enable us to intervene in the middle of the FT process and divert the product path from paraffin to olefin in a more direct and efficient way.

### B. Intermediate Steps and Elementary Reactions

It is important to understand intermediate steps of a FT which is known as elementary reactions if we are to propose control at molecule-level. Figure 2 shows how elementary reactions convert  $H_2$  and  $CO$  to olefins and paraffins [16]. All reactions take place on the surface of a catalyst. Before a reaction can take place, reactants must first be adsorbed onto (or adhered to) the catalytic surface. The surface of a catalyst contains numerous sites or special locations (denoted by  $s$  in Figure 2) where reactants can be adsorbed. A  $CO$  molecule breaks into one  $C$  and one  $O$  molecules each adsorbing onto a separate site. Similarly, a  $H_2$  breaks into two  $H$ . These  $H$  atoms play key roles in all subsequent product development.

A  $CH_2$  is formed when a  $C$  reacts with two nearby  $H$  atoms and a  $CH_2$  in turn may react with a nearby  $H$  to form

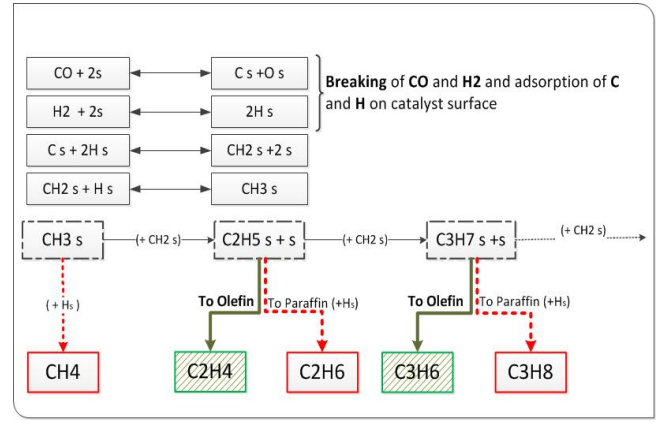


Fig. 2. Fischer-Tropsch elementary reactions, chain growths and path to olefins and paraffins.

a  $CH_3$ . A  $CH_3$  can either react with a  $H$  to form a  $CH_4$  (methane) or it can kick start a *chain growth* by reacting with a  $CH_2$ , forming intermediate species  $C_nH_{2n+1}$ , which are the precursors to both olefins and paraffins. A  $C_nH_{2n+1}$  can either lead to olefin, i.e.,  $C_nH_{2n}$ , through desorption or dehydrogenation (losing one  $H$ ), or paraffin, i.e.,  $C_nH_{2n+2}$ , through hydrogenation (adding one  $H$ ). In order to improve the FT selectivity, our NSN aims to reduce the occurrence of the latter reaction which produces paraffins.

### C. Reaction Rates and Kinetic Constants

The rate  $r$  at which two reactants,  $A$  and  $B$ , react to produce the output species is given by the law of mass action [17]

$$r = k \times \theta_A \times \theta_B \quad (1)$$

where  $k$  is a kinetic constant of this specific reaction and  $\theta_A$  and  $\theta_B$  are volumes or counts of species on the catalyst surface (species concentration). The value of  $k$  is difficult to predict, but it may be affected by the process and catalyst factors. In addition,  $k$  can be given a probabilistic interpretation in the sense at the probability that a reaction will take place is proportional to  $k$ . Equation (1) suggests that for a given concentration level, we could control rates of specific elementary reactions if we found a way to influence the kinetic constants of those reactions. The proposed NSN aims to achieve such control by monitoring catalysis at molecular level.

## III. PROPOSED NANO SENSOR NETWORK

The main goal of the proposed NSN is to cutoff the path to the production of paraffin and redirect surface effort to increase olefin productivity. From Figure 2 we see that  $H$  plays a key role to the product selectivity. Paraffin is produced only when a  $H$  reacts with  $C_nH_{2n+1}$ , which is the elementary reaction called hydrogenation to paraffin (HTP). Therefore, the aim of NSN would be to prevent  $H$  from reacting with  $C_nH_{2n+1}$  as much as possible.

Assuming that a  $H$  and  $C_nH_{2n+1}$  would react only if these two species come *close* to each other, say reside

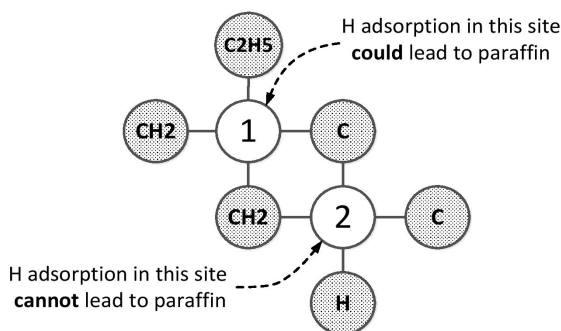


Fig. 3. An example snapshot from the surface of a catalyst. There are two unoccupied sites, each having four neighbours occupied with different molecule species. Hydrogen adsorption in site 1 can lead to a paraffin because of the presence of an  $C_2H_5$  in the neighbourhood, but its adsorption in site 2 cannot produce any paraffin.

in neighbouring sites of a catalyst surface, then one way to reduce reaction rates of HTP would be to control the location of H adsorption on the catalyst surface based on the knowledge of current content of each site. For example, Figure 3 shows the contents of a number of sites (represented by circles) on the catalyst surface. There are two unoccupied sites, marked as 1 and 2. If an H is adsorbed at site 1, then it could lead to paraffin due to presence of  $C_nH_{2n+1}$  ( $C_2H_5$ ), but an adsorption of an H at site 2 cannot lead to the production of paraffin. Therefore, the NSN should allow H adsorption for site 2, but prevent it for site 1.

Such spatial control of H adsorption on a catalyst surface could be achieved with an NSN in the following ways. Let us assume that we can deploy a nano-sensor at each site as part of catalyst preparation<sup>1</sup>. Each nano-sensor is capable of sensing two events: (1) a  $C_nH_{2n+1}$  has just formed at the site from an elementary reaction and (2) an H is attempting to adsorb to the currently unoccupied site. In detection of the first event, the nanosensor should update a local binary variable, which indicates whether the site currently contains a  $C_nH_{2n+1}$ . In detection of the second event, the nanosensor queries all the neighbouring nanosensors to learn whether there exists a  $C_nH_{2n+1}$  in the neighbourhood (neighbourhood search). If  $C_nH_{2n+1}$  is present in the neighbourhood, the nanosensor should prevent the H adsorption (say by reflecting it back to the feed gas which would be a basic actuation task), but allow the adsorption otherwise.

It is clear that success of the proposed NSN in exerting spatial control over H adsorption would depend in turn on its sensing, actuation, and communication capabilities. While NSNs are in their embryonic stages of development, there would be issues with all three dimensions, sensing, actuation, and communication. In this paper, our focus is on the communication performance of NSNs and how communication reliability affects FT selectivity assuming perfect sensing and actuation capabilities.

#### IV. EFFECTS OF SENSOR COMMUNICATION RELIABILITY

Theoretically, if H atom are never allowed to adsorb near  $C_nH_{2n+1}$ , then HTP reactions cannot take place. In other words, if the NSN is 100% successful in its mission, we can expect a zero reaction rate (or zero production) of paraffins irrespective of the level of H and  $C_nH_{2n+1}$  concentrations on the catalyst surface. Thus, based on Equation (1) the net effect of a perfect NSN, which never allows an H to come close to  $C_nH_{2n+1}$ , is to yield  $k = 0$ .

In practice, the performance of NSN, i.e., its ability to successfully control the adsorption location of H atoms, will depend on its sensing, actuation, and communication reliability. In this paper, we are interested in modelling the effect of communication reliability on FT catalysis performance, hence we will assume perfect sensing and actuation performance for the NSN. We will, however, assume that communication between two neighbouring nanosensors is not 100% reliable. More precisely, we will assume that a packet (a unit of communication) will be lost with probability  $p$  and will be successfully received with  $(1 - p)$ . Our goal is then to derive FT selectivity as a function of  $p$ . Note that the packet loss probability  $p$  is used to model loss due to bit error, packet collision and late response. For simplicity, we assume that the value of  $p$  does not vary across the catalyst surface; more realistic models will be used in future work.

As discussed earlier, the effect of NSN can be modelled by deriving a modified  $k$ , denoted by  $k'$ , for the HTP reactions. For extreme values of  $p$ , the corresponding  $k$  can be readily obtained. We have  $k' = 0$  for  $p = 0$ , i.e., when the NSN works perfectly. Similarly,  $k' = k$  for  $p = 1$ , i.e., when the NSN is totally ineffective. To model  $k'$  for the more general case of  $0 < p < 1$ , we have to examine the communication protocol used by the nanosensors to search the neighbourhood in more details.

##### A. Neighbourhood Searching Protocol

When a nanosensor residing at an unoccupied site detects the event that an H is attempting to adsorb to the site, it must immediately query its neighbourhood to establish whether or not the neighbourhood contains a  $C_nH_{2n+1}$ . The responses of this query will help the nanosensor to make the right decision about whether to allow the H to adsorb to this site or not. However, The nanosensor may fail to learn the presence of  $C_nH_{2n+1}$  in the neighbourhood if communication is not reliable, leading to less than perfect outcome for the NSN.

We call this query and response protocol the neighbourhood searching protocol, whose steps are as follows. A nanosensor broadcasts a query message to all its neighbours asking whether they contain  $C_nH_{2n+1}$  and waits for a predetermined period to hear from them. Upon receiving a query message, each neighbour responds immediately if it contains a  $C_nH_{2n+1}$ . If the originator does not receive any positive response from any of its neighbours within the given period, it concludes that the neighbourhood does not contain  $C_nH_{2n+1}$  and allows the H to adsorb to its site (deflect it otherwise). Successful two-way communication with a neighbour therefore would require that the broadcast

<sup>1</sup>Catalysts are routinely prepared in nano-scale[11], [12]

message was successfully received by the neighbour, and the response from the neighbour was also successfully received by the originator.

### B. Derivation of Modified Kinetic Constant

We consider a neighbourhood consisting of  $N$  neighbours. Let us use the following notations and probabilities:

- $f$ : probability of any of the  $N$  neighbours containing a  $C_nH_{2n+1}$
- $P(N, i) = \binom{N}{i} f^i (1-f)^{N-i}$ : probability of  $i$  neighbours containing  $C_nH_{2n+1}$
- $\sum_i^N P(N, i)$ : probability that the neighbourhood contains  $C_nH_{2n+1}$
- $q = 1 - (1-p)^2$ : Probability of failing to communicate with a given neighbour
- $(1-q)^i$ : Probability of successful communication with at least one of the  $i$  neighbours
- $s$ : probability of successfully learning the existence of  $C_nH_{2n+1}$  in the neighbourhood (probability of successfully deflecting a H) given that the neighbourhood contains  $C_nH_{2n+1}$

We obtain  $s$  as :

$$s = \frac{\sum_i^N P(N, i)(1-q^i)}{\sum_i^N P(N, i)} \quad (2)$$

Modified reaction constant  $k$  is then derived as:

$$k' = k(1-s) : 0 < f \leq 1, 0 \leq q \leq 1 \quad (3)$$

At any given time  $t$ ,  $f$  could be obtained from the molecule concentrations in the reactor as:

$$f(t) = \frac{N_{C_nH_{2n+1}}(t)}{N_{\text{total}}(t)} \quad (4)$$

Where  $N_{C_nH_{2n+1}}(t)$  and  $N_{\text{total}}(t)$  are, respectively, the number of  $C_nH_{2n+1}$  molecules and total number of molecules at time  $t$  in the reactor. For  $f = 0$  (if there is no  $C_nH_{2n+1}$  in the system),  $k' = k$ .

Equation (3) allows us to derive the modified kinetic constant due to spatial control of H adsorption on the catalyst surface as a function of packet loss probability  $p$  for any communication between the nanosensors. To model the effect of NSN on FT selectivity, the next step would be to model selectivity as a function of the kinetic constants of all reactions, which is a challenging problem due to the complicated nature of chemical reactions. Instead, we use simulation.

### V. SIMULATION

Chemical reactions can be modelled by Markov chains and a standard algorithm to simulate them is the Stochastic Simulation Algorithm (SSA) by Gillespie [18]. The standard SSA algorithm takes three inputs: (1) a set of reactions, (2) a set of kinetic constants corresponding to each reaction, and (3) initial counts for each species, which define the *initial state* of the simulation.

In general, the state in SSA is defined as the number of each chemical species in the system. At each state,

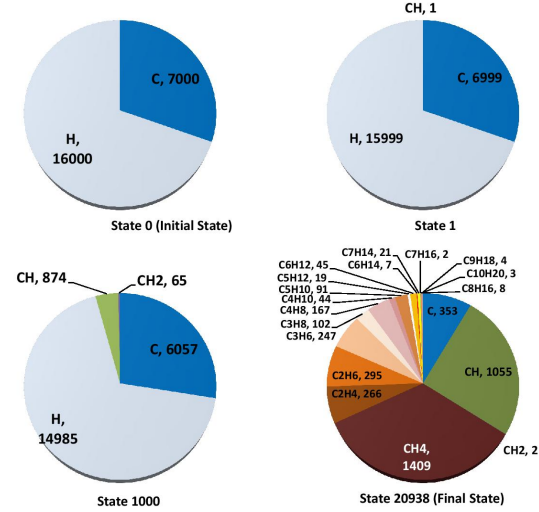


Fig. 4. A snapshot of state changes during simulation. Species concentrations continue to evolve as elementary reactions take place.

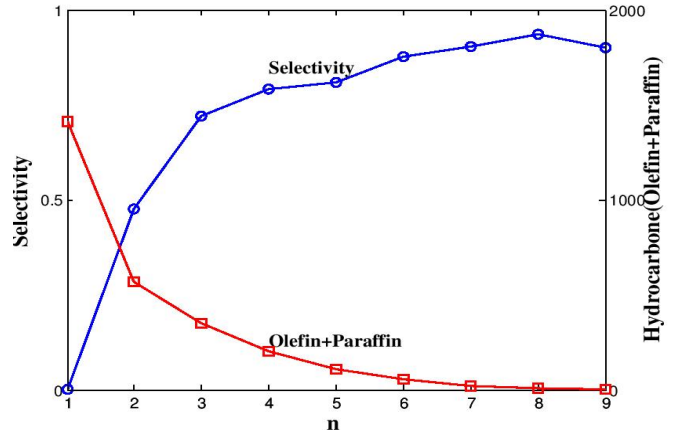


Fig. 5. Selectivity and number of species molecules (Olefin+Paraffin) for different chain length ( $n$ ).

the simulator determines a candidate reaction to take place from the set of all possible reactions and updates the state (species counts) accordingly. Simulation stops when there are no reactions to take place. The output of the algorithm is the number of each chemical species at the end of the simulation. Note that the standard SSA algorithm assumes that the kinetic constant does not change over time, but we have modified the algorithm so that the kinetic constants for the HTP reaction depends on packet loss probability  $p$  and also probability of any of the  $N$  neighbours containing a  $C_nH_{2n+1}$ ,  $f$ , according to Equation (3).

We consider a FT catalysis with  $n \leq 10$  meaning that the chain growth is limited to  $C_{10}H_{21}$ , i.e., the longest olefin and paraffin produced would be, respectively,  $C_{10}H_{20}$  and  $C_{10}H_{22}$ . Thus, based on Figure 2 we have a total of 32

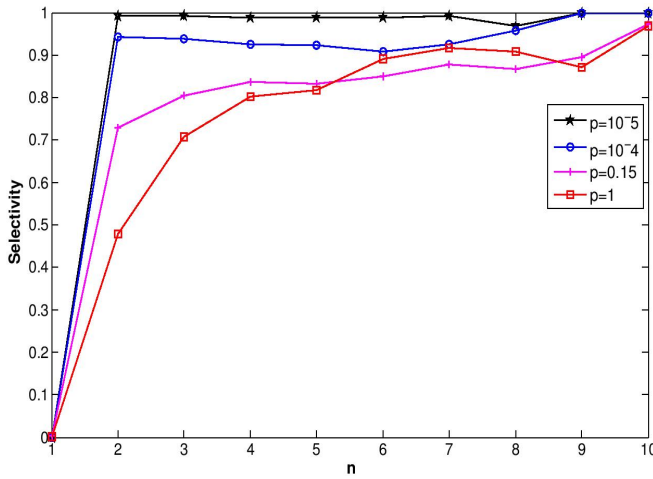


Fig. 6. Selectivity improvement using NSN as a function of different chain length ( $n$ ).

reactions (3 chain initiation, 9 chain growth, 10 hydrogenation to paraffin and 10 dehydrogenation to olefin). Similarly, we have a total of 33 species including C, H, CH, 10 as ( $C_nH_{2n+1}$ ), 10 as ( $C_nH_{2n}$ ), and 10 as ( $C_nH_{2n+2}$ ). We set the initial species counts as  $C=7000$ ,  $H=16000$ , and zero for all other species. Figure 4 shows how the states change for a given simulation ( $p = 1$ ) after forming different species via different reactions. At the end of the simulation, we analyse all olefin and paraffin products and the resulting selectivity for the simulated FT catalysis.

Although there are a total of 32 kinetic constants for 32 reactions, for simplification, it can be assumed that all reactions in a given category have the same kinetic constant. We simulated a total of 4 different sets of kinetic constants. We only present results for the set where all  $k = 7$ , as the results for other settings were similar. To simulate the effect of a given packet loss probability  $p$ , we run a new simulation with a modified  $k_{ip}$  (Kinetic constant of HTP group of reactions) according to Equation (3), while all other kinetic constants remain the same. For each of the 4 sets of kinetic constants, we simulated a total of 200 different packet loss probabilities ranging from  $p = 0$  to  $p = 1$ , leading to a total of  $4 \times 200 = 800$  simulation configurations. For each configuration, the simulation is repeated 10 times and the average results over 10 runs are presented.

Assuming sites are structured as a regular grid [19] on the surface of a catalyst, the value of  $N$  (neighbourhood size) could vary from 4, 8, 16, etc., depending on whether H could react with a  $C_nH_{2n+1}$  located in the immediate (left-right-up-down) grid positions or beyond. We present results for  $N = 4$ , but we found that the conclusions drawn in this paper are not affected for larger values of  $N$ .

## VI. RESULTS

First, we examine selectivity for each individual hydrocarbon chain length  $n$ . For  $p = 1$ , Figure 5 shows the individual selectivity alongside the total hydrocarbon product volume

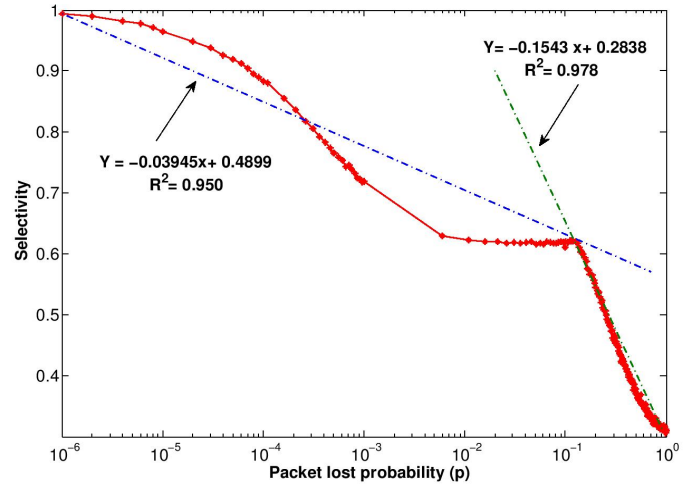


Fig. 7. Overall selectivity as a function of packet lost probability  $p$ .

(olefin+paraffin) for each  $n$ . We see that selectivity and volume have opposite trends, which is a natural phenomenon in FT catalysis [15]. Therefore, if NSN is to have a major impact on the *overall* selectivity, it must boost selectivity for small values of  $n$ , where FT selectivity is naturally low, yet the product volume is high. Figure 6 plots individual selectivity for different values of  $p$ . Indeed we find that the proposed NSN can successfully boost selectivity for most values of  $n$ , especially when  $n$  is small. The smaller the packet loss probability, the higher the impact on selectivity.

In Figure 7, we investigate improvement in overall selectivity as a function of  $p$ . We find that the selectivity has a logarithmic dependence on  $p$ , which means that we need exponential reduction in packet loss probability to achieve a linear increase in FT selectivity. We also observe that the rate of improvement in selectivity is not uniform over the entire range of  $0 \leq p \leq 1$ . We can divide the entire range roughly in two segments,  $10^{-1} \leq p \leq 1$  and  $0 \leq p \leq 10^{-1}$ . The rate of selectivity improvement is much faster in the former than in the latter. It means that we can quickly improve selectivity up to certain point by simply achieving a packet loss rate of about  $10^{-1}$ , but improvement beyond that becomes much harder, which would require more significant improvement in  $p$ . For example, selectivity could be increased from 0.3 to 0.6 by ensuring that packet loss probability does not exceed beyond 10%, but to increase selectivity from 0.6 to 0.9, we would need to keep packet loss below 0.01%. These results therefore suggest that although the concept of NSN holds great promise for improving FT selectivity, the performance requirement of such NSNs in terms of communication reliability is extremely high.

## VII. CONCLUSION

We have proposed a nano-scale sensor network to monitor and control chemical reactions at molecular-level with the ultimate goal of improving product selectivity of FT catalysis. We have derived expressions to model kinetic constants of key chemical reactions as a function of packet

loss probability in the underlying sensor network. Finally, the impact of packet loss probability on FT selectivity is investigated using molecule-level simulations of chemical processes.

Our simulations revealed that FT selectivity has a logarithmic dependence on packet loss probability, and that we need an exponential reduction in packet loss probability to achieve a linear increase in FT selectivity. This finding suggests that for NSNs to be highly effective in improving selectivity, nanosensors must be able to communicate with each other at all times with high reliability.

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