DEVELOPMENT OF A GAS COMBUSTION SYSTEM FOR RADIOCARBON ANALYSIS OF HYDROCARBON GASES

Ąžuolas Ivickas[1], Žilvinas Ežerinskis[1], Laurynas Bučinskas[1], Laima Kazakevičiūtė-Jakučiūnien[1], Justina Šapolaitė[1]

[1]Lithuania, Center for Physical Sciences and Technology, Department of Nuclear Research, Accelerator Mass Spectrometry Laboratory
azuolas.ivickas@ftmc.lt

Radiocarbon (^{14}C) analysis is a powerful tool for distinguishing between biogenic and anthropogenic carbon sources, enabling the ability to track carbon dynamics in various systems and plays a critical role in carbon accounting, renewable energy certification, and greenhouse gas mitigation strategy development. Radiocarbon measurements of hydrocarbon gases such as methane are particularly valuable in differentiating between the contemporary and fossil fractions of gaseous carbon emissions. However, accurate ^{14}C determination in gaseous samples remains challenging as they require quantitative oxidation to CO_2 prior to accelerator mass spectrometry (AMS) or graphitization, as well as small sample sizes and the risk of isotopic fractionation during combustion [1].

Existing methods typically rely on catalyst-based oxidation systems that are time-consuming, prone to contamination, and require extensive laboratory infrastructure [1]. Therefore, the aim of this study was to develop and evaluate a novel gas combustion system (GCS) for the efficient and reliable conversion of hydrocarbon gases to CO_2 for AMS analysis. An alternative, catalyst-free GCS was designed and implemented utilizing controlled electric discharge ignition to oxidize sample gases mixed with oxygen, enabling more rapid sample preparation. The resulting CO_2 was compatible with automated graphitization equipment (AGE-3, Ionplus AG). The system was optimized to ensure complete combustion, high carbon recovery, minimal cross-contamination, and preservation of isotopic integrity.

Performance evaluation demonstrated quantitative combustion of methane at CH_4:O_2 ratios ≥ 1:2, yielding CO_2 recoveries of 78–88 % with negligible $\delta^{13}C$ fractionation. Cross-contamination between fossil and biogenic samples remained below 0.1 %, and analytical precision of radiocarbon measurements was sufficient to resolve mixed carbon sources. Environmental methane samples exhibited fully biogenic radiocarbon signatures, confirming system applicability under real-world conditions.

These results demonstrate that the developed GCS provides a rapid, robust, and reproducible alternative to conventional catalyst-based combustion systems. The GCS significantly enhances analytical capability for routine ^{14}C analysis of gaseous samples and supports improved source attribution of hydrocarbon gases in environmental, regulatory, and energy-related applications.

[1] C. Espic, M. Liechti, M. Battaglia, D. Paul, T. Röckmann, and S. Szidat, “Compound-Specific Radiocarbon Analysis of Atmospheric Methane: A New Preconcentration and Purification Setup,” Radiocarbon, vol. 61, no. 5, pp. 1461–1476, Jul. 2019, doi: https://doi.org/10.1017/rdc.2019.76.