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在线全文

# 卫生检验样品前处理新技术与应用发展*

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**【摘要】** 卫生检验工作涉及的样品形态多、基质复杂，待测成分多、量值不一，应用场景多、要求各异。作为卫生分析和检测的重要环节及关键步骤，卫生检验样品前处理不仅与检验方法的重现性和检验效率有关，而且直接影响检验结果的准确性、灵敏度、可靠性及后续分析可行性。传统样品前处理技术存在效能和通量低、通用性差、有机溶剂用量大、后续分析衔接受限等问题，难以满足公共卫生与预防医学研究及实践需求。围绕提高效率和自动化程度、增强选择性和灵敏度、强化在线检验、实现绿色环保等目标，近年来卫生检验样品前处理技术正逐步实施升级改造，并涌现出众多新技术。本文重点结合新材料发展、人工智能应用、在线处理衔接及特定场景检验需求驱动等，综合评述卫生检验样品前处理新技术及应用，同时关注到卫生检验组学和质谱成像样品处理方法。为卫生检验样品前处理的绿色、高通量发展和卫生检测创新体系构建提供借鉴。

**【关键词】** 卫生检验　样品前处理　新技术　人工智能　应用

# New Technologies and Application Developments in Sample Pretreatment for Public Health Laboratory Testing

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**[Abstract]** Public health laboratory testing involves a wide range of sample types, complex matrices, diverse target analytes with varying concentrations, and multiple application contexts with different analytical requirements. As a critical step in public health laboratory analysis and testing, sample pretreatment plays a decisive role in ensuring the reproducibility and efficiency of the analytical methods. It directly affects the accuracy, sensitivity, and reliability of testing results, as well as the feasibility of downstream analyses. Traditional sample pretreatment techniques face persistent challenges, including low efficiency, limited throughput, restricted universal applicability, high organic solvent consumption, and poor compatibility with downstream analytical procedures. These limitations constrain their capacity to meet the evolving demands of research and practice in public health and preventive medicine. In recent years, technological advances have focused on improving efficiency and automation, enhancing selectivity and sensitivity, facilitating online testing capabilities, and promoting environmental sustainability. Sample pretreatment techniques in public health laboratory testing have been undergoing progressive upgrades, and numerous novel technologies have emerged. The paper provides a comprehensive review of new technologies and applications in the field. We focused on the development of new materials, the application of artificial intelligence, connections for online processing, and the approaches tailored to the demands of specific testing settings. We also discussed sample processing for omics analyses and mass spectrometry imaging methods relevant to public health laboratory testing. These advances are expected to support the development of greener and higher-throughput sample pretreatment and foster innovation in the public health laboratory testing system.

**[Key words]** Public health laboratory testing　Sample pretreatment　Novel technology　Artificial

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intelligence Application

一个完整的卫生检验过程，通常包括采样、样品前处理、分离、检测、数据分析五个环节，其中样品前处理是用时最长、对分析结果影响最大的关键步骤，特别是在处理突发公共卫生事件或开展应急检验工作时，及时、正确的样品处理是卫生分析"一锤定音"的关键控制点。样品前处理（sample pretreatment）是指为满足待测物检验和维护后续仪器分析性能需要，对样品进行适当前处理（如提取、浓缩、净化、衍生化等），使被测组分转变成可测定形式的过程及方法。卫生检验涉及的样品种类多、组成复杂、形态多样、被测成分含量差异大，需要结合分析目标和后续检测方法对样品进行适当前处理才能获得满意结果。同时，公共卫生现场检验及突发公共卫生事件处置等特定场景检验、人群队列研究和流行病学调查产生的大样本检测及高通量组学分析等对卫生检验提出了更高要求。但传统的样品前处理技术，例如，用于有机物分析的固相萃取（solid-phase extraction, SPE）、液-液萃取（liquid-liquid extraction, LLE）等操作繁琐、处理效率低，溶剂消耗大、对操作人员和环境不友好，灵敏、选择性捕捉目标物的能力以及前处理技术与后续分析方法的协调性无法满足现代卫生检验与检疫需要[1-2]。

近年来，新材料合成持续推动新型萃取剂和吸附剂快速发展、人工智能辅助样品前处理设计并提升装置的自动化水平、物理及化学学科进步推进了在线（on-line）前处理接口不断优化，大大驱动卫生检验科技进步。为了提高检验效率和自动化程度、增强检测方法选择性和灵敏度、推进在线检验发展进程、实现绿色环保目标，卫生检验样品前处理技术正逐渐实现对传统方法的升级改造和技术创新。本文将重点综合评述卫生检验样品前处理新技术及应用，并关注卫生工作特定场景的样品预处理方法，为卫生检验样品前处理的绿色、高通量发展和检测体系创新提供借鉴。

## 1 新型SPE技术

SPE技术的核心是固相萃取剂。为缩短传统SPE技术前处理时间、减少有机溶剂和萃取剂用量、增强吸附剂通用性和特异性、提升自动化和智能化水平，围绕萃取剂与目标分子间相互作用，目前开发出了适于多场景应用的功能材料，包括碳纳米材料（carbon nanomaterials, CNMs）、磁性纳米颗粒（magnetite nanoparticles, MNPs）、金属有机框架（metal organic frameworks, MOFs）、共价有机框架（covalent organic frameworks, COFs）、分子印迹聚合物（molecularly imprinted polymers, MIPs）、离子液体（ionic liquids, ILs）等[1-18]。作为SPE萃取剂，碳纳米管（carbon nanotubes, CNTs）和石墨烯等碳基纳米材料对非极性目标化合物有较好吸附作用，并且进一步功能化后能够获得更高的选择性。例如，CNMs-ILs复合材料表现出高选择性吸附能力[9-10]。近年来，新材料及其功能化已形成众多优良的固相萃取剂，并以不同的应用模式产生了多种新型SPE技术（图1）。例如，有研究[8]合成了氨基功能化MOFs材料（UiO-66-$NH_2$）作为SPE吸附剂，已用于高效、灵敏测定茶饮料中卤化乙酸消毒副产物。

### 1.1 分散固相萃取技术

分散固相萃取（dispersive solid-phase extraction, dSPE）是基于传统柱固相萃取技术发展起来的一种前处理方法。将固相萃取剂直接分散于含有待测成分的样品溶液中，使固体吸附剂与目标成分充分接触并高效富集，然后利用离心或过滤等方式将吸附剂与溶液分离，再使用合适的溶剂将固相吸附剂上的目标成分洗脱下来。常用吸附剂（如$C_{18}$、活性炭等）对非极性化合物有一定吸附效果，但特异性差。新型dSPE萃取剂分散性好、吸附能力和特异性强、结构稳定。基于功能化COFs、COFs-MOFs、表面羧基化和氮原子掺杂MOFs材料的dSPE技术结合串联质谱已成功用于环境及生物组织卫生检验[11-12, 19-20]。

### 1.2 磁固相萃取技术

磁固相萃取（magnetic solid-phase extraction, MSPE）技术是以磁性或可磁化的材料作为吸附剂基质的一种新型SPE技术。该技术能够克服传统SPE小柱在样品处理过程中易堵塞和操作繁琐等问题，也可避免dSPE离心或过滤步骤。MSPE的核心是磁性吸附剂及磁性材料功能化。例如，经典$Fe_3O_4$基材料表面修饰$SiO_2$（$Fe_3O_4@SiO_2$）能增强其化学稳定性，避免酸性环境溶解，增加卫生分析应用场景适用性[4, 6]。目前，碳基MNPs、MOFs衍生物磁性纳米材料、磁性COFs纳米颗粒、磁性MIPs材料、功能化聚合物MNPs等作为吸附剂已广泛应用于MSPE分析中[21-26]。

### 1.3 分子印迹固相萃取技术

分子印迹固相萃取（molecularly imprinted SPE, MISPE）是以MIPs作为固相萃取剂的一种高选择性、高灵敏度新型固相萃取技术。针对样品复杂基质中目标分子的结构设计"模板孔穴"，实现特异性吸附[1]。应用中可制备分子印迹柱，也可以dSPE和MSPE模式进行操作[12, 25]。基于MIPs材料的分子印迹柱组装较复杂，且不利于大体

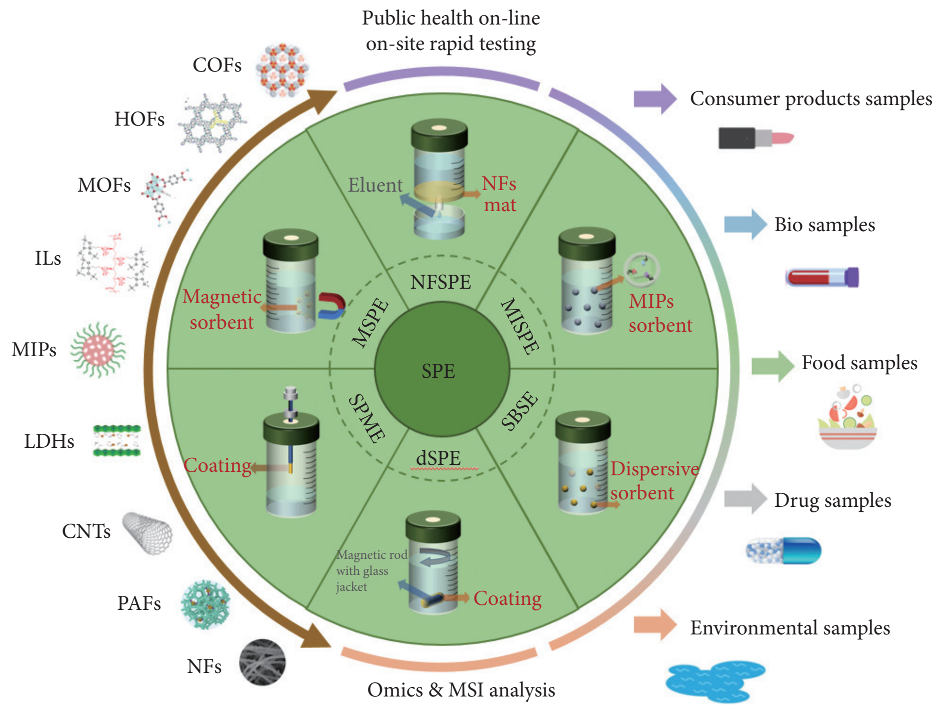


**图 1　固相萃取新技术及应用**

**Fig 1　New technologies and applications of solid phase extraction**

COFs: covalent organic frameworks; HOFs: hydrogen-bonded organic framework materials; MOFs: metal–organic frameworks; ILs: ionic liquids; MIPs: molecularly imprinted polymers; LDHs: layered double hydroxides; CNTs: carbon nanotubes; PAFs: porous aromatic frameworks; NFs: nanofibers; dSPE: dispersive solid-phase extraction; MSPE: magnetic solid-phase extraction; MISPE: molecularly imprinted solid-phase extraction; NFSPE: nanofibers solid-phase extraction; SPME: solid-phase microextraction; SBSE: stir bar sorptive extraction; MSI: mass spectrometry imaging.

积样品前处理，将NMPs结合MIPs形成磁性MIPs作为萃取剂更有利于环境、食品和生物样品分析的实际应用[25-26]。为满足MISPE在水相介质样品前处理中应用，有人制备了比表面积大和易于功能化修饰的表面印迹微球，使其对水相目标分子的识别和吸附能力更强[27]。基于MOFs的分子印迹SPE吸附剂（MOFs@MIPs）是以MOFs为核、MIPs为壳复合而成的新型材料，该材料将 MIPs高选择性与MOFs高吸附容量相结合，能显著提高对目标分子的处理效率且可重复多次使用[28]。

### 1.4　纳米纤维固相萃取技术

纳米纤维固相萃取（nanofibers SPE, NFSPE）是以功能化纳米纤维为萃取剂的新型SPE技术。纳米纤维具有高比表面、高孔隙率以及表面化学性质可调控等显著优势，使其作为SPE介质在样品前处理领域展现出巨大的应用潜力[6]。通过优化制备方法和功能化修饰，可实现对纳米纤维直径、孔隙率、取向度等微观结构的精准调控并具备多重吸附机制，为复杂样品痕量目标物的高效萃取提供技术支撑[17]。目前，聚多巴胺（polydopamine, PDA）功能化聚苯乙烯纳米纤维、热交联聚乙烯醇纳米纤维、MOF-808修饰的PDA功能化聚丙烯腈纳米纤维及基于磺酸化聚苯胺纳米纤维的96孔板SPE技术均已成功应用于环境及食品污染物检测[29-31]，大大缩短了样品前处理时间。

### 1.5　固相微萃取技术

不同于一般SPE吸附剂（固定相）对目标物的选择性吸附与洗脱，固相微萃取技术（solid-phase microextraction, SPME）技术是建立在固相（或涂层）-样品基质间分配平衡基础上的新型处理技术，已形成MIPs-dSPME[12]、管内固相微萃取（in-tube SPME）[13]、ILs-SPME[7, 14]、膜固相微萃取[17] 等多种应用模式。作为SPE技术的进一步发展，SPME是一种集采样、萃取、浓缩和进样于一体、不使用或少使用有机溶剂的微萃取技术[3]。SPME无需淋洗和浓缩，装置简单便携，纤维可重复使用，操作快速且易实现后续在线联用分析[14]。近年来，在SPME探针的固相涂层材料及涂渍技术、萃取模式、联用技术等方面快速发展，在环境、食品和生物样品检验中得到广泛应用[32-43]。

### 1.6　搅拌棒吸附萃取技术

搅拌棒吸附萃取（stir bar sorptive extraction, SBSE）技术是一种环境友好型固相萃取新技术，它利用固相萃取搅拌棒涂层吸附材料与目标成分之间的化学吸附作用，

将目标物质从样品中分离出来,然后再根据目标成分的挥发性选用浸入式或顶空萃取方式将目标物质从吸附涂层上溶解或解吸下来。SBSE将搅拌与吸附目标分子同步进行,使用简便。与SPME相比,SBSE涂层量大,因此具有更高的萃取效率[4]。SBSE的核心是介质涂层,应用中应重点关注搅拌棒涂层的稳定性和萃取选择性[11]。目前,功能化MOFs及MIPs等新型涂层材料已成功用于食品、生物样品SBSE处理中[44-46]。

在卫生检验领域,自动、便捷、高效的样品前处理装置是提升检测通量和速度、减少人为误差和实现标准化的关键。近年来,随着磁性材料、微流控技术、人工智能辅助能化设计的发展,一系列创新装置和高通量样本处理平台已投入应用或处于研发阶段[22-27, 41]。全自动SPE工作站集成样品加载、柱活化、上样、淋洗、洗脱等步骤,支持多通道并行处理(如96孔板格式),能高通量处理复杂样品[31, 47]。MSPE自动化系统利用功能化MNPs(如$Fe_3O_4$@MOFs、$Fe_3O_4$@MIPs)吸附目标物,通过磁分离取代离心/过滤,大幅简化步骤,并提高回收率[21-26]。另一方面,将前处理模块直接耦合到分析仪器,实现“样品进-结果出”,发展样品前处理-分析仪器on-line联用技术,可实现样品前处理与后续仪器检测的无缝对接,防止或减少人工操作中间环节产生的误差,提高后续分析的灵敏度、准确性与重现性[48-49]。

## 2 新型液相微萃取技术

液相微萃取(liquid phase microextraction, LPME)凭借其操作简单、快速、环境友好和成本低的优势,受到广泛关注。近年来,液相微萃取技术不断发展和完善,衍生出了多种新技术(图2)。

### 2.1 分散液液微萃取

分散液液微萃取(dispersive liquid-liquid microextraction, DLLME)是将萃取剂与分散剂混合并注入样品中形成乳浊液,增加萃取剂与水相的接触面积,从而提高萃取效率。DLLME具有高效、低溶剂消耗、操作简便等优点,广泛应用于环境、水样、食品及生物样品中的污染物分析[50]。超声辅助DLLME(ultrasound-assisted DLLME, UA-DLLME)能通过超声波促进溶剂的分散,进一步提高萃取效率,并有效减少溶剂用量,已在农药残留、环境污染物及生物样品药物检测中表现出较高的灵敏度和良好的富集能力[51-52]。目前,一些环境友好型溶剂,如低共熔溶

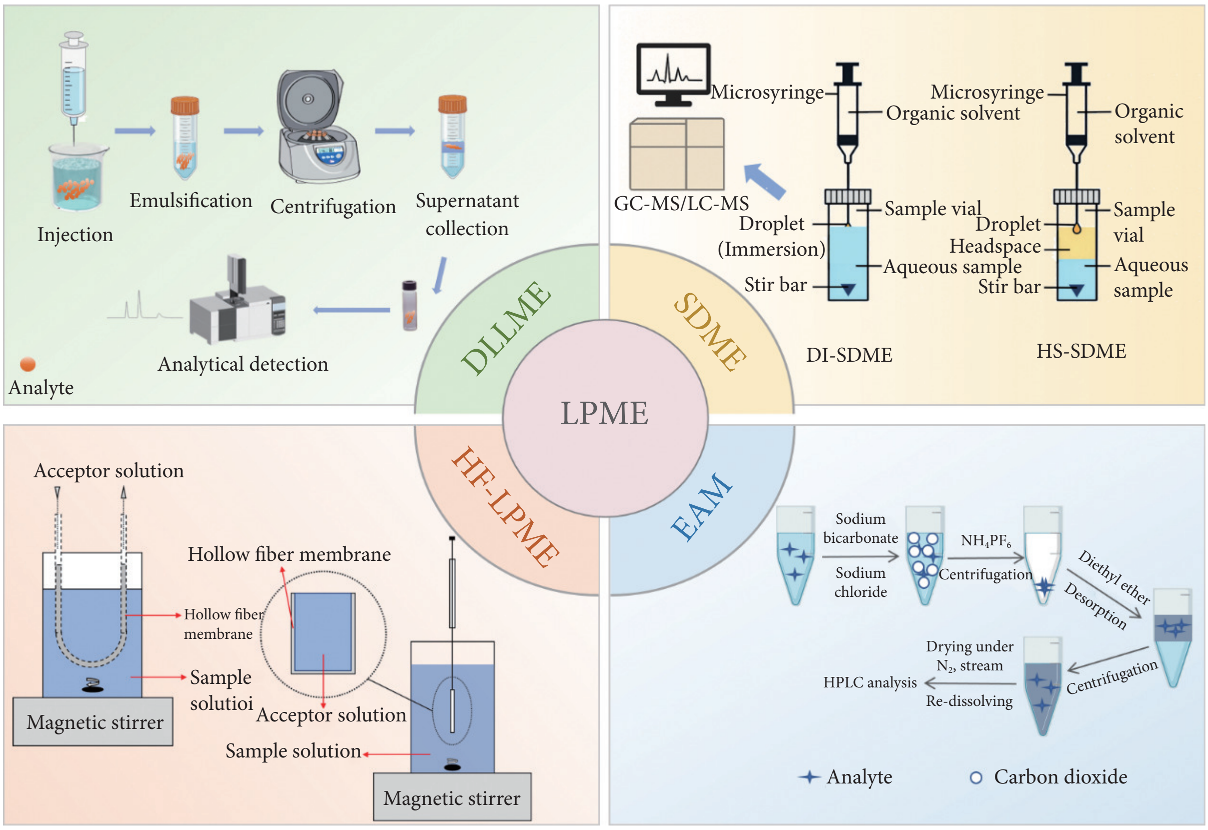


图 2 液相微萃取

**Fig 2 Liquid phase microextraction**

LPME: liquid phase microextraction; DLLME: dispersive liquid-liquid microextraction; SDME: single-drop microextraction; HF-LPME: hollow fiber liquid-phase microextraction; EAM: effervescence-assisted microextraction.

剂(deep eutectic solvents, DESs)和ILs被引入DLLME, 替代了传统的有机溶剂[53], 实现绿色处理。

### 2.2　单滴液相微萃取

单滴液相微萃取(single-drop microextraction, SDME)是一种基于传统液液萃取发展而来的绿色微萃取技术。该技术通过在微量注射器针尖悬挂微升级别的有机溶剂液滴, 实现对目标组分进行富集和浓缩, 具有操作简便、溶剂用量少、成本低和绿色环保等优点。根据操作方式不同, SDME可分为直接浸没式与顶空式两种形式, 前者适用于非挥发性或中等极性的目标物质, 而后者则用于易挥发或复杂基质中挥发性分析物的萃取[54]。目前, SDME已广泛应用于环境和生物样品分析领域, 常与质谱联用以提高检测灵敏度与选择性[55-56]。

### 2.3　中空纤维液相微萃取

中空纤维膜-液相微萃取(hollow fiber liquid-phase microextraction, HF-LPME)利用中空纤维膜作为有机试剂的固定载体, 实现对样品的高效萃取[57]。目前常用的HF有聚丙烯、聚四氟乙烯等。HF-LPME不仅解决了传统单滴液相微萃取中液滴稳定性不足的问题, 而且在处理复杂样品基质方面表现出色, 集净化、萃取、浓缩处理于一体, 在卫生检验中应用广泛[58]。

### 2.4　泡腾辅助微萃取

泡腾辅助微萃取(effervescence-assisted microextraction, EAM)是一种新型绿色样品前处理技术, 其核心机制是利用$CO_2$供体、$H^+$供体在溶液中充分反应产生$CO_2$气泡, 加速新型萃取剂在溶液中快速分散, 增大与目标物的接触面积, 进而提高萃取效率。相较于传统方法, EAM具有操作简便、有机溶剂用量少、萃取时间短等优势, 避免了传统微萃取中对超声波和漩涡混合等外部能量源的依赖, 在环境和食品安全分析中已广泛应用[59]。

## 3　QuEChERS技术革新及应用

QuEChERS(Quick, Easy, Cheap, Effective, Rugged, and Safe)技术在吸附材料优化、自动化集成及应用范围扩展等方面取得显著进步, 已成为食品、环境及生物样品中多残留分析前处理的主流方法。

### 3.1　新型吸附剂及绿色提取剂的应用

目前, 新型吸附剂、低毒性溶剂替代提取溶剂或可生物降解DESs已逐渐应用于QuEChERS技术[53, 60-61]。微型化QuEChERS结合低体积提取, 可减少溶剂用量。近年来, 在吸附剂基础上陆续开发出QuEChERS 净化柱, 例如Sin-QuEChERS Nano 兽药残留分析净化柱、QuEChERS-HF净化柱等, 使用操作简单、处理快捷[62]。我国学者将特定纳米结构填料与固相吸附相结合作为净化材料, 填充于针管式柱管中制备SinChERS净化柱, 可一步完成净化过程, 实现环保、快速、特异性前处理目标[63]。

### 3.2　自动化、在线检测前处理技术

全自动QuEChERS处理平台能实现自动添加提取溶剂、内标或保护剂、自动涡旋样品和振荡、自动完成样品离心操作、移取提取液并进行基质分散固相萃取操作、自动移取净化后的上清液供质谱分析[60]。自动QuEChERS与便携式GC-MS联用适于野外环境样品分析[64]。另一方面, 发展在线QuEChERS处理与后续检测联用分析技术, QuEChERS-HPLC-MS/MS可用于农药残留分析和安全评价、纳米银增强基底QuEChERS-拉曼光谱实现药物的快速筛查[61, 65]。

QuEChERS技术通过材料创新和自动化不断突破传统局限, 已从农产品检测的快速前处理拓展到环境和生物样本污染物及药物残留监测领域, 但复杂样品中共提取物的干扰和基质效应仍需制订针对性净化策略[64]。

## 4　针对特定场景和代谢组学研究需求的样品前处理新技术

### 4.1　公共卫生现场检验需求及前处理新技术

公共卫生现场对环境、生物样本等检测或对突发公共卫生事件处理时需要与检测仪配套的样品处理方法或待测成分快速浓缩技术, 并尽可能简单、快速、高效、规范[47]。例如, 对环境水体进行污染物调查分析时, 因采样量大、运输繁琐、待测成分含量低, 需要在采集现场将水中目标成分高倍富集于吸附剂或萃取小柱上, 带回实验室后洗脱检测。前述SPE固相萃取新技术及快速处理装置, 可针对目标成分结构特征设计填料或吸附剂, 增强前处理选择性[48-49]。同样, 对于空气样品的滤膜处理, 可结合新型纳米材料的合成和功能化改良, 开发适于空气污染物分析的集气管装置[17, 30, 57-58]。

目前, 公共卫生现场检验应用较多的新型前处理技术包括SPE技术对环境/食品新污染物的筛查、基于纳米材料增强的微生物富集技术、免疫磁珠核酸/蛋白和试剂盒快速提取技术用于现场病原体和食品安全检测、微流控集成化前处理芯片技术可在纳升级流体尺度下实现目标分析物的分离、纯化和提取等样品前处理步骤等, 降低了样本运输和储存成本, 缩短了检测周期[47-49, 64-65]。人工智能设计和3D打印定制化装置可快速制作微流控芯片或适配器, 满足特定样品处理需求[66]。

### 4.2　代谢组学和空间代谢组学分析前处理

代谢组学分析中的样品前处理技术直接影响卫生检

测的覆盖度、灵敏度和重现性。近年来，为应对复杂生物样本挑战，新型纳米材料吸附、SPME、MSPE、在线SPE、QuEChERS技术等都已广泛应用于代谢组学研究[18, 67]。如前所述样品前处理技术在线检测或目标物提取衍生化技术在提升代谢物检测灵敏度方面效果显著，实施多技术融合提取能够有效提升代谢物覆盖度[48-49, 68]。

空间代谢组学和质谱成像技术揭示代谢物的原始空间分布信息和代谢异质性，在卫生检验领域具有重要价值[69]。空间代谢组学分析样品前处理技术需兼顾代谢物稳定性、空间分辨率和检测灵敏度，同时匹配后续检测技术，特别是针对MALDI-MSI的基质组合及喷涂优化的前处理方法应用广泛[70]。空间代谢组学前处理技术核心在于原位固定、精准提取、低损检测。人工智能算法有助于预测最佳切片厚度、基质条件或溶剂组合，减少人为前处理误差。软件工具能自动校正基质结晶不均或组织处理变形带来的信号偏差。未来随着MSI和纳米材料的进步，空间分辨率与检测通量将进一步提升，为卫生检验研究提供更精细的代谢图谱[71]。

## 5 总结与展望

卫生检验工作不仅涉及复杂的环境和食品样本，而且要面对大量人群和生物组织，要根据不同样本基质、待测成分性质、仪器分析要求等进行针对性样品前处理。本文重点介绍了新型萃取剂的发展、应用模式变化及其衍生的SPE和LPME技术，评述了QuEChERS技术在集成柱净化及在线检测一体化方面的革新及应用进展，并结合现代卫生检验前沿科学问题，探讨了代谢组学和空间代谢组学分析样品前处理关注的内容和技术指标，为卫生检验前处理技术创新及应用拓展提供借鉴。近年来，新材料、人工智能、绿色环保试剂等推动样品前处理技术和卫生检测体系不断创新，但面对公共卫生和预防医学需求，卫生检验样品前处理技术的通用性和检测通量有待加强、自动化和智能化水平有待进一步提升、集成化和微型化是需要重点攻克的瓶颈。

* * *

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