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リュウ, タクヒ

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KYUSHU UNIVERSITY

Name　：LIU ZHUOFEI（リュウ　タクヒ）

Title　：Study on the application of graphite nanomaterials for laser desorption/ionization-mass spectrometry in food analysis
（食品分析のためのレーザー脱離イオン化―質量分析法におけるグラファイトナノ材料の応用に関する研究）

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Thesis Summary
論文内容の要旨

Food components are key determinants of food safety and quality. Numerous analytical technologies have been developed to characterize them. However, conventional analytical methods, such as gas chromatograph-mass spectrometry (GC-MS) and liquid chromatograph-mass spectrometry (LC-MS), generally require time-consuming and tedious pretreatment procedures. Moreover, although spatial heterogeneity of flavor compounds is considered to affect sensory perception, these methods are unable to retain spatial information of analytes. Therefore, to address these limitations, we aimed to develop surface-assisted laser desorption/ionization mass spectrometry (SALDI-MS) techniques based on graphite nanomaterials for comprehensive food component analysis.

Firstly, a graphite sheet-assisted LDI-MS method was developed to analyze small organic compounds with a variety of chemical properties. A grooved nanostructure was observed on the surface of graphite sheets with well-developed sp^2-conjugated carbon atoms. Compared to highly oriented graphite plates, the graphite sheet showed higher UV absorption and efficient ionization capability in LDI-MS. The detectable mass range of the present method covered 100–2,000 *m/z* in positive and negative ion modes. Using the present method, representative food compounds, such as amino acids, peptides, synthetic polymers, organic acids, fatty acids, nucleotides, sugars, advanced glycation end products, and polyphenols, were successfully detected. When applied to commercially available food, pharmaceutical, and biological samples, the present graphite sheet-assisted LDI-MS detected approximately 10-fold more MS peaks (~500 MS peaks) than matrix-assisted LDI (MALDI)-MS in both ion modes.

Secondly, a graphite carbon black (GCB)-assisted LDI-MS imaging technique was designed to visualize the spatial distribution of flavor compounds in food tissues. To improve the reproducibility of the GCB spraying, optical density (OD) was used as the quantitative indicator of the amount of sprayed GCB. OD showed a good linear correlation with the weight of sprayed GCB (R^2 = 0.9326). Using banana tissue as a model sample, the highest intensity of [$^{13}C_5$, $^{15}N_1$]glutamate was achieved at the OD value of ~200. Compared with the conventional cycle-number-controlled spraying method (RSD = 24.9%), the OD-assisted spraying method markedly improved reproducibility (RSD = 5.0%). Under these optimized conditions, the GCB-assisted LDI-MS imaging method successfully visualized heterogeneous distributions of food compounds such as sugars, amino acids, and organic acids, on the banana tissue. Moreover, the number of MS peaks detected by GCB-assisted LDI-MS (98 peaks in positive mode and 111 peaks in negative mode)

was significantly larger than that obtained by MALDI-MS imaging.

In conclusion, the present study established graphite nanomaterial-assisted LDI-MS methods for food analysis. The graphite sheet-assisted LDI-MS technique provides a robust platform for comprehensive analysis of small organic compounds in food and biological samples. In addition, the GCB-assisted LDI-MS imaging, combined with OD-assisted spraying, enables highly reproducible imaging and effective visualization of spatially heterogeneous compounds in food tissues. These analytical techniques are expected to serve as valuable tools for food quality and safety assessment.