Temperature oscillations of methane oxidation in a jet stirred reactor

Yu Song[a], Oliver Herbinet[a], Frederique Battin-Leclerc[a,*], Marco Lubrano Lavadera[b], Pino Sabia[b], Mara de Joannon[b], Tiziano Faravelli[c]

[a] Laboratoire Réactions et Génie de Procédés, CNRS, Université de Lorraine, 1 rue Grandville, BP 20451, 54001 Nancy Cedex, France
[b] Istituto di Ricerche sulla Combustione, C.N.R., Napoli, Italy
[c] CMIC Department, Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano, Italy

* ***Corresponding author:*** frederique.battin-leclerc@univ-lorraine.fr

Introduction

The global energy demand is growing rapidly, and biogas from household and agriculture wastes, sewage sluge, and animal manure will play an important role in future. Biogas is a promising energy source, which can take place of fossil fuels in power and heat generation. Moreover, it has attracted attention of being used as a vehicle fuel.

Methane is the main component of biogas. There exists dynamic behaviors which are hidden in traditional systems involving methane oxidation. Significant temperature oscillation phenomenology for a wide range of inlet temperatures and C/O ratios has reported in previous studies. It could have a negative effect on the experimental results accuracy, on the other hand, the presence of such oscillatory regimes could raise high frequency in combustion chamber, which is very harmful to gas turbine burners. However, due to the special composition of biogas, the oscillations of methane oxidation under CO_2-bath gas need to be studied.

Purpose

The main aim of this work is to investigate the oscillatory regimes for biogas oxidation with various methane mole fractions and temperatures under fuel-lean to fuel-rich conditions with different carrier gases.

Experimental approach

The measurements were performed in a jet-stirred reactor designed following the rules established by the Villermaux team. Its main advantage is that it can be easily modeled as a 0 dimensional ideal perfectly stirred reactor as the temperature and the composition in the reactor are homogenous. The experiments were carried out at a constant pressure of 1.06 bar, at a fixed residence time of 2 s, with temperatures ranging from 950 to 1200 K. The bath gas were helium and carbon dioxide in this work. In order to measure the temperature inside the JSR reactor, a Pt13%Rh thermocouple (0.2 mm bead size) which was marked on the silica-coated envelope to measure how further it was inserted inside the reactor from the outlet of the reactor.

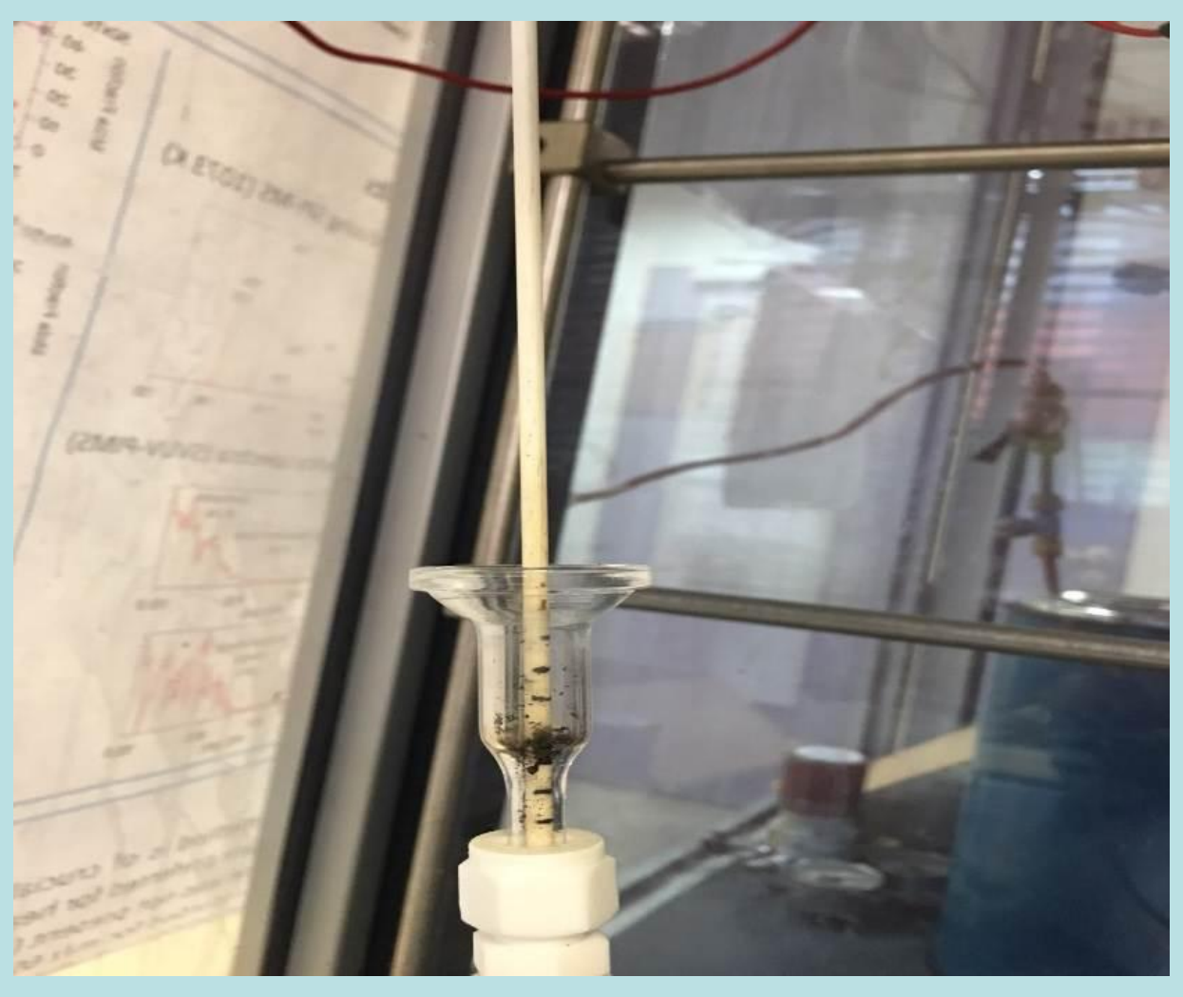


Fig. 1. The marked silica-coated envelope thermocouple

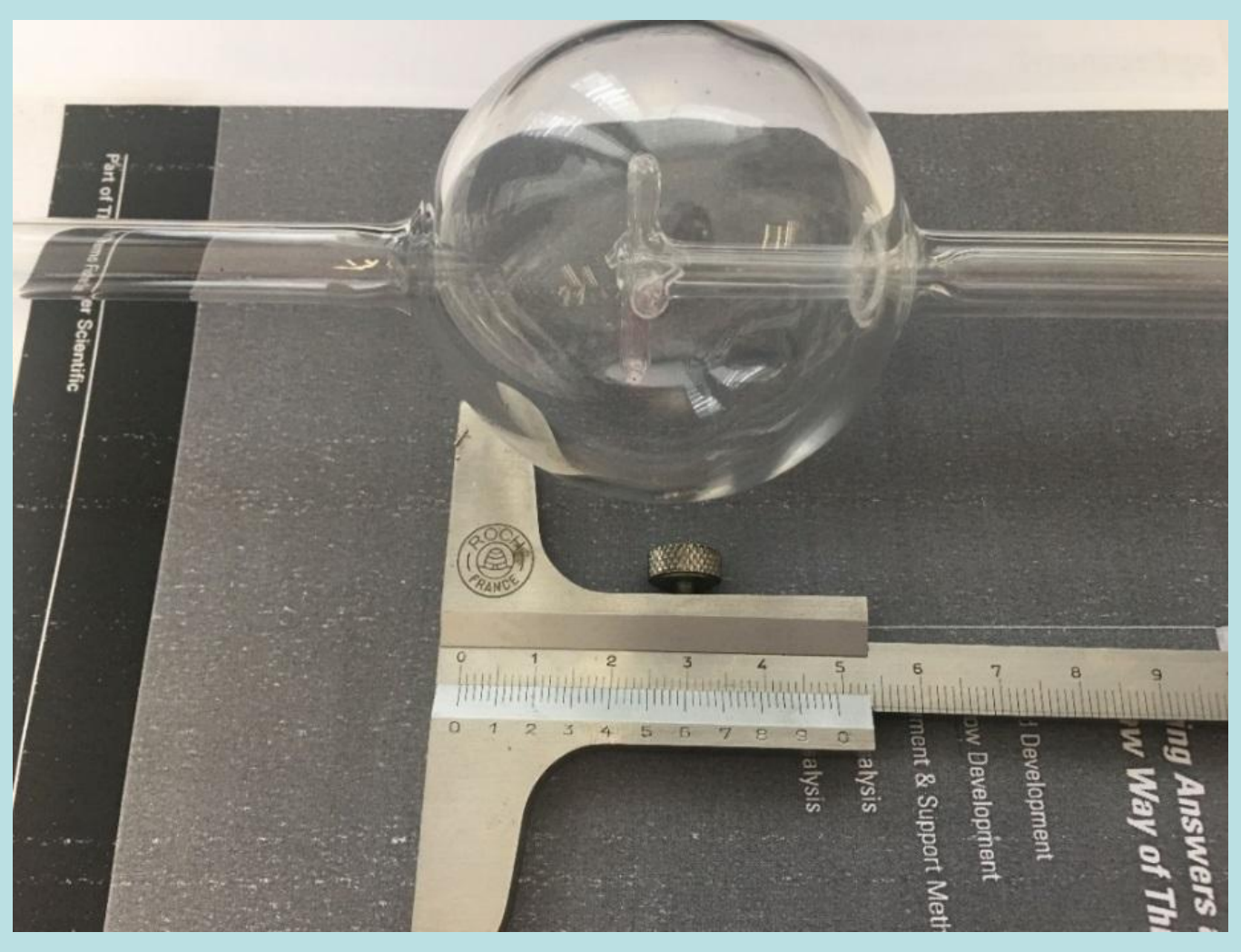
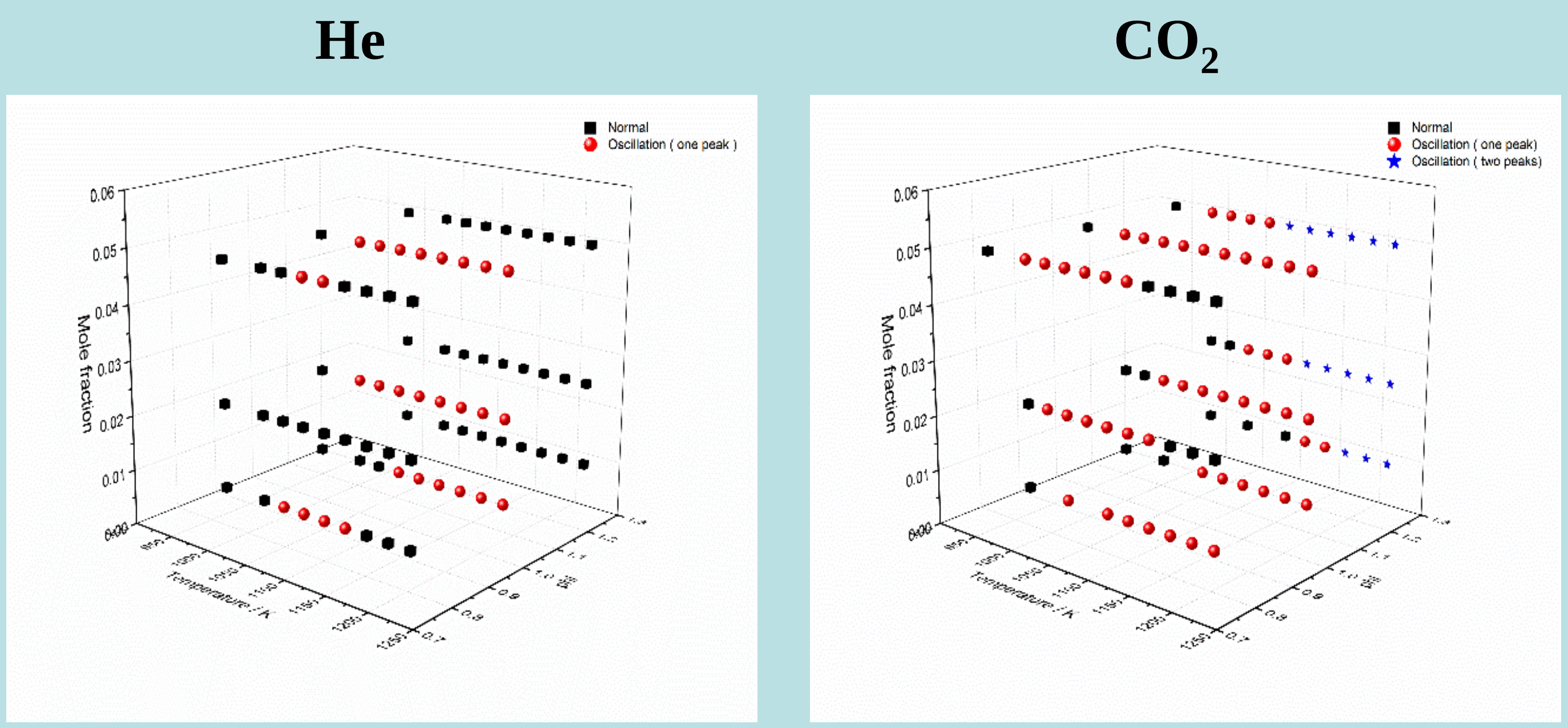


Fig. 2. The scale of jet stirred reactor

Results and discussion



He

CO_2

Fig. 3. The maps of experimental dynamic behaviors with fixed residence time 2 s for helium and carbon dioxide dilution

With carbon dioxide dilution, steady conditions appeared when the temperature was below 1000 K or beyond 1100 K under fuel-lean conditions. One peak oscillation behaviors occurred at stoichiometric ratio which was independent of dilution gas. Moreover, one peak oscillation behaviors were also detected under fuel-lean condition with carbon dioxide dilution. Furthermore, two peaks oscillation behaviors were only found under fuel-rich condition with carbon dioxide dilution.

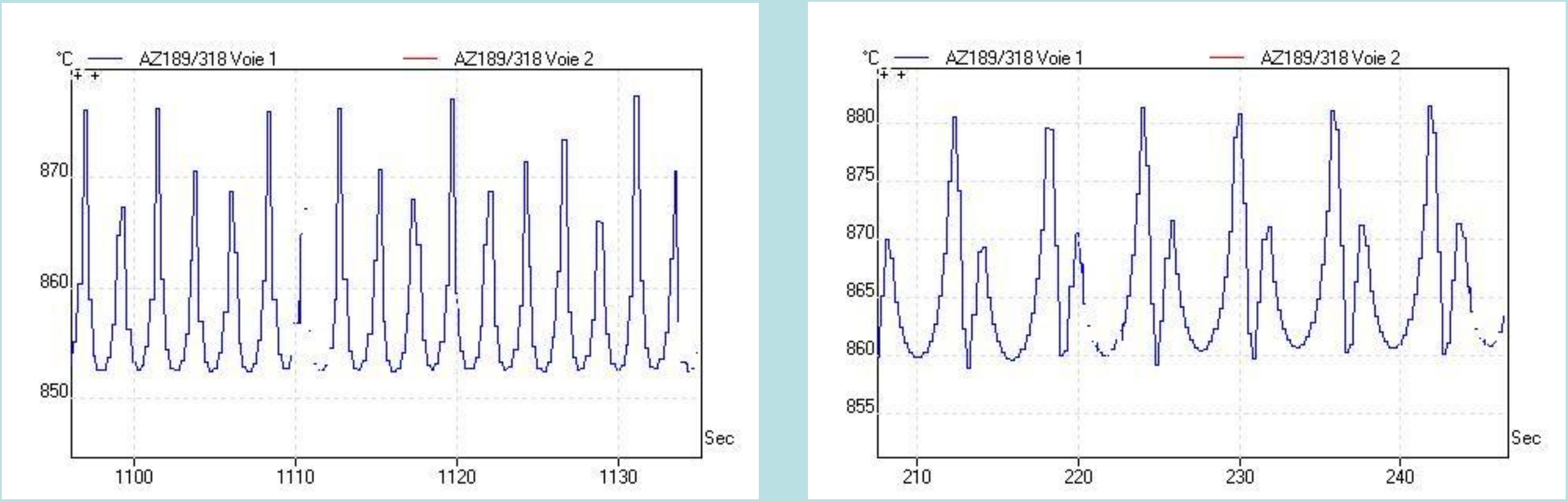


Fig. 4. The shapes of oscillations : one peak (Xch4=0.025; phi=1;T=1150 K; CO_2) and two peaks (Xch4=0.025; phi=1.25;T=1150 K; CO_2)

"Two peaks" experimental oscillation behavior occurred under fuel-rich conditions

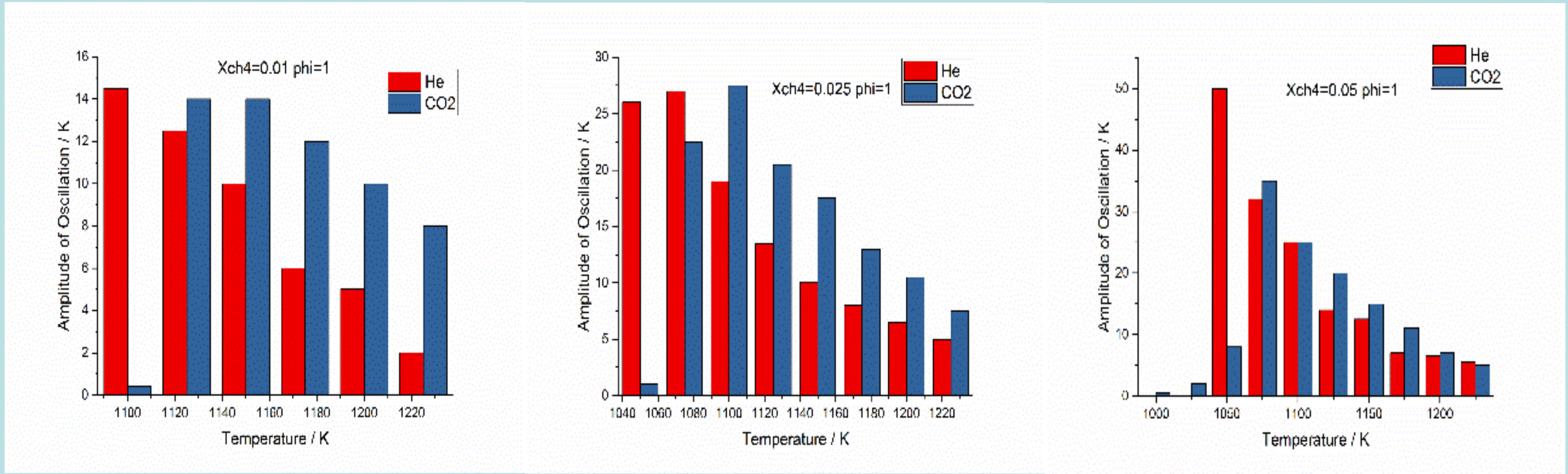


Fig. 5. The experimental oscillation amplitudes as a function of temperature with various methane concentration (0.01-0.05) under bath gas helium and carbon dioxide respectively.

The onset oscillation temperatures under bath gas helium conditions were 25 K lower than those under bath gas carbon dioxide conditions, which was independent of methane mole fraction. However, the oscillation amplitudes under bath gas carbon dioxide conditions were much higher than those under bath gas helium conditions. It demonstrated that the bath gas played a significant role on the dynamic behaviors.

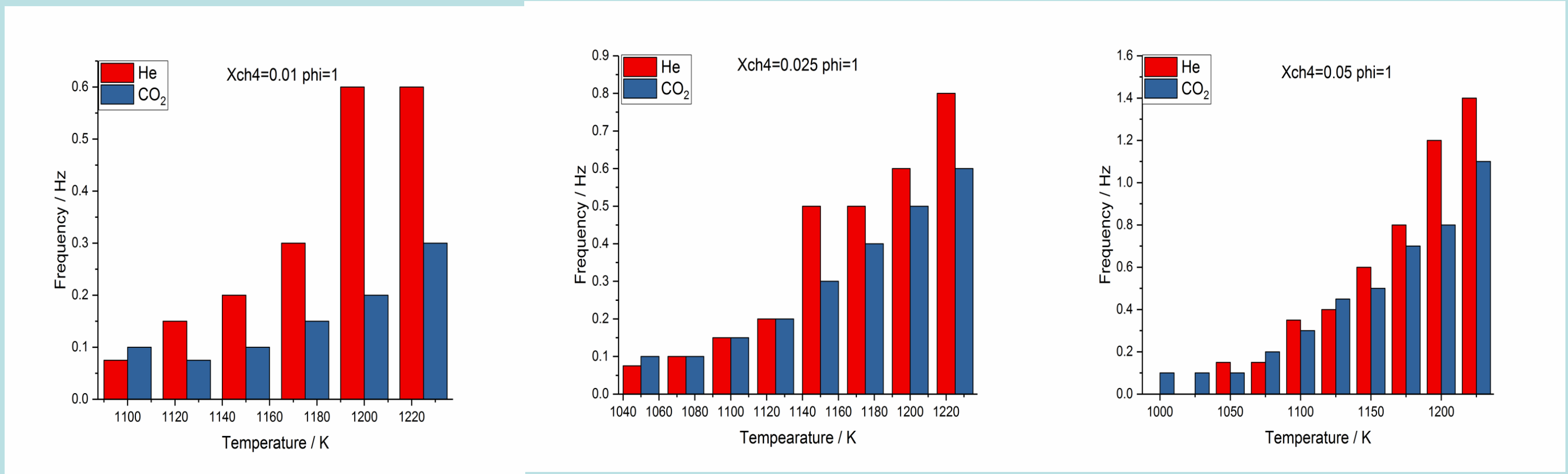


Fig. 6. The experimental oscillation frequency as a function of temperature with various methane concentration (0.01-0.05) under bath gas helium and carbon dioxide respectively.

The lower the oscillation amplitude, the higher the oscillation frequency.

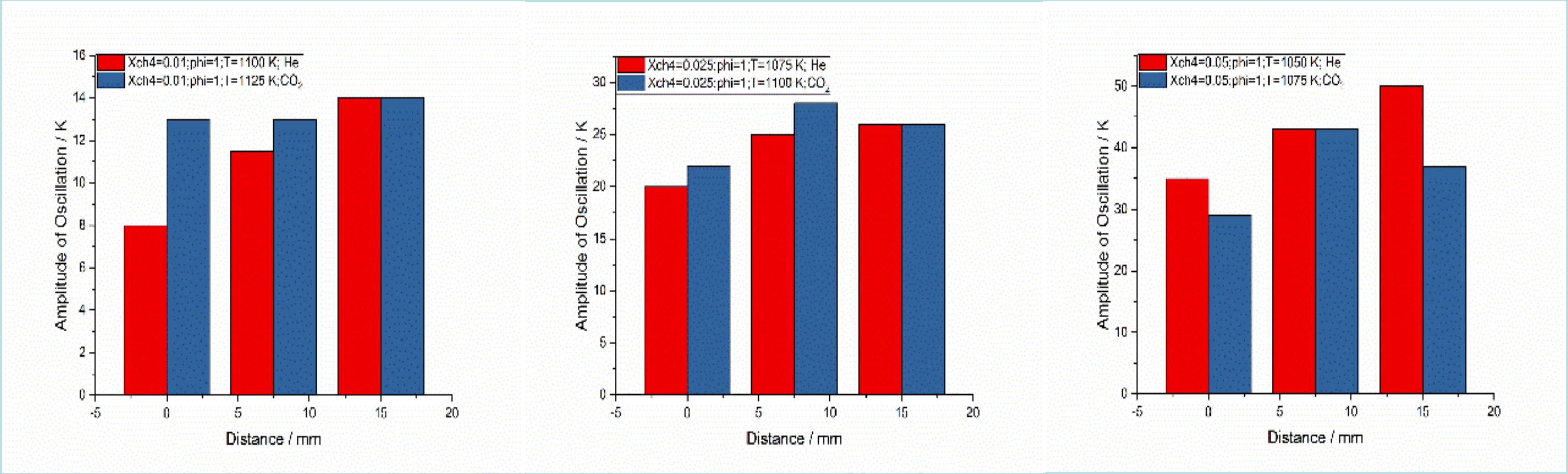


Fig. 7. The experimental oscillation amplitudes as a function of thermocouple position with various methane concentration (0.01-0.05) under bath gas helium and carbon dioxide respectively.

The oscillation amplitude increased monotonically in helium as a bath gas when the thermocouple moved from the JSR outlet to the core area.

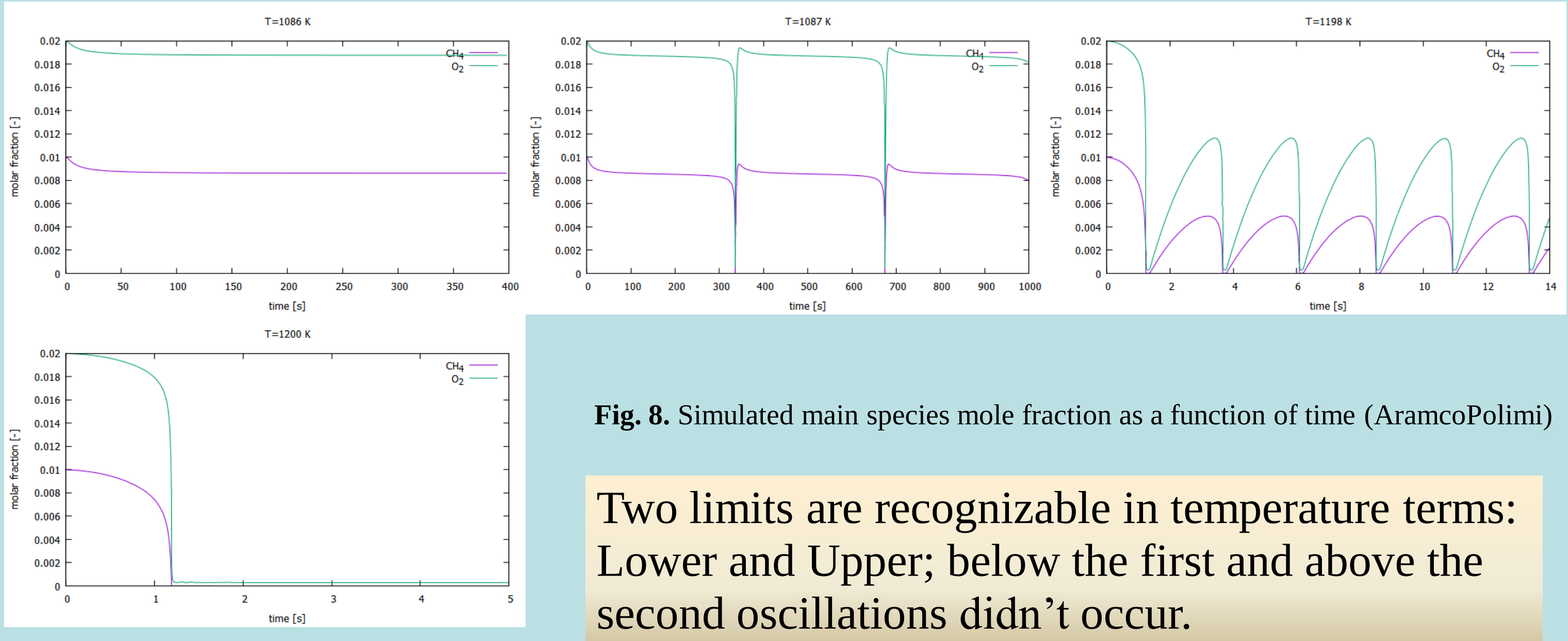


Fig. 8. Simulated main species mole fraction as a function of time (AramcoPolimi)

Two limits are recognizable in temperature terms: Lower and Upper; below the first and above the second oscillations didn't occur.

Conclusions

- The high concentration of CO_2 widened the oscillation range.
- Oscillation amplitudes under bath gas carbon dioxide conditions were much higher than those under bath gas helium conditions
- The lower the oscillation amplitude, the higher the oscillation frequency.

ACKNOWLEDGEMENTS

The work leading to this intervention has received funding from the European Union H2020 (H2020-SPIRE-04-2016) under grant agreement n°723706 and from the COST Action CM1404 "Chemistry of smart energy carriers and technologies".