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The combustion mitigation of methane as a non-CO_2 greenhouse gas

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ABSTRACT

Anthropogenic emissions of non-CO_2 greenhouse gases such as fugitive methane contribute significantly to global warming. A review of fugitive methane combustion mitigation and utilisation technologies, which are primarily aimed at methane emissions from coal mining activities, with a focus on modelling and simulation of ultra-lean methane oxidation/combustion is presented. The challenges associated with ultra-lean methane oxidation are on the ignition of the ultra-lean mixture and sustainability of the combustion process. There is a lack of fundamental studies on chemical kinetics of ultra-lean methane combustion and reliable kinetic schemes that can be used together with computational fluid dynamics studies to design and develop advanced mitigation systems. Mitigation of methane as a greenhouse gas calls for more efforts on understanding ultra-lean combustion. Recuperative combustion provides a promising means for mitigating ultra-lean methane emissions. Progress is needed on effective methods to ignite and to recuperate and retain heat for oxidation/combustion of the ultra-lean mixtures. Catalysts can be very effective in reducing the temperatures required for oxidation while plasmas may be utilised to assist the ignition, but thermodynamic/aerodynamic limits of burning ultra-lean methane remain unexplored. Further technological developments may be focussed on developing innovative capturing technology as well as technological innovations to achieve effective ignition and sustainable oxidation/combustion.

Contents

Abbreviations: CFC, chlorofluorocarbon; CFD, computational fluid dynamics; CFRR, catalytic flow reversal reactor; CMM, coal mine methane; CMR, catalytic monolith reactor; CO_2-eq, CO_2-equivalent; CSIRO, Commonwealth Scientific and Industrial Research Organization; DNS, direct numerical simulation; EDC, eddy dissipation concept; EEC, excess enthalpy combustion; EGR, exhaust gas recirculation; GHG, greenhouse gas; GT, gas turbine; GWP, global warming potential; HFC, hydrofluorocarbon; LEM, linear-eddy model; LES, large eddy simulation; LLGHG, long-lived greenhouse gas; MD, molecular dynamics; MILD, moderate or intense low-oxygen dilution; ODS, ozone-depleting substance; ODT, one-dimensional turbulence; PDF, probability density function; PFC, perfluorocarbon; PSA, pressure swing adsorption; RANS, Reynolds-averaged Navier–Stokes; SGS, sub-grid scale; TFRR, thermal flow reversal reactor; TST, transition state theory; UQ, uncertainty quantification; VAM, ventilation air methane.

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1. Background

Addressing climate change associated with anthropogenic emissions, either from human activities or from processes that have been affected by human activities, of greenhouse gases (GHGs) is a global challenge. Earth's climate is adversely affected as a result of the emissions of GHGs with carbon dioxide (CO_2) being the largest contributor, mainly from the utilisation of fossil fuels in combustion applications for energy conversion. Anthropogenic emissions of non-CO_2 greenhouse gases [1], such as methane (CH_4), nitrous oxide (N_2O), ozone-depleting substances (ODSs), hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), sulphur hexafluoride (SF_6) and nitrogen trifluoride (NF_3), also contribute significantly to warming. In terms of the abilities to absorb available infrared radiation and their persistence in the atmosphere, these non-CO_2 GHGs are very different from CO_2. The global warming potential (GWP), defined as the climate influence integrated over time and expressed relative to that of an equivalent mass of CO_2 emission, can be used to indicate the effectiveness of these non-CO_2 GHGs on global warming. All the major non-CO_2 GHGs have very large GWPs, e.g. CH_4 has a value of 25, N_2O has a value of 298, SF_6 has a value of 22,800 and NF_3 has a value of 17,200 over a 100-year time horizon [2].

In the global combat against the adverse effects of GHG emissions, non-CO_2 GHGs must be taken into account, as non-CO_2 GHGs currently account for about one-third of total CO_2-equivalent (CO_2-eq) emissions and 35–45% of total climate forcing from all long-lived GHGs (LLGHGs) [1]. Since most anthropogenic emissions of the non-CO_2 GHGs are linked to society's fundamental needs for food, energy and industrial products, their emissions will continue to increase and further warm the earth unless substantial efforts are undertaken to reduce them worldwide. Although the major GHGs CO_2, CH_4, and N_2O occur naturally in the atmosphere, human activities have greatly changed their atmospheric concentrations. From the pre-industrial era ending at about 1750 to the present, concentrations of these three greenhouse gases have increased globally by 43, 152, and 20%, respectively [2]. This can cause a huge problem for the climate as the natural balance in our environment can be broken. For example, the global carbon cycle is made up of large carbon flows and reservoirs. Billions of tons of CO_2 are absorbed by carbon "sinks" such as oceans and living biomass, meanwhile they are emitted to the atmosphere through various "sources". When in equilibrium, the emissions of CO_2 to and the removals of CO_2 from the atmosphere are roughly balanced. Once the balance is broken, adverse or even catastrophic consequences might happen. The climate can also be adversely affected if the balances of non-CO_2 GHGs are broken.

Among the various non-CO_2 GHGs, N_2O is mainly produced by biological processes that occur in soil and water and by a variety of anthropogenic activities in the agricultural, energy-related, industrial and waste management fields. Agricultural soil management, manure management, mobile source fuel combustion and stationary fuel combustion have been the major common sources of N_2O emissions. Presently anthropogenic N_2O emissions account for 3.1 ± 0.8 $GtCO_2$-eq per year [1], which is about 9% of the total radiative forcing (the difference of solar irradiance absorbed by the Earth and energy radiated back to space). The enhanced use of fertiliser could increase N_2O emissions, and most emissions of N_2O are associated with feeding the world's growing population. Because of the relevance to food production, careful measures need to be taken when reducing the N_2O emissions. The N_2O mitigation strategies [1] could include more judicious application of fertiliser, increasing nitrogen uptake efficiency by crops, expanding the use of nitrification inhibitors, improving manure management strategies and expanding access to sewage treatment [3–5].

In the global effort to control non-CO_2 GHG emissions, there was a success associated with the significant reduction in ODSs of about 5 $GtCO_2$-eq per year since 1990 [1], primarily because of the effectiveness of the Montreal Protocol on Substances that Deplete the Ozone Layer [6]. ODSs are man-made chemicals that damage the ozone layer in the upper atmosphere (the stratosphere), including chlorofluorocarbons (CFCs), halons, etc. Measures following the Montreal Protocol such as banning the use of CFCs and limiting the critical use of halons had certainly helped. HFCs and PFCs are families of synthetic chemicals that do not deplete the stratospheric ozone layer; thus, have been used as acceptable alternatives for ODSs under the Montreal Protocol. These compounds, however, along with SF_6 and NF_3, which are used in industrial sectors such as electrical transmission and distribution, semiconductor manufacturing, aluminium production, magnesium production and processing, are potent greenhouse gases. In addition to having high GWPs, SF_6 and PFCs have extremely long atmospheric lifetimes, resulting in essentially irreversible accumulation in the atmosphere once emitted. Although HFCs, PFCs, SF_6 and NF_3 are generally of small amounts [1], their controlled use is certainly important in the combat against global warming.

In the context of energy utilisation and GHG emissions, CH_4 and CO_2 are of paramount importance. Due to the vast amounts of literature available on CO_2 as a GHG, this review will not be focussed on CO_2 but on CH_4, which is not only the predominant constituent of natural gas as one of the major fossil fuel sources but also the second largest GHG next to CO_2. Presently CH_4 is the most abundant non-CO_2 GHG in the atmosphere. Methane's atmospheric increase since 1750 implies anthropogenic emissions of 340 ± 50 $TgCH_4$ per year or 8.5 ± 1.3 $GtCO_2$-eq per year [1], which accounts for about 20% of the total radiative forcing from all of the long-lived and globally mixed GHGs, estimated at around 40–50 $GtCO_2$-eq per year. Agriculture and fossil fuel exploitation together account for about 230 $TgCH_4$ per year or 5.8 $GtCO_2$-eq per year, or two-thirds of all human-derived CH_4 emissions. The energy sector is a significant contributor to anthropogenic methane emissions, at around 30% [7]. The main activities causing methane emissions in the energy sector include oil and natural gas systems, coal mining and biomass combustion. Meanwhile, waste treatment and other industrial processes lead to smaller amount of CH_4 emissions.

Greenhouse gas emissions including those from non-CO_2 sources are not entirely understood. Although the GHG emission inventory (the percentage contributions of gases to anthropogenic GHG emissions) provides a solid foundation for the development of a more detailed and comprehensive strategy for the global action against