Response to reviewers:

Reviewer #1: This work describes a new modification of an infrared reflection absorption spectroscopy (IRRAS) system to enable transient kinetics measurements by adding a fast gas pulsing valve and synchronizing it with a fast data acquisition system. This allows obtaining IRRA spectra with a time resolution down to ~ 67 ms with a signal-to-noise ratio that would normally require orders of magnitude longer acquisition times. The utility of this modification was demonstrated by following the kinetics of adsorption and desorption of CO from a clean Pd(111) crystal at elevated temperatures, both in ultra-high vacuum and in the presence of background O2 in the system. This shows how kinetics of fast processes can be captured with this approach, which can in turn provide insights into the elementary steps of reaction mechanisms when coupled with the development of modelling methodologies. Additionally, the approach used here can be easily extended to other surface science  
spectroscopies, such as X-ray photoelectron spectroscopy.  
This work is a significant improvement in the time resolution achievable using surface sensitive spectroscopy, which is of broad interest in the catalysis and surface science communities. As such, I recommend accepting this article for publication in Catalysis Today with the following minor corrections:

1.      The text in Figure 4 (Axis labels) is too small  
Zubin address.  
  
2.      In the supporting information, there are several references to "Figure M" but there is no Figure M in the SI or in the manuscript  
To Zubin: Figure M should be Figure 2 from the paper: CO adsorption on Pd(111): the effects of temperature and pressure.   
  
3.      A more detailed description of the model fitting procedure should be described beyond just "Fitting this regression line amounts to applying ordinary least squares." If you linearized the data using equation 3 and then fit a straight line to each set of data (450, 475, 500), then please show the data with the linear fits in the SI. The model does not seem to fit the data well at 475 K in particular, and this suggested figure should provide some more insight.

Thank you for your comment. We have added the details for obtaining the optimal k value using ordinary least squares. This includes the cost function that we are optimizing and the exact solution for k we obtain.   
  
We also thank you for noticing the misfit. There was a small error in our code and we have refitted all the curves. Additionally, we tried to resize the points to make the figure clearer. As per your suggestion, we have also included figures of the linear fittings (as well as the corresponding curved fittings) separately for each temperature for better visual assessment.

To Zubin: These 2 figures should replace the one in the manuscript.

Chart, line chart

Description automatically generatedChart, line chart, scatter chart

Description automatically generated

The following figures should be added in the Supporting Material:

Chart, scatter chart

Description automatically generatedChart, line chart

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Chart, scatter chart

Description automatically generatedChart, scatter chart

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Chart, scatter chart

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4.      Activation energy is in units of kcal/mol (on p. 11, it's just kcal)

Thank you for your careful reading. We have now fixed the units.  
  
  
Reviewer #2: The manuscript entitled ¨Addition of Transient Kinetics Capabilities to an Infrared Reflection Absorption Spectroscopy System through Synchronized Gas Pulsing and Data Acquisition¨ by Darbari et al. is a short but nice communication demonstrating the capabilities for studying transient kinetics on reversible processes on surfaces by a simple addition/modification of an IRRA system. This preliminary results shown by the authors are very promising in the sense that they might want to improve the instrument and data aquisition system in some ways to make it available for study other kind of processes (the authors briefly mentioned at the end the possibility of studying reaction mechanisms like carbon monoxide oxidation) and to acquire meaningful data in shorter times (improving time resolution).  
In general terms the manuscript is well written, easily understood for a broad audience and the results presented support the Conclusions.  
The Highlights section clearly states the main aspects of the manuscript.  
There are a few typos and mistakes, that need to be corrected, please check. One of them:  
- Introduction: First time citing Ref.6 should be in superscript.

Zubin address.  
  
Also some of the Figures could be improved to make them easier to understand the results:  
- Figure 1. The instrument drawing could be enlarged so is easier to understand the section that was modified. And probably rearrange the entire Figure to easier reading.

Zubin address.

- Figure 3b: Absorbance numbers are too small, make spacing larger to fit bigger numbers. Wavenumber axis numbers are difficult to read int he way presented. Time axis numers are missing.

Zubin address.  
  
An Arrhenius plot with only 3 point to fit and determine an Activation Energy for the process is too risky, but is understandable the authors do not purse to obtain and exact result of Ea but only to demonstrate the usage of their method.

Thank you for acknowledging this point. To make it clearer for readers, we have added a sentence stating this, and that we will report thorough analysis with sufficient number of data in a future work.