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论文题目：微流控界面聚合制备相变微胶囊及其储热性能研究

微流控界面聚合制备相变微胶囊及其储热性能研究

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摘要

移动电子终端向轻薄化、高集成化、高算力方向发展,随着人工智能与边缘算力的普及,芯片热流密度持续攀升,局部热积聚已成为制约器件运行稳定性、使用寿命与算力释放的核心瓶颈。传统主动散热难以适配终端热管理低碳高效的升级需求,基于相变微胶囊的被动储热技术成为缓解局部热积聚的重要路径。尺寸均一可控的相变微胶囊可有效削减温度峰值,其储热性能高度依赖于液滴稳定成形、壳层界面构筑及限域效应;但传统机械搅拌法制备的微胶囊普遍粒径分布宽、壳层结构可控性弱、储热性能批次差异大,无法从界面聚合反应层面对微胶囊壳层结构实现调控。

本文采用微流控界面聚合法,以异佛尔酮二异氰酸酯 (IPDI) 和二乙烯三胺 (DETA) 为反应单体制备聚脲-十八烷相变微胶囊;该方法可生成分散性良好的液滴,构建出高度均一的油-水反应界面,使 IPDI 与 DETA 的加成聚合限定在规整可控的液-液界面上进行,实现界面聚合反应进程与聚脲壳层结构的有效调控。本文结合计算流体动力学 (CFD) 模拟分析和在线显微图像识别研究方法,定量表征微胶囊前驱体的粒径大小及分布,重点研究液滴稳定成形、聚脲壳层形成及芯材相变行为的调控机制,其中的核心化学问题在于揭示 IPDI 与 DETA 在油-水界面聚合反应如何决定聚脲壳层的结构。

研究表明,毛细管数调控的界面张力决定液滴的形态转变,当分散相毛细管数 Ca_d 为 0.0072-0.0216 时,可形成稳定的球形微胶囊前驱体,为界面聚合提供均一的反应模板;IPDI 与 DETA 在油-水界面发生逐步加成聚合,形成致密连续的聚脲壳层,壳层交联稳定性与反应单体配比直接相关。随着十八烷 (OD) /IPDI 配比由 3:1 增至 7:1,芯材十八烷的结晶温度由 17.8°C 升至 20.8°C,熔融焓由 151.1J/g 提升至 202.2J/g,其本质是核壳反应单体配比变化改变了壳层对相变芯材的限域效应,进而调控微胶囊整体的相变焓值;经 100 次冷热循环后,样品相变峰形与焓值无明显衰减,表明聚脲壳层具有优异的结构稳定性与限域持久性。本研究建立了相变微胶囊的微流控界面聚合可控制备方法,揭示了从液滴成形到限域相变的连续作用机制,对高性能相变储能微胶囊的可控制备开辟了新途径。

关键词: 微流控; 界面聚合; 聚脲微胶囊; 壳层限域; 相变行为

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1. 引言

随着移动通信与人工智能技术的快速迭代，手机、笔记本电脑等移动终端持续向轻薄化、高集成度、高算力方向发展，加之AI边缘计算、高帧率渲染等功能的普及进一步推高了芯片、电池及电源管理模块的热流密度^[1]；同时，终端内部散热空间随机身轻薄化持续压缩，局部热量积聚已成为制约器件运行稳定性、使用寿命与算力释放的核心瓶颈^[2,3]。

当前终端热管理体系仍以风扇散热、金属导热片、热管等主动/导热方案为主体，在整机热负荷调控中发挥核心作用。但这类方案依赖流体或固体的宏观导热散热，针对电子元件局部高发热点和热积聚，难以实现精准快速的温度峰值抑制与温度均化。随着终端能效标准的持续收紧与产业低碳发展导向，电子信息产业正加速向高能效、低功耗方向升级；仅依靠传统主动散热方案，已难以兼顾“局部精准温控”与“系统能效提升”的双重需求。**因此，高效被动式储热技术成为补充主动散热体系、解决局部过热问题、提升热管理系统整体能效的重要技术路径。**

相变材料可在特定温度区间内通过可逆固-液相变过程吸收与释放潜热，实现热能的时空转移与温度调控，是被动式热管理的核心储热介质。将相变芯材包覆于聚合物壳层中形成的相变微胶囊，凭借核-壳结构的空间限域作用，既能有效降低液态芯材的泄漏风险、提升冷热循环稳定性，又能完整保留芯材的高潜热特性，为移动终端的局部热点温控与温度均化提供了材料基础^[4,5]。

相变微胶囊的制备工艺经历了从批量粗放制备向精准调控的发展历程。传统制备方法以搅拌乳液聚合、原位聚合、喷雾干燥等为主，其中搅拌乳液聚合法通过机械剪切作用形成油水分散体系，诱导单体在液滴界面发生聚合形成壳层，是目前应用最广泛的技术路线^[6]。**但该方法中剪切场分布不均，液滴粒径分布宽、球形度差异大，壳层厚度、界面结构与致密性难以精准调控，最终导致产品储热性能一致性差，难以满足高精度局部热管理的应用需求^[7]；喷雾干燥等其他传统方法同样存在粒径可控性弱、壳层缺陷多等共性问题，难以实现微胶囊结构与性能的定向设计。**

微流控技术为相变微胶囊的精准可控制备提供了全新的技术路径。在微米级受限通道中，通过流体剪切力与界面张力的动态平衡，生成尺寸高度均一、单分散性优异的油-水液滴体系；其既可为界面聚合提供稳定可控的反应模板，也为定量研究界面张力、流体黏性与剪切作用对液滴形成、断裂过程的调控规律提供了理想的研究平台^[8]。

国内外学者围绕微流控制备相变微胶囊开展了大量探索性研究。Hao等^[9]利用同轴微流控装置实现了相变微胶囊的连续可控制备,验证了两相流速对液滴粒径的连续调控作用。安妮等^[10]进一步系统探究了核壳流速比对微胶囊核壳比例及宏观储热性能的影响规律。现有研究已充分证实微流控技术在提升微胶囊粒径均一性、调控核壳比例方面的显著优势,但现有工作更多集中于“如何制得尺寸均一、性能较好的微胶囊”,对液滴形成之后的界面聚合以及聚合壳层对芯材结晶/熔融的影响仍多作为独立的工艺或性能问题讨论。换言之,从瞬态流体界面到稳定聚合物限域界面的连续演化机制,以及这种界面演化如何决定最终相变行为,仍缺少系统认识。

近年来,人工智能与计算机视觉方法正逐渐进入微流控研究领域。例如Tang等^[11]采用流动聚焦法制备了水包油包水双乳液,建立了内核液滴与双乳液粒径的机器学习预测模型,为双包埋液滴的可控制备提供了量化基础。但将视觉识别用于相变微胶囊液滴颈缩、断裂、成球全过程的在线量化,并与界面受力模型直接对应的研究仍较少。

从材料化学的视角来看,相变微胶囊的储热性能并非单纯由芯材的本征特性决定,而是液滴界面形成、界面聚合反应、壳层限域效应等前后衔接的关键环节共同作用的结果。油-水界面处的界面聚合反应直接决定微胶囊壳层的化学结构与微观形貌,而聚合物壳层形成后产生的空间限域作用,又会进一步影响材料的相变温度、过冷度与相变焓值等核心储热参数。

基于上述研究现状,本文将聚脲-十八烷微胶囊视为一个由流体界面、反应界面、限域界面连续演化的体系,重点探讨三个相互关联的科学问题:(1)黏性/剪切作用与界面张力的竞争如何控制前驱液滴的失稳、断裂与稳定成球;(2)水相 DETA 与油相 IPDI 如何在油-水界面发生聚合并形成连续聚脲壳层,核壳组成如何影响壳层完整性;(3)聚脲壳层形成后,空间限域如何进一步影响十八烷的结晶温度、过冷度及相变焓。本文采用 CFD 模拟仿真与在线显微图像识别建立微胶囊前驱体成球过程的“模拟-实验”对应关系,结合 FTIR、XRD 和 SEM 分析界面聚合与核-壳结构,并利用 DSC 建立核壳组成、限域程度与相变行为之间的构效关系。

本文创新之处在于:(1)建立了相变微胶囊的微流控界面聚合可控制备方法。相较传统搅拌乳液聚合法,该方法通过微流控生成单分散液滴构建均一反应界面,实现了界面聚合反应由传统无序、随机状态向分散、定量可控状态的关键转变,解决了聚脲壳层厚度与交联结构难以精准调控的技术难题。(2)采用 CFD 模拟与在线显微图像识别相结合的研究方法,揭示了微胶囊前驱体成球机理。本文为通过界面聚合化学设计主动调控相变微胶囊的储热性能提供了实验基础,对高性能相变储能微胶囊的可控制备开辟了新途径。

2. 实验设计与研究方法

2.1 实验材料

本实验所用主要药品及试剂包括十八烷（OD）、二乙烯三胺（DETA）、异佛尔酮二异氰酸酯（IPDI）、聚乙烯醇（PVA）和十二烷基硫酸钠（SDS），均为分析纯（AR），由上海麦克林生化科技股份有限公司提供；实验用水为去离子水。本实验主要需要配置两种液体，分散油相和连续水相，如图 1 所示。

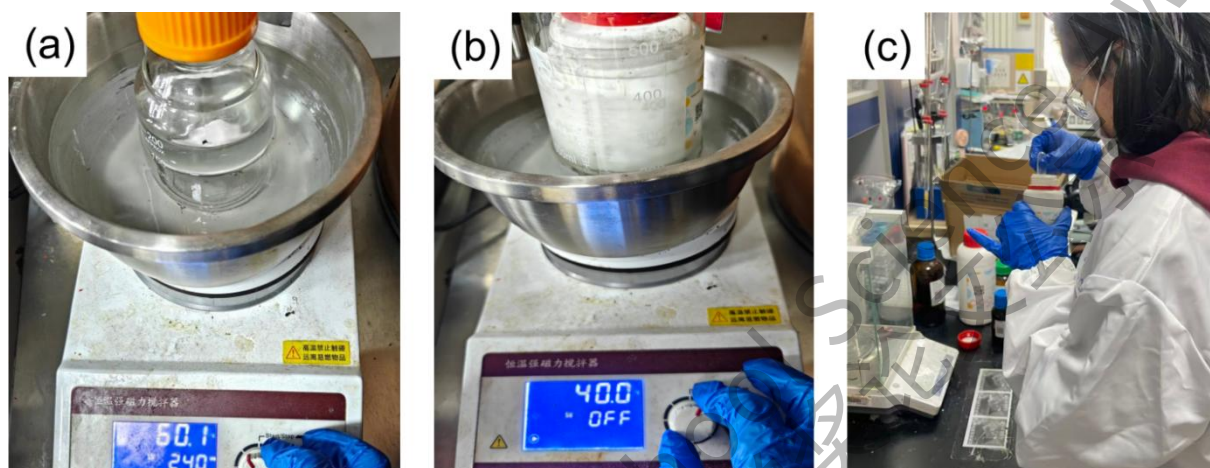


图 1. 实验原料溶液配制过程

(a) PVA、SDS 加热水溶解；(b) 十八烷水浴加热溶解；(c) 分散油相配制

连续水相配制：水相中 PVA 主要用于提高水相黏度，SDS 用于降低油-水界面张力、促进液滴形成。二者共同提高液滴在微流控成球及后续固化过程中的稳定性，减少液滴并聚、黏连和破裂。按照设定的质量分数，将 PVA、SDS 和二乙烯三胺（DETA）加入去离子水中配制连续水相。配制时首先向洁净容器中加入约 80% 的水，在持续搅拌条件下缓慢加入 PVA，避免粉末结团。随后将溶液加热至 60°C ，直至 PVA 完全溶解，形成澄清且无可见颗粒的溶液。待溶液冷却至室温后，依次加入 SDS 和 DETA，并用去离子水补足至设定总质量。连续水相中 DETA、PVA 和 SDS 的质量分数分别为 4.0wt%、1.5wt% 和 1.0wt%。

分散油相配制：OD 作为相变芯材，通过固-液相变实现用于实现潜热储存与释放；IPDI 作为油溶性壳材前驱体，与熔融 OD 混合形成分散油相，实验前，OD 置于 40°C 水浴中充分熔融，待其转变为澄清、均一液体后使用。DETA 作为水溶性交联反应单体，由连续水相向油-水界面扩散，并与 IPDI 的异氰酸酯基在油-水界面发生聚合反应，逐步形成聚脲壳层。IPDI 具有较强的水分敏感性，取用过程中尽量缩短其暴露于潮湿空气中的时间，取样后立即密封保存。分散油相按照设定的 OD/IPDI 质量比配制。以 OD/IPDI 质量比为 3:1 为例，称取质量为 3 g 的熔融 OD 后，加入质量为 1 g 的 IPDI，在 40°C 条件下 300 r/min 搅

拌 30 min，直至体系均一且无可见分层。

各溶液均以当次实验的实际称量结果为计算基准，采用质量法确定各组分的配比，所用容器在称量前保持洁净、干燥。连续水相使用前采用静置脱泡，以减少气泡进入芯片后引起的流动波动和通道堵塞；油相配制完成后加盖保温，并尽量缩短其在进样前的停留时间，以降低 IPDI 与环境水分发生副反应的可能性。

2.2 主要仪器与装置

微胶囊实验装备（定制，购买自上海澎赞生物科技有限公司）主要由压力控制系统、流动聚焦微流控芯片、恒温水浴、液滴收集与搅拌装置、光学显微镜（M500C 型，购买自深圳市华显光学仪器有限公司）；后处理使用抽滤装置和鼓风干燥箱，采用内径 0.6 mm PTFE 的连接管将原料管和流动聚焦微流控芯片连接(图 2)。液滴动态形成过程采用 M500C 型光学显微镜及配套图像采集系统进行连续记录。正式采集前，在与实验观察相同的物镜倍率、相机分辨率和图像裁剪范围下完成空间标定。以 100 μm 的液滴为例，根据软件界面记录，图像采集帧率为 9.5 fps，同时在画面中加入 100 μm 标尺，显微镜的观察倍率为 10 倍。

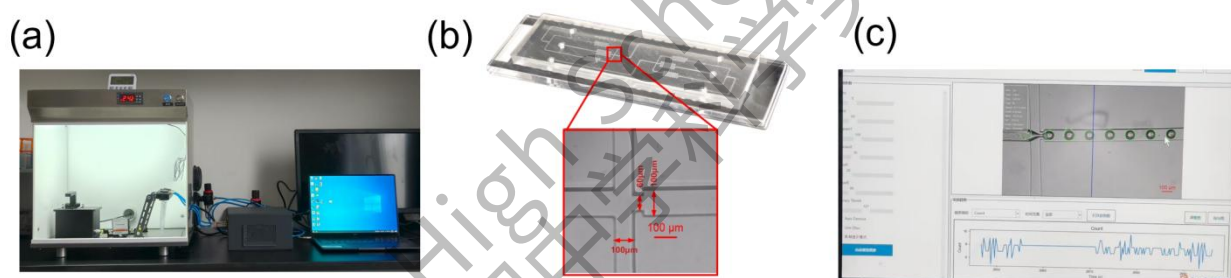


图 2. 实验装置及在线监测

(a) 微流控实验装置；(b) 流动聚焦芯片；(c) 液滴生成动态监测

每组工况开始记录前，先调整光源强度、焦距和曝光参数，使微通道边缘及液滴轮廓清晰可辨。随后固定显微镜、镜头、相机和微流控芯片的位置，在保持相同视野、倍率、分辨率及裁剪比例的条件下进行图像采集。改变两相流量后，待液滴断裂位置、生成周期和下游运动状态基本稳定，再连续采集有效图像。显微图像按帧读取后，依据标定系数将像素尺寸换算为实际尺寸。液滴生成频率根据固定观察区域内单位时间通过的有效液滴数量计算。

2.3 实验设计及步骤

本研究以聚脲-十八烷核壳体系作为研究对象，重点关注微胶囊成球、界面成壳及壳层限域对相变行为的内在规律（如图 3），围绕成球、成壳和限域相变设置三组相互衔接的变量。第一组改变分散相与连续相流动条件，以分散相毛细管数 Ca_d 表征黏性作用与界面

张力的竞争，研究相变微胶囊前驱液滴的成球过程；第二组改变 OD/IPDI 质量比，分析壳材前驱体原料配比对泄漏、壳层完整性及聚脲特征峰的影响；第三组比较不同核壳组成以及不同尺度样品的晶型和热行为，探究相对壳层含量和颗粒尺度所产生的空间限域影响。

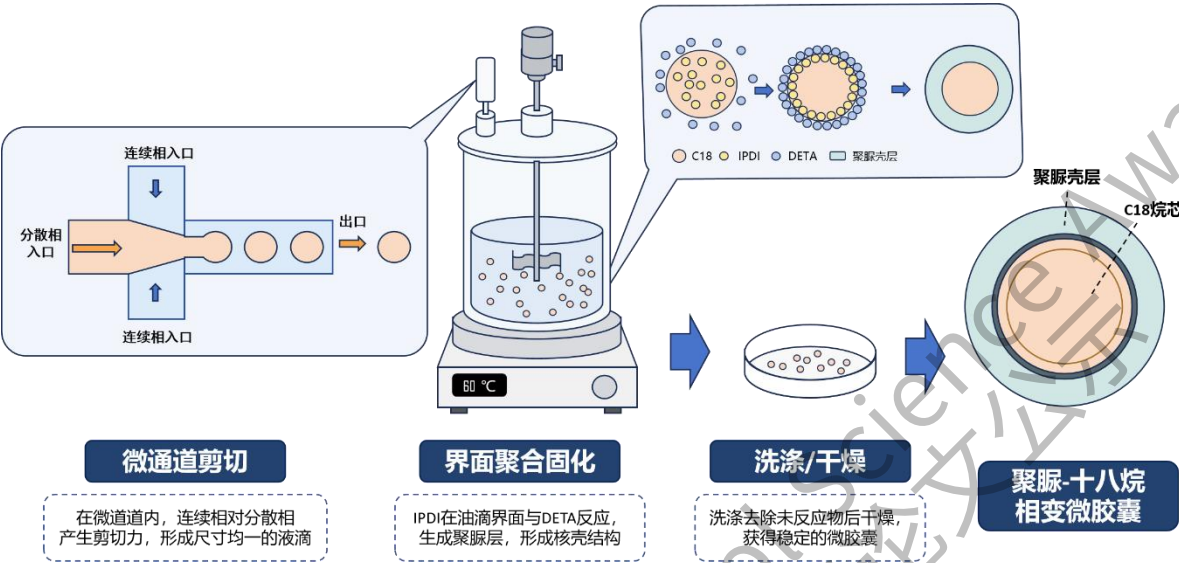


图 3. 聚脲-十八烷相变微胶囊制备流程示意图

2.3.1 相变微胶囊前驱液滴成球实验研究

为揭示界面受力对相变微胶囊前驱液滴稳定成球的调控机制。将 OD 于 40 °C 水浴中熔融，并按设定 OD/IPDI 质量比加入 IPDI，配制分散油相；连续水相由 PVA、SDS 和 DETA 的水溶液组成，其中 DETA 浓度调节至 4 wt%。两相分别通入流动聚焦微通道，通过改变两相流动条件改变黏性/剪切作用与界面张力的相互作用。

实验过程中连续采集液滴生成的显微图像，并采用实验设备配套软件，在线显微图像识别提取液滴轮廓、平均粒径、标准差、CV 值及生成频率等参数，以此和 CFD 模拟仿真预测的界面形变和断裂状态相互验证，从动态实验角度判定稳定成球对应的毛细管数范围。

2.3.2 IPDI/DETA 界面聚合与微胶囊成壳实验

本部分实验旨在阐明 IPDI/DETA 界面聚合形成聚脲壳层的反应机制。微流控生成的前驱液滴连续进入含 DETA 的水相收集体系，水相 DETA 向油-水界面扩散，其氨基与油相 IPDI 中的异氰酸酯基发生加成反应并生成脲键。由于聚脲在油、水两相中的溶解度较低，初生聚合物优先在界面沉积；随着单体持续迁移和反应，局部沉积层逐渐连接并形成连续包覆层，从而把液态 OD 固定在壳层内部。

为研究壳材前驱体配方比例对壳层形成的影响（如图 4），在保持其他条件一致的基础上，设置不同 OD/IPDI 质量比（3:1、5:1 和 7:1 等），比较壳材前驱体相对含量变化所对应的泄漏、黏连、壳层完整性及聚脲特征变化。随后将收集液置于 60 °C 恒温水浴中保

温反应 6 h，并在恒定 60 r/min 转速下、温和搅拌下维持颗粒分散，使界面反应继续进行并完成壳层固化。

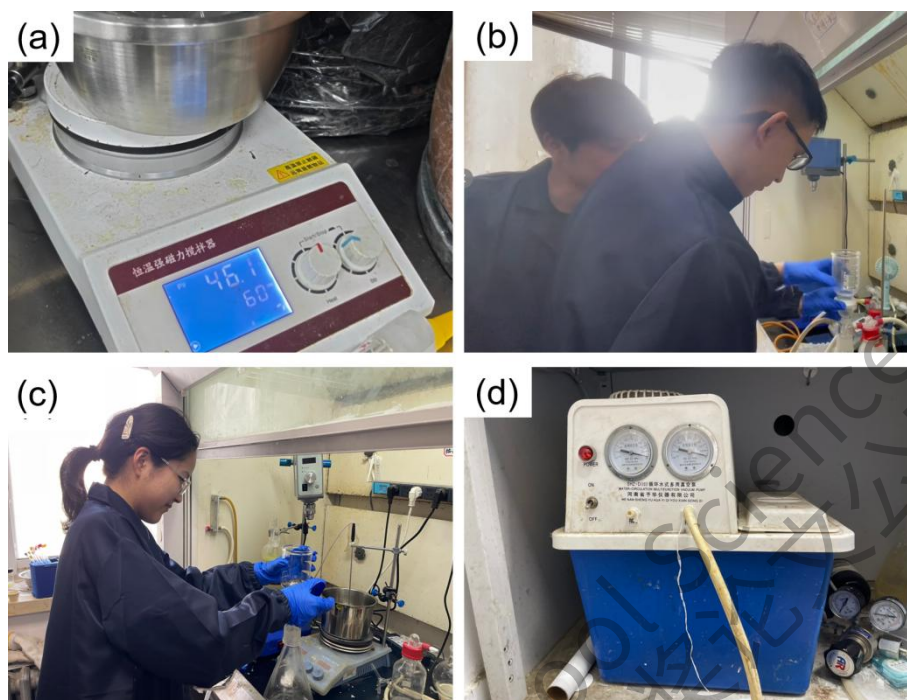


图 4. 固化实验研究

(a) 加热固化升温阶段；(b-c) 抽滤装置组装；(d) 真空抽滤用真空泵

反应完成并冷却至室温后，采用抽滤收集微胶囊。滤饼先以去离子水洗涤以除去水相中的残留物，再以正己烷洗涤以除去颗粒表面未包覆的 OD 和油溶性残留物；去离子水洗与正己烷洗交替进行 3-5 次。洗涤后的样品置于 60 °C 鼓风干燥箱中干燥 12 h，冷却后密封保存，供后续测试研究。

2.3.3 核壳组成与限域相变实验

该部分实验用于分析聚脲壳层限域对十八烷相变行为的调控机制。选取不同 OD/IPDI 质量比以及 100、300 和 500 μm 尺度的微胶囊作为对比对象，并以纯 OD 为基准样品。在芯材化学组成不变的前提下，先通过 FTIR 确认各样品的特征官能团，再通过 XRD 比较特征衍射峰位置及相对强度，判断壳层形成和颗粒尺度变化是否改变 OD 的基本晶型；随后利用 DSC 比较熔融温度、结晶温度、过冷度及相变焓。

2.4 表征方法

2.4.1 微胶囊及其前驱液滴形貌表征

利用 M500C 型光学显微镜记录液滴形成过程，在固定视野和稳定流动阶段采集图像，并结合在线显微图像识别软件获得平均粒径和生成频率。干燥微胶囊采用 S-4700 型扫描

电子显微镜（SEM，日本日立公司）观察整体球形度、粒径一致性、表面连续性、黏连和破损情况，为界面聚合是否形成稳定核-壳颗粒提供形貌证据。SEM 测试前，将充分干燥的微胶囊轻撒于导电胶上，去除未黏附颗粒后进行喷金处理。

2.4.2 傅里叶红外光谱测试

采用傅里叶红外光谱（FTIR，Agilent Technologies Cary630）分析纯 OD 及干燥微胶囊的官能团组成，光谱范围 $4000-500\text{ cm}^{-1}$ 。重点比较 $2950-2850\text{ cm}^{-1}$ 、 1460 cm^{-1} 和 720 cm^{-1} 附近的 OD 烷烃特征吸收，以及 1650 cm^{-1} 和 1550 cm^{-1} 附近的聚脲相关吸收。通过不同 OD/IPDI 条件下峰位是否出现、相对峰强如何变化，判断聚脲壳层是否形成以及壳材相对含量的变化趋势。



图 5. 对样品进行傅里叶红外光谱仪测试

2.4.3 X 射线衍射测试

采用 X 射线衍射（XRD，Bruker D8 ADVANCE）分析纯 OD 与各组干燥微胶囊的晶体结构，扫描范围 $2\theta=5-90^\circ$ 、扫描速率 $5^\circ/\text{min}$ ， $\text{Cu K}\alpha$ ， $\lambda=0.154\text{ nm}$ 。XRD 测试前将干燥样品轻柔研散并均匀铺入样品槽，保持各组样品铺展状态一致，比较 $2\theta=10^\circ-30^\circ$ 范围内 OD 特征衍射峰的位置、峰形及相对强度。峰位是否偏移用于判断界面聚合和空间限域是否诱导新的晶型变化，相对强度变化则作为结晶程度及芯材相对含量变化的辅助证据。

2.4.4 差示扫描量热仪测试

采用差示扫描量热法（DSC，德国耐驰公司）表征纯 OD 及微胶囊的结晶/熔融行为，测试温区为 $-10-70^\circ\text{C}$ ，升温和降温速率均为 $10^\circ\text{C}/\text{min}$ ，并在 N_2 保护下测试。由升温曲线获得熔融峰温及熔融焓，由降温曲线获得结晶峰温及结晶焓；相变焓通过相应吸、放热峰面积积分得到，过冷度按熔融峰温与结晶峰温之差计算。不同样品采用一致的温度程序和数

据处理方法，以便比较 OD/IPDI 质量比及粒径变化对相变温度和储热能力的影响。

DSC 测试时称取 5-10 mg 样品置于铝坩埚中密封，以空坩埚作为参比；氮气流量为：40 mL/min。各样品的积分基线、起止温度和峰温读取方法保持一致，并保留原始热流曲线。热循环稳定性实验采用相同的升降温边界对样品进行 100 次冷热循环，循环前后分别比较 DSC 峰位、峰形和相变焓。

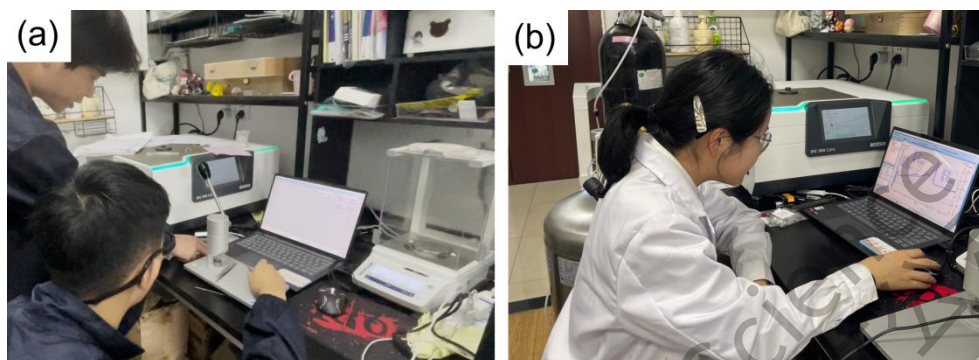


图 6. 对样品进行 DSC 热性能测试

3. 结果与讨论

3.1 相变微胶囊前驱液滴的界面成球机理

受限微通道中的液滴成球并非简单的流速优化，而是液滴界面在黏性/剪切作用与界面张力共同作用下发生形变、颈缩和断裂的过程。本节采用 CFD 仿真分析方法，研究不同毛细管数下的界面形态演变，并结合实验结果分析界面受力与液滴成球状态之间的关系。

3.1.1 仿真方法与界面受力参数

本文采用流动聚焦微通道研究聚脲-十八烷前驱液滴的形成过程。如图 7 所示，油相为分散相，水相为连续相，连续相从上下两侧对分散相进行挤压和剪切；通道宽度和深度均为 240 μm 。

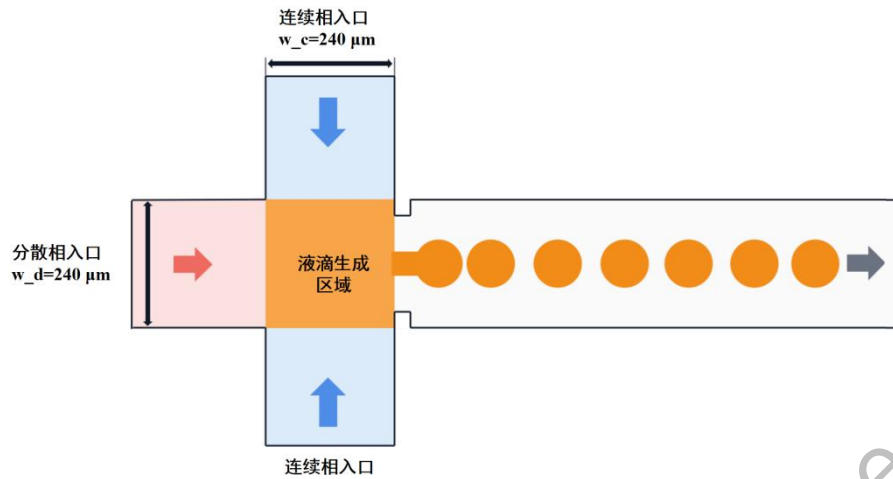


图 7. 微流控通道成球过程示意图

采用 COMSOL 瞬态两相流模型，并通过 VOF 方法追踪油-水界面的动态变化。模型采用经典质量与动量守恒方程，假设两相不可压缩、互不相溶且处于层流状态，忽略重力影响；两相物性参数见表 1。仿真的重点在于比较不同界面受力条件下液滴形态的演变，而非对经典流体力学模型本身进行修正。

为表征分散相黏性作用与界面张力之间的相对关系，采用分散相毛细管数 Ca_d 作为关键无量纲参数：

$$Ca_d = \frac{\mu_d v_d}{\sigma} \quad (1)$$

式中， μ_d 为分散相动力黏度， v_d 为分散相流速， σ 为油-水界面张力。

Ca_d 反映黏性作用相对于界面张力的强弱，其变化可用于判断液滴界面由稳定恢复向持续变形、拖尾和射流转变的趋势。

3.1.2 仿真结果与分析

在固定连续相流速 $v_c = 1.0 \times 10^{-2} \text{ m/s}$ 的条件下改变分散相流速 v_d ，使 Ca_d 随之变化；两支路截面积相同时，流量比 q 可近似表示为 v_d/v_c 。材料参数见表 1，不同 Ca_d 对应的液滴形态见表 2 和图 8。

表 1 两相材料模拟仿真参数

项目	分散相（油相）	连续相（水相）
密度 $\rho/(\text{kg} \cdot \text{m}^{-3})$	890	1000
动力粘度 $\mu/(\text{Pa} \cdot \text{s})$	0.117	0.072
界面张力参数 $\sigma/(\text{N} \cdot \text{m}^{-1})$	0.02	0.02

在连续相条件保持不变时，分散相流速及 Ca_d 的增加会改变液滴界面受力平衡，使液滴由周期性断裂逐渐向拖尾和射流状态转变。

表 2. 固定连续相流速下的液滴成型参数及状态

v_c (m/s)	v_d (m/s)	Ca_d	q	液滴成型状态
0.01	0.002	0.0072	0.2	液滴较小，形成周期较长，下游液滴数量少
	0.004	0.0144	0.4	稳定成球，液滴大小及间距较均匀
	0.006	0.0216	0.6	液舌明显延长，断裂位置向下游移动并出现拖尾
	0.008	0.0288	0.8	拖尾更加显著，出现液滴粘连、局部融合现象
	0.010	0.036	1.0	形成连续射流，观察时间内未发生完整断裂

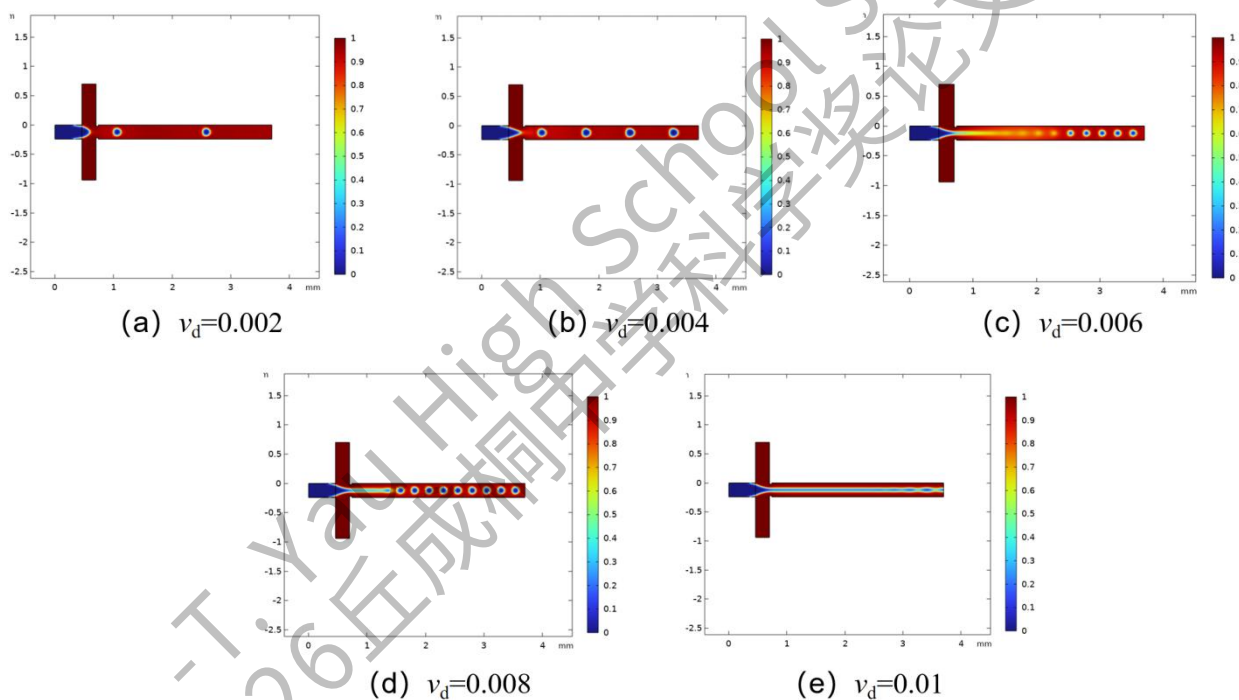


图 8. 分散相流速对液滴成球的仿真影响（图中单位为 m/s）

当 $Ca_d=0.0072$ 时，界面张力相对占优，液滴能够断裂，但分散相补充较慢，形成周期较长；当 Ca_d 增至 0.0144 时，黏性/剪切作用与界面张力达到较协调的动态平衡，液滴周期性经历“膨胀-颈缩-断裂-回缩”，形成较规则的液滴；当 Ca_d 进一步增至 0.0216 及以上时，分散相持续推进使界面变形增强，颈缩位置向下游移动，并逐步出现拖尾和射流。由此可见，液滴形态转变来源于界面变形驱动力与界面恢复力之间的竞争，而不是流速单一增加带来的工艺改善。图 9 进一步概括了上述受力机制。在本体系中， $Ca_d=0.0072\sim0.0216$ 范

围内液滴仍可完成断裂成球，其中适中的 Ca_d 更有利于形成规则、周期性的液滴；当 Ca_d 继续增大时，界面张力难以及时恢复液滴形态，体系逐渐向拖尾和射流状态转变。

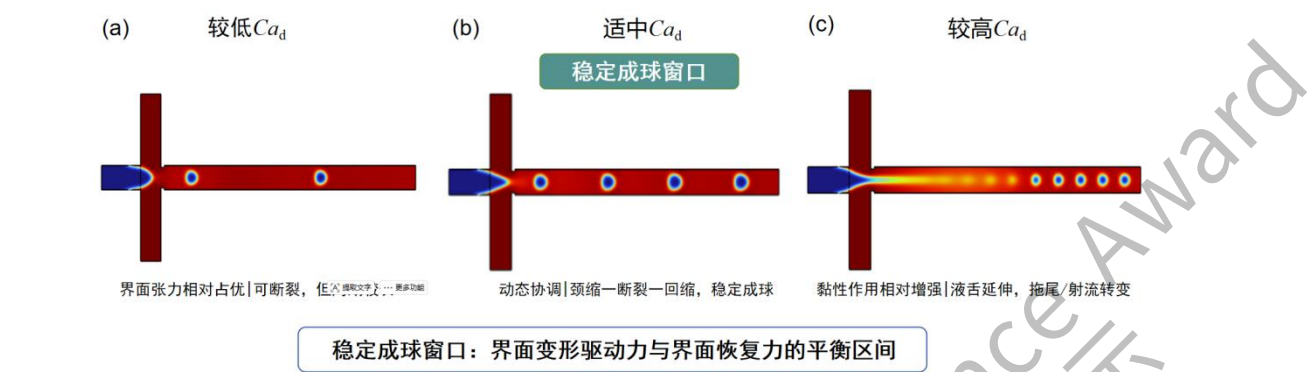


图 9. 受限微通道中界面受力竞争驱动的液滴成球机理示意图

3.2 在线显微图像识别辅助的液滴成球行为实验表征

为检验 CFD 所揭示的“界面受力-液滴形态”关系，本文采用在线显微图像识别对微流控显微图像进行定量分析。采用实验设备配备的显微图像在线识别软件提取液滴粒径、CV 值和生成频率，用于判断液滴是否处于规则、周期性的断裂成球状态，识别结果见图 10。随着分散相流速提高，平均粒径由 $77.6\text{ }\mu\text{m}$ 增至 $136\text{ }\mu\text{m}$ 和 $194.9\text{ }\mu\text{m}$ ，同时液滴生成频率增加，说明分散相供给增强后液滴在断裂前经历更明显的界面扩张，与 CFD 中观察到的液舌延伸趋势一致。

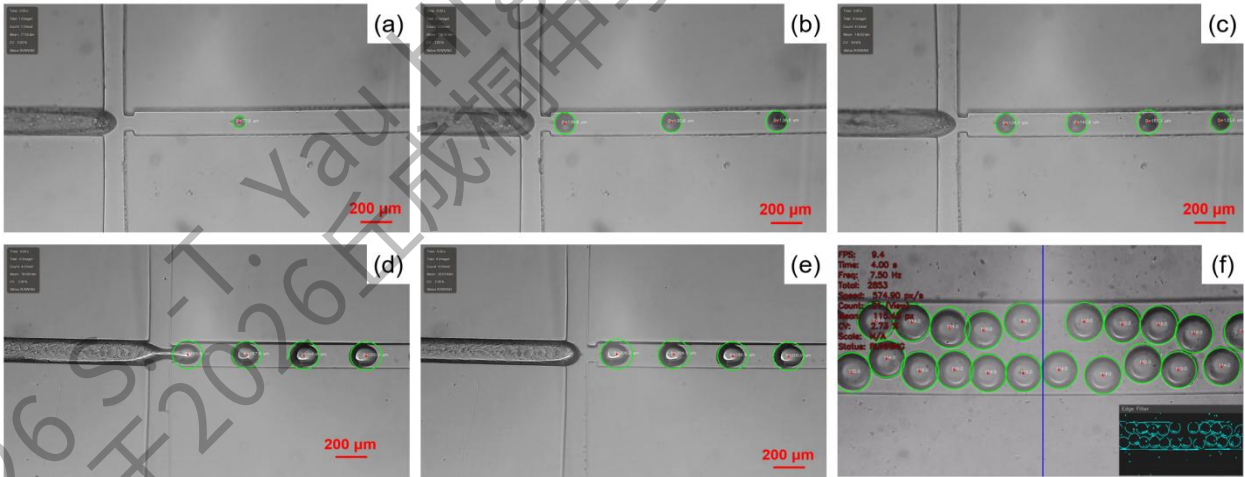


图 10. 在线显微图像识别微球成型结果展示

进一步选取连续相流速 0.01 m/s 、分散相流速 0.005 m/s 进行实验与模拟对比（图 11）。显微图像在线识别得到液滴平均粒径 $196.6\text{ }\mu\text{m}$ 、CV 值 1.88% ，实验中液滴能够持续、规则生成；相同条件下的模拟同样表现出周期性断裂。两者的一致性为界面受力控制液滴成

球状态的分析提供了动态实验佐证，而不是单纯用于筛选最优工艺参数。

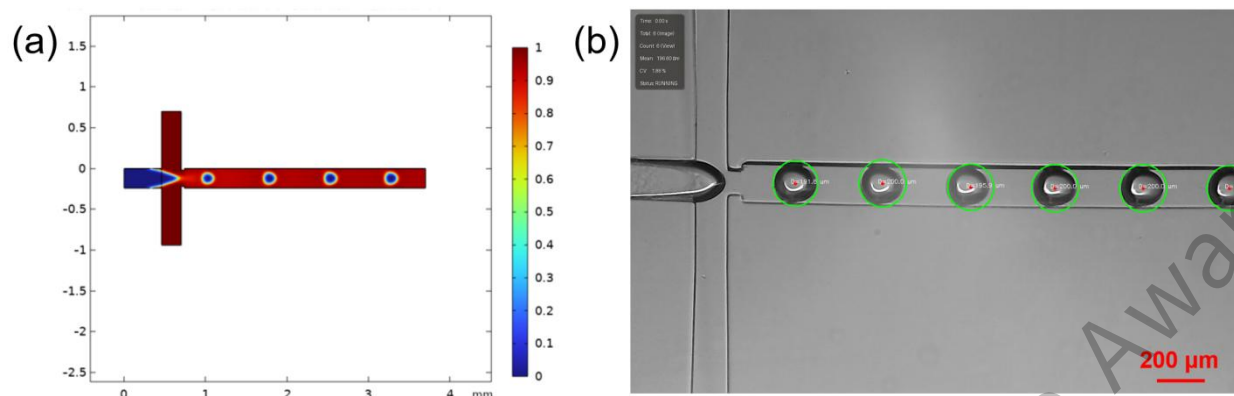


图 11. 微球成型仿真结果与实验验证

3.3 微胶囊的界面聚合与结构表征

为分析 IPDI/DETA 对比对聚脲壳层形成及微胶囊形貌的影响，对不同配比下制备的微胶囊进行 SEM 表征，结果如图 12 所示。随着 IPDI 相对用量增加，微胶囊的成球状态逐渐改善：如图 12（a）所示 IPDI 用量较低时，样品中出现较多不规则颗粒、破损结构和黏连团聚，部分颗粒表面存在塌陷或包覆不完整现象；如图 12（b）所示，适当提高 IPDI 比例后，具有完整球形轮廓的微胶囊明显增多，颗粒间的黏连有所减轻，但仍可观察到少量形貌不规则的微胶囊；如图 12（c）所示，当 IPDI 用量进一步增加至适宜范围时，大部分微胶囊呈现较规则的球形，表面包覆较为连续，颗粒边界清晰，破损和团聚现象明显减少。该变化表明，增加油相中的 IPDI 含量能够为界面聚合提供更多-NCO 基团，使其与水相中的 DETA 充分反应，从而促进聚脲在油-水界面持续沉积，形成较为连续、完整的壳层。

当 IPDI 相对不足时，参与界面成壳反应的-NCO 基团数量减少，生成的聚脲难以完全包覆 OD 液滴，进而形成较薄或局部不连续的壳层，导致熔融 OD 从壳层缺陷处渗出，如图 12（d）所示。泄漏的 OD 会使微胶囊表面发黏，并在后续固化、过滤和干燥过程中引起颗粒黏连、桥联和结块，这与图 12（e-f）观察到的不规则颗粒及团聚现象相对应。实验结果表明，当 DETA 浓度保持为 4 wt%、OD/IPDI 质量比不高于 7: 1 时，所得微胶囊整体球形度和分散性较好，壳层表面较为连续，未观察到明显的芯材泄漏；当 OD/IPDI 质量比进一步增大时，单位质量 OD 所对应的 IPDI 用量减少，导致壳材前驱体相对不足，微胶囊的壳层完整性下降，芯材泄漏、黏连和结块的风险增加。SEM 形貌与样品宏观状态的变化基本一致，共同说明适当提高 IPDI 比例有利于聚脲壳层的形成和微胶囊结构稳定性的提高。

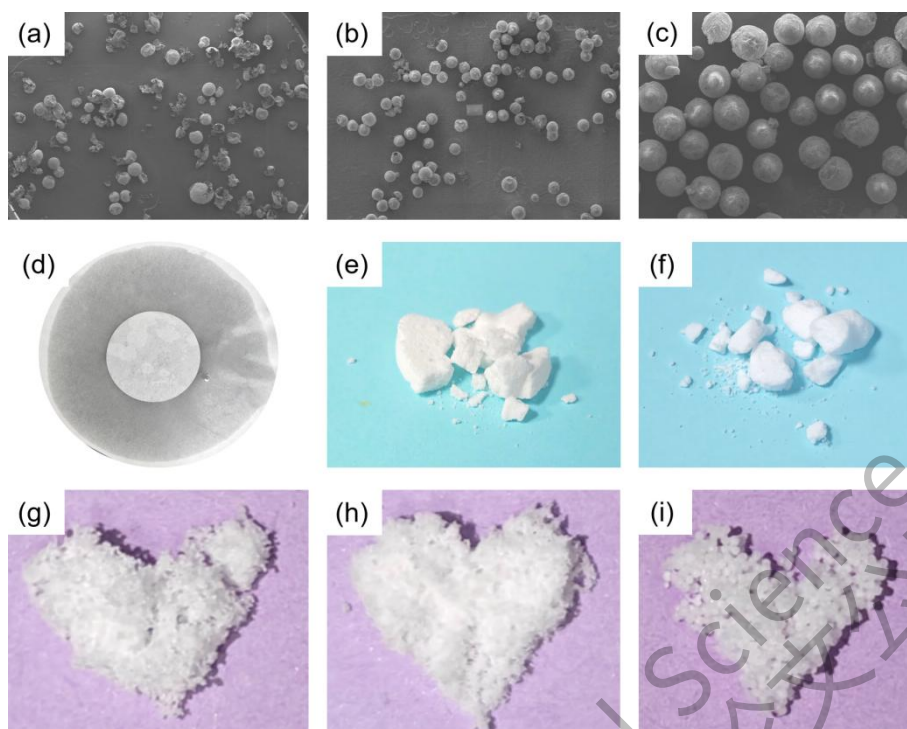


图 12. 聚脲-十八烷相变微胶囊形貌

(a-c) 不同 IPDI 相对用量下聚脲-十八烷相变微胶囊的 SEM 形貌；(d-f) IPDI/DETA 质量比不适宜条件下出现芯材泄漏、颗粒黏连及结块的微胶囊宏观形貌；(g-i) 壳层完整、无明显泄漏的 100、300 和 500 μm 正十八烷@聚脲相变微胶囊宏观形貌

水相 DETA 扩散至油-水界面后，其氨基与油相 IPDI 的异氰酸酯基发生加成反应，生成脲键；生成的聚脲溶解性较低，因而优先在油-水界面沉积。该界面聚合反应路径与 Yang 等报道的 IPDI-DETA 界面聚合机制一致^[12]。结合本研究微胶囊的核-壳形貌，可以推断随着反应持续，界面聚脲由局部沉积逐渐演化为连续壳层，最终将液态油滴固定为稳定的核-壳结构，其形成过程如图 13 所示。

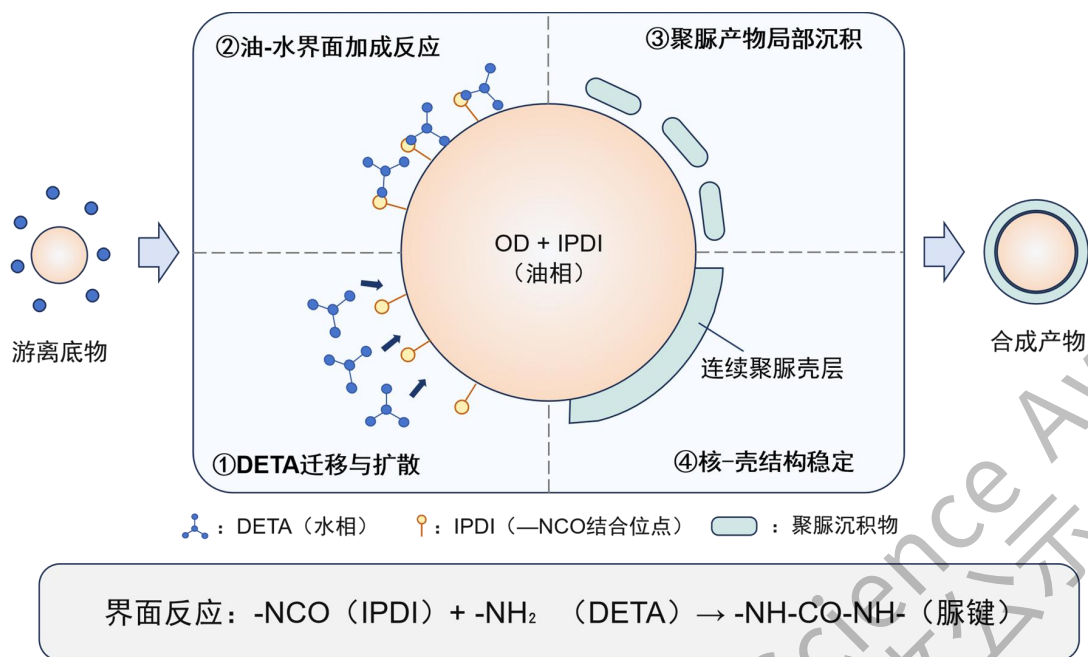


图 13. IPDI/DETA 在油-水界面的聚脲成壳机理示意图

为探究不同 OD/IPDI 配比聚脲-十八烷相变微胶囊的化学组成、界面反应行为与芯材结晶特性。对样品进行了傅里叶红外光谱 (FTIR) 与 X 射线衍射 (XRD) 表征, 结果如图 14 所示。

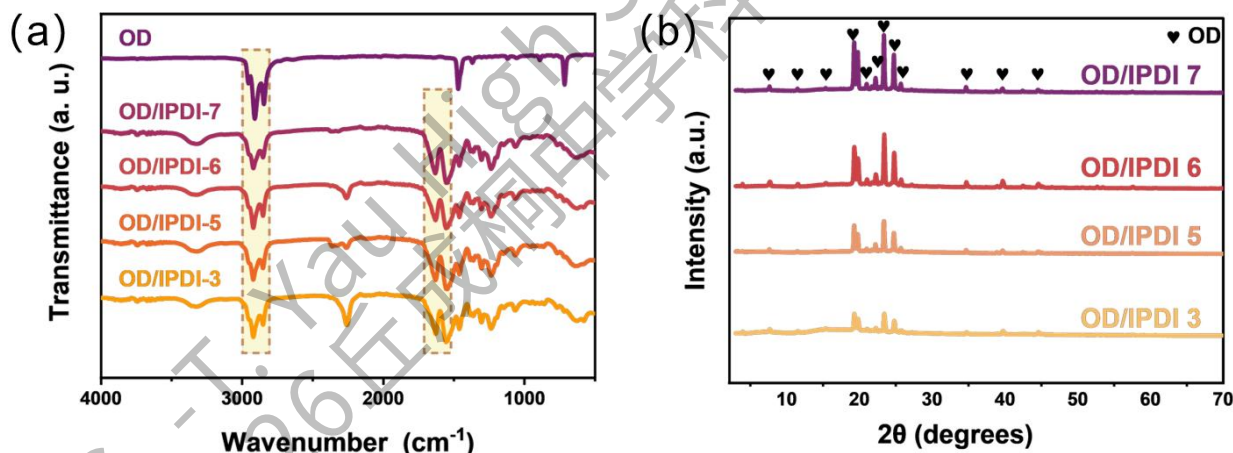


图 14. 不同 OD/IPDI 配比聚脲-十八烷微胶囊的化学结构与晶体结构表征

(a) 不同核壳质量比下 100 μm 级微胶囊的傅里叶红外光谱; (b) 不同核壳质量比下 100 μm 级微胶囊的 X 射线衍射图谱

图 14 (a) 为不同 OD/IPDI 核壳配比的微胶囊红外光谱 (FTIR) 图, 由图可知分别在 $2950\text{-}2850\text{ cm}^{-1}$ ($\text{-CH}_2\text{-CH}_3$ 的不对称与对称伸缩振动)、 1460 cm^{-1} (-CH_2 弯曲振动) 及 720 cm^{-1} (长链烷烃面内摇摆振动) 处, 出现明显的 OD 特征峰^[13]; 随着体系中 IPDI 比例提高, 1650 cm^{-1} (脲基 C=O 伸缩振动) 与 1550 cm^{-1} (N-H 弯曲振动) 处的聚脲特征峰逐

渐增强^[14]，证明成功制备生成了聚脲结构。与此同时，OD 特征峰的相对强度减弱，与芯材质量分数降低、聚脲壳层相对含量增加的变化趋势一致。图 14（b）为不同 OD/IPDI 配比微胶囊的 XRD 图谱。纯 OD 在 2θ 为 10° - 30° 的范围内呈现明显的特征衍射峰。不同配比的微胶囊样品的 OD 特征衍射峰位置未发生明显偏移，说明聚脲包覆未改变 OD 的基本晶型，芯材仍能保持其结晶与相变能力。

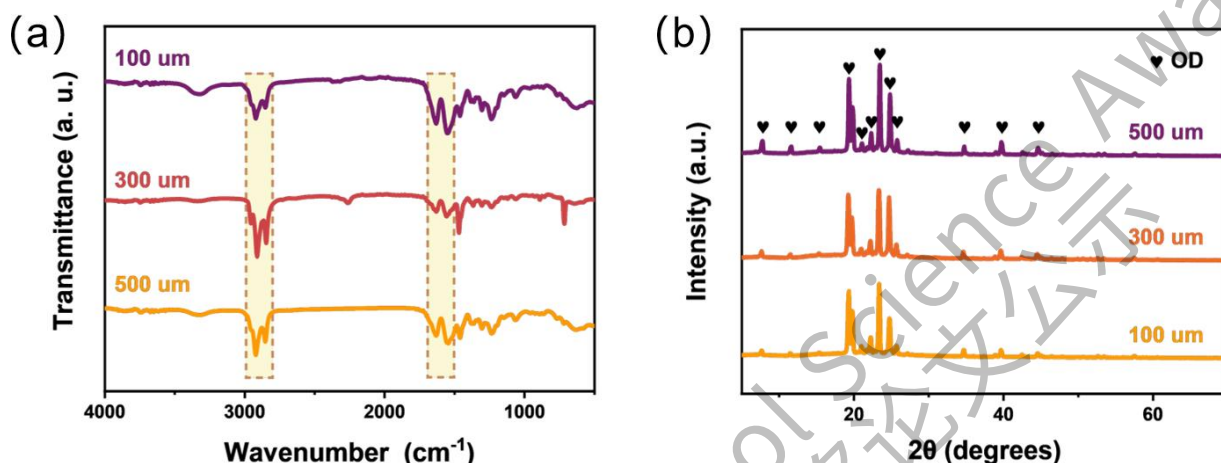


图 15. 不同粒径的聚脲-十八烷微胶囊的化学结构与晶体结构表征

(a) 100、300 和 500 μm 级微胶囊的傅里叶红外光谱；(b) 100、300 和 500 μm 级微胶囊的 X 射线衍射图谱

图 15（a）为粒径为 100、300、500 μm 微胶囊 FTIR 光谱。所有样品均同时呈现 OD 的烷烃特征峰与聚脲的特征吸收峰，且主要峰位基本一致，表明改变微胶囊粒径未引起其主要化学组成和官能团结构的明显变化，各粒径条件下均能发生 IPDI/DETA 界面聚合反应。图 15（b）为不同粒径微胶囊的 XRD 图谱，不同粒径样品均呈现与纯 OD 完全匹配的特征衍射峰，表明不同粒径微胶囊中的 OD 均保持原有晶体结构。较大粒径样品的 OD 特征衍射峰相对较强，说明其结晶有序程度可能有所提高。这可能是由于微胶囊粒径增大后，单位体积芯材受到的壳层空间限域作用相对减弱，从而有利于 OD 分子的有序排列与结晶。

较大粒径胶囊中 OD 受聚脲壳层的空间约束作用更弱，结晶度略有提升，为微胶囊的相变储热性能提供了保障。从构效关系看，IPDI 比例增加首先表现为聚脲特征峰增强和壳层相对含量提高，而 OD 晶型峰位基本保持不变，说明核壳组成变化主要改变的是界面与限域环境，而非诱导十八烷发生新的晶型转变；这为后续从壳层限域角度解释相变温度、过冷度和焓值变化提供了结构依据。

对成功制备的不同尺度的相变微胶囊进行了形貌与结构表征。图 16（a-c）的光学显微图像表明，不同粒径的微胶囊均呈规整球形，粒径均匀，无明显团聚、粘连或破碎现象，

证明微通道乳化结合界面聚合的制备工艺可实现微胶囊尺寸的精准调控。图 16 (d-f) 的扫描电子显微镜图像进一步证实了不同粒径的微胶囊表面均呈现致密的表面形貌，无明显裂纹、针孔或壳层缺陷，表明界面聚合反应充分，形成了完整、连续的聚脲包覆层，可有效维持微胶囊结构。因此，完整连续的聚脲壳层不仅是界面聚合形成核-壳结构的形貌证据，也是后续产生稳定空间限域、限制液态芯材迁移并维持可逆相变的结构前提。

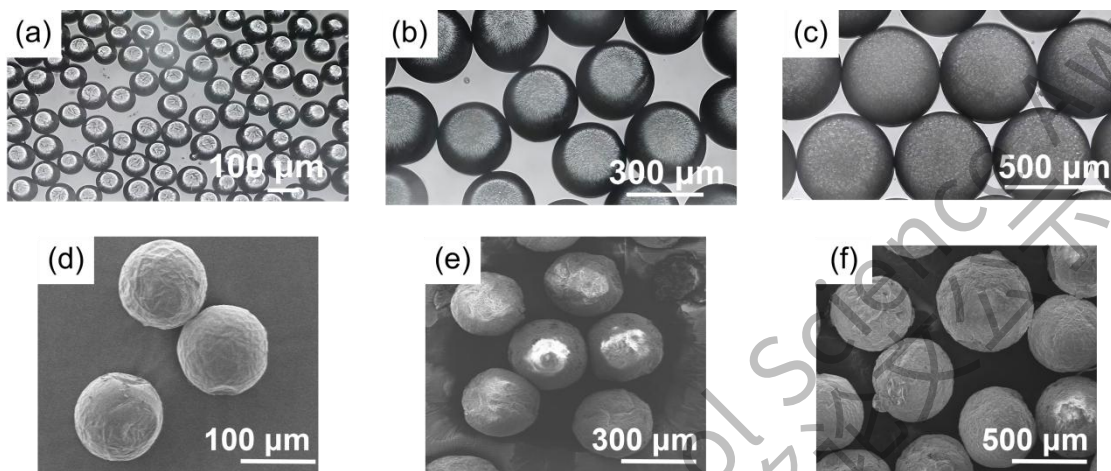


图 16.不同尺度的微胶囊图像

(a-c) 光学显微镜图像；(e-f) 不同尺度的微胶囊扫描电子显微镜图像

3.4 微胶囊相变行为与壳层限域作用

为探究相变微胶囊相变储热性能，对样品进行了差示扫描量热(DSC)表征(如图 17)。DSC 升温曲线(图 17a)与降温曲线(图 17b)显示，所有配比的微胶囊样品在升温过程中均呈现明显的吸热峰，降温过程中出现对称的放热峰，表明聚脲壳层的包覆未抑制其结晶-熔融行为，具备相变储热功能。微胶囊的熔化温度 T_m 整体分布在 32.3-34.1 °C 区间(如图 17c)，略高于纯十八烷的熔点(28.2 °C)。结晶温度 T_c 随 OD 配比升高呈单调上升趋势，从 3:1 的 17.8 °C 升至 7:1 的 20.8 °C，对应的过冷度从 16.0 °C 降至 11.5 °C，表明芯材占比越高，过冷度越小，这是因为高芯材含量下，十八烷分子受聚脲壳层的空间约束减弱，结晶成核的位阻降低[15]，成核过程更易发生，更利于实际储热应用。图 17d 表明，随着 OD/IPDI 配比提高，微胶囊的熔化焓 ΔH_m 从 151.1 J/g 显著提升至 202.2 J/g，凝固焓 ΔH_c 从 151.1 J/g 增加至 203 J/g，证明相变过程可逆性良好，无明显热损失或不可逆结构变化。焓值随芯材占比升高而增大，与相变焓和芯材质量分数的理论正比关系一致，说明通过调整 OD/IPDI 配比可有效调控微胶囊的储热密度。

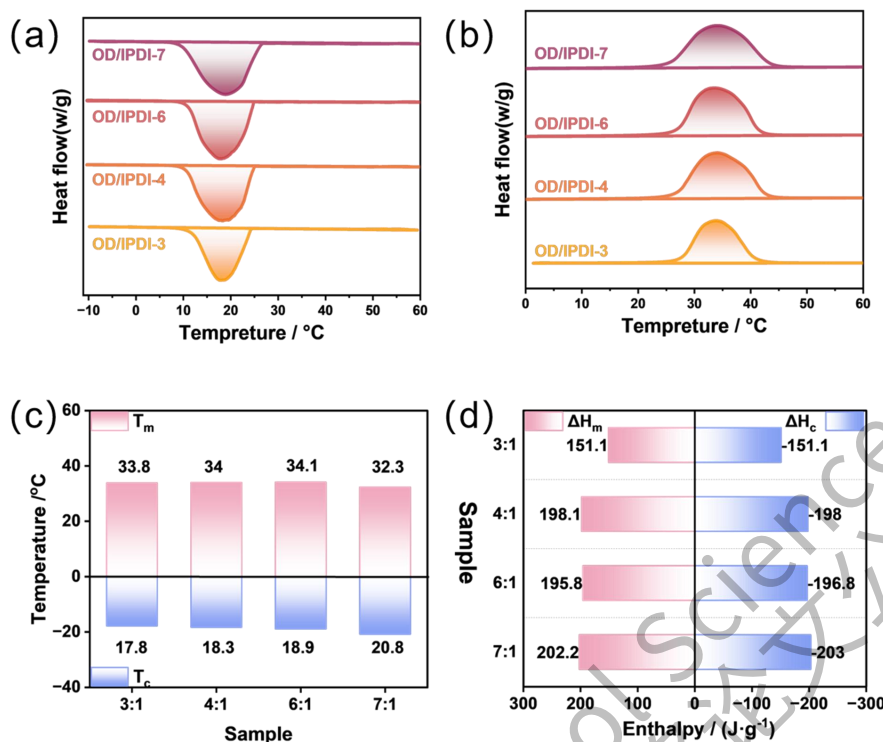


图 17 不同核壳比的 100μm 微胶囊热性能

(a) 差示扫描量热升温熔融曲线；(b) 差示扫描量热降温结晶曲线；

(c) 熔化温度及结晶温度；(d) 熔融焓与结晶焓

图 18 进一步揭示了壳层限域条件下十八烷的可逆相变过程。升温时，十八烷晶体的有序结构逐渐破坏，并在熔融过程中吸收潜热；熔融后的十八烷仍被封装于聚脲微腔内。降温时，十八烷分子内重新排列，并经历成核与晶体生长过程，同时释放潜热。结合不同 OD/IPDI 配比的 DSC 结果可知，壳层相对占比提高时空间限域增强，结晶重排受到更明显约束；OD 比例提高后限域相对减弱，结晶温度升高、过冷度降低。由此形成核壳组成、限域程度、结晶行为与储热响应的构效映射。

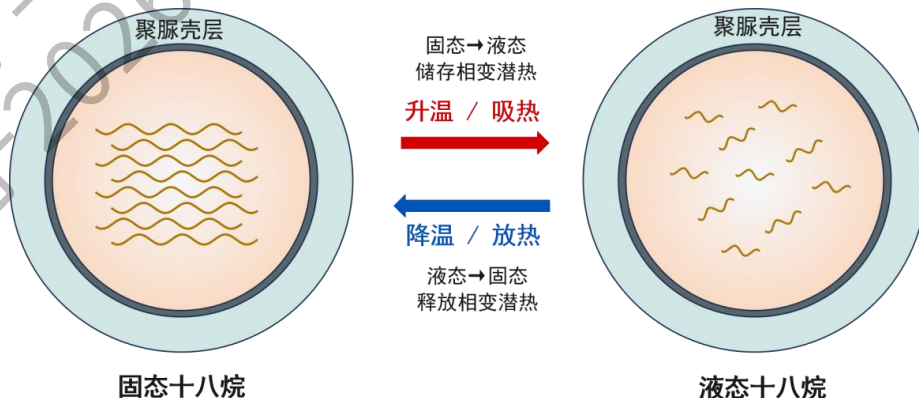


图 18 聚脲限域十八烷的相变吸热/放热与热量调节机理示意图

为评估聚脲-十八烷相变微胶囊的热循环稳定性和储热性能耐久性，对粒径分别为 $100\ \mu\text{m}$ 、 $300\ \mu\text{m}$ 和 $500\ \mu\text{m}$ 的微胶囊进行了 100 次冷热循环测试。由图 19 可知，循环后三种粒径微胶囊的熔融吸热峰和结晶放热峰仍清晰、完整，峰位未发生明显偏移，熔融温度和结晶温度仅出现较小变化，表明微胶囊的相变过程具有良好的可逆性。同时，相变焓值未出现显著减小，说明其潜热储存能力在循环过程中基本保持稳定。上述结果表明，聚脲壳层能够承受十八烷反复固-液相变所产生的体积变化，其包覆作用和空间限域效应未发生明显弱化。DSC 曲线在循环前后保持稳定，进一步说明微胶囊核壳结构稳定性与相变储热性能的稳定之间具有对应关系。

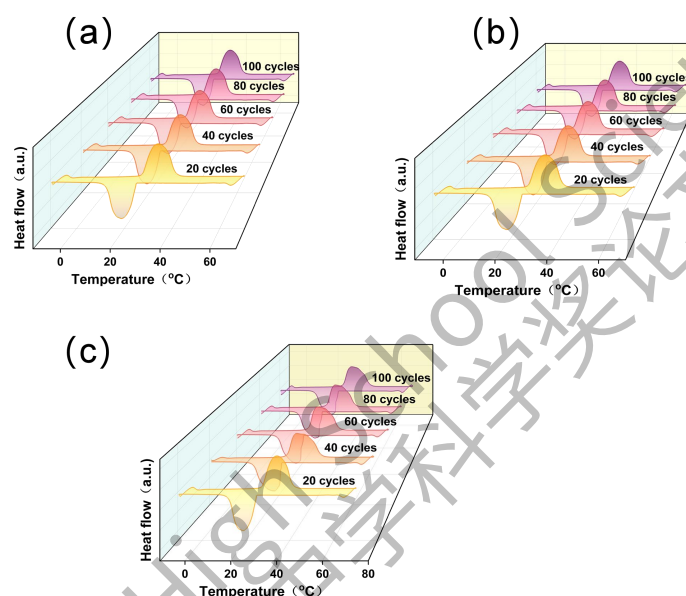


图 19. 相变微胶囊不同热循环次数下的差示扫描量热相变曲线

(a) $100\ \mu\text{m}$ 级； (b) $300\ \mu\text{m}$ 级； (c) $500\ \mu\text{m}$

4. 结论与展望

4.1 结论

针对电子终端对尺寸可控的相变微胶囊提升热管理能效的迫切需求，本研究采用微流控界面聚合技术制备了尺寸精准可控的聚脲-十八烷相变微胶囊，系统探究了液滴前驱体稳定成球、油-水界面聚合成壳、壳层限域调控芯材相变行为三个连续环节的内在规律，揭示了相变微胶囊形成与储热过程的界面作用机制，主要结论如下：

(1) 在受限微通道中毛细管数是调控微胶囊前驱体成球行为的核心无量纲参数。当分散相毛细管数 Ca_d 处于 $0.0072\sim 0.0216$ 区间时，可生成尺寸均一、球形度良好的稳定液

滴，当液滴平均粒径为 $196.6\mu\text{m}$ 时，CV 值仅为 1.88%；

(2) 微胶囊聚脲壳层形成过程如下：水相 DETA 向油-水界面迁移，并与油相 IPDI 在界面处发生加成聚合反应，生成的聚脲在界面持续富集和沉积，逐步形成连续致密的聚合物壳层；

(3) 微胶囊聚脲核壳反应单体配比变化改变了壳层对相变芯材的限域效应，进而调控其结晶过程与相变焓值；随着 OD/IPDI 质量比由 3:1 提升至 7:1，芯材十八烷的结晶温度升高，过冷度显著降低，相变焓由 151.1J/g 提升至 202.2J/g ；经过 100 次冷热循环后，微胶囊的 DSC 峰形与相变焓仍保持稳定，表明其具有良好的循环稳定性。

本文建立了相变微胶囊的微流控界面聚合制备方法，实现了界面聚合反应由传统无序、随机状态向分散、可控态的关键转变，从反应层面突破了聚脲壳层结构难以精准调控的共性难题，为通过界面聚合化学设计主动调控相变微胶囊的储热性能提供了实验基础，对高性能相变储能微胶囊的可控制备开辟了新途径。

4.2 展望

(1) 微流控制备相变微胶囊具有精确可控、性能稳定的优势，未来可结合 AI 图像识别方法优化成型工艺，发展多通道并行制备体系，实现大规模批量精准制造。

(2) 依托微流控方法制备的相变微胶囊尺寸均一可控的独特优势，除了人工智能芯片与 5G/6G 设备散热应用外，在新能源汽车及动力电池热安全、光伏与储能系统温度调节、低能耗建筑和智能纺织品等领域具有广阔应用前景；结合绿色壳材技术的发展，其可通过高效储放热降低温控能耗、减少碳排放，助力实现碳达峰、碳中和目标，成为新一代低碳热管理材料。

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致谢

本课题由八十中学国际部冯甄钰、艾星佑共同申报、独立完成，研究周期为 2025 年 11 月至 2026 年 8 月，历时 10 个月。

课题始于冯甄钰的亲身经历：奶奶瘫痪卧床无法言语，发烧时常难以及时察觉，这份牵挂让她在初中就思考：能否做一种不用电、一眼就能看出发烧的材料？后来在北京化工大学潘凯教授组会中，她首次知道相变微胶囊技术，开始研究，并成功制备可用于发烧预警的热敏相变微胶囊，项目荣获北京市青少年科技创新大赛一等奖、全球发明大会中国区总决赛一等奖。但她没有停止研究，琢磨怎么突破传统制备工艺的局限，并将其用到更多地方。在王珩老师课上讲到移动终端芯片局部过热的现实问题——手机发烫、笔记本电脑过热等，让她打开了思路：相变材料能否给芯片降温、缓解局部过热？基于此，她邀请化学社社长艾星佑一起将研究逐步推进到界面聚合精准调控与壳层限域相变机制的更深层面。

王珩是冯甄钰的 AP 化学教师、艾星佑的 A-Level 化学教师，也是我们的年级组长。选题时她结合两校实验室条件，与我们共同讨论方向可行性；确定方向后，我们加入她的荣誉导师工作室，利用课余时间系统学习实验操作、实验设计与数据处理方法；论文撰写阶段，她通过 IB 学术论文写作课程给予我们具体修改意见与方向引导。整个研究周期她全程跟进，在我们遇到瓶颈时耐心引导思路。北京化工大学潘凯教授为课题提供深层次学术引领，从选题论证、机理梳理到实验设计、论文架构均给予系统性指导；正是与他深入讨论后，我们最终确立了微流控界面聚合的核心技术路线。

实验期间，我们常与北京化工大学实验室的陈轲老师研讨聚脲-十八烷微胶囊的制备方案；热性能、循环稳定性等核心专业知识，并通过文献检索自主学习实践。基于实验室的安全管理规定，李一学长每次都等我们完成实验、整理清洗器材后才一起离开。电子显微镜、傅里叶红外光谱仪、X 射线衍射仪、差示扫描量热仪等各类大型仪器的操作与测量方法，都由他亲手示范讲解。百次冷热循环测试阶段，仪器昼夜运行，我们常实验至深夜，他始终耐心陪同。

以上提到的所有人的指导与帮助均为无偿提供。实验操作、原始数据采集记录与论文写作，均由我们独立完成。课题分工明确，我们优势互补、协同推进。冯甄钰负责课题选题与前期调研，完成 CFD 仿真，探究液滴稳定成球的调控规律以及不同 OD/IPDI 配比下的微胶囊成型过程；艾星佑负责基于机器视觉的液滴在线成型验证，以及微胶囊的热性能和循环热稳定性。论文初稿由冯甄钰执笔，艾星佑补充完善，最终两人共同修改定稿。

实验过程我们遇到三个核心难题。实验初期前驱体液滴难以稳定成球，通过查阅大量文献、反复调试流速与界面张力，将分散相毛细管数精准控制在 0.0072–0.0216，实现球形前驱体稳定生成。针对人工计数微胶囊效率低、误差大的问题，我们引入微流控微球实时视觉分析系统，实现粒径数据实时采集与自动统计。针对制备流程繁琐、难以连续稳定产出的问题，我们通过不停优化工艺参数，最终制得平均粒径 196.6 μm 、粒径变异系数 (CV) 仅 1.88% 的单分散微胶囊。

AI 使用情况声明：课题涉及的 AI 相关技术均事先经指导老师确认后使用，所有结果均经人工逐一核验。实验所用微流控微球实时视觉分析系统 (V1.0，软件著作权登记号：2026SR0625514) 由北京化工大学提供，基于 OpenCV 图像处理算法开发，可实时识别液滴与微胶囊的尺寸、形态和数量，自动计算频率、体积分布及 CV 值等指标，属于实验监测工具，非生成式 AI。

最后，衷心感谢北京市中学生英才计划的科研平台，感谢两校各位老师的悉心指引。现在只是开始，未来我们想做的更多。材料层面可优化聚脲壳材结构与组分，精准调控相变性能；工艺层面可结合 AI 图像识别技术优化成型控制，探索多通道批量制备；应用可延伸至低能耗建筑温控、动力电池热管理等领域，助力低碳发展。

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Thesis Title: Synthesis of Phase-Change Microcapsules via
Microfluidic Interfacial Polymerization and Their
Thermal Energy Storage Properties

Synthesis of Phase-Change Microcapsules via Microfluidic Interfacial Polymerization and Their Thermal Energy Storage Properties

Zhenyu Feng, Xingyou Ai

Abstract

Mobile electronic terminals are evolving towards thinner, more highly integrated, and higher computing power devices. With the widespread adoption of artificial intelligence and edge computing power, chip heat flux density continues to rise, and local heat accumulation has become a major challenge to device reliability, service life, and computational performance. Conventional active cooling methods struggle to meet the low-carbon and high-efficiency requirements for next-generation terminal thermal management. Passive thermal storage based on phase-change microcapsules therefore represents a promising strategy for mitigating localized heat accumulation. Phase-change microcapsules with uniform and controllable sizes can effectively suppress temperature peaks, while their heat storage performance depends critically on the stable formation of droplet precursors, formation of the polyurea shell, and shell confinement. However, microcapsules prepared by conventional mechanical stirring methods generally exhibit wide particle size distribution, weak controllability of shell structure, and batch-to-batch variability in heat storage performance. [More importantly, these methods provide limited control over shell architecture at the level of interfacial polymerization.](#)

[In this article, microfluidic interfacial polymerization was employed to prepare n-octadecane@polyurea phase-change microcapsules using isophorone diisocyanate \(IPDI\) and diethylenetriamine \(DETA\) as reactive monomers.](#) Well-dispersed droplets were generated, constructing a highly uniform oil-water reaction interface. The addition polymerization of IPDI and DETA to occur exclusively at a well-defined and controllable liquid-liquid interface. This enabled effective control over the interfacial polymerization process and the resulting polyurea shell structure. By combining computational fluid dynamics (CFD) simulations with real-time microscopic image recognition, this article quantitatively characterized the particle size and distribution of the droplet precursors, focusing on the control mechanisms governing the stable formation of droplet precursors, formation of the polyurea shell, and phase-change behavior of the octadecane core. [The core chemical question is to reveal how the interfacial polymerization reaction of IPDI and DETA at the oil-water interface determined the structure of the polyurea shell.](#)

The results indicated droplet morphology was governed by the balance between viscous and interfacial forces, as characterized by the capillary number. When the dispersed phase capillary number Ca_d ranges from 0.0072 to 0.0216, stable spherical droplet precursors can be formed, providing a uniform reaction template for interfacial polymerization. IPDI and DETA undergo stepwise addition polymerization at the oil-water interface, forming a dense and continuous polyurea shell. The crosslinking stability of the polyurea shell was directly related to the n-octadecane-to-IPDI feed mass ratio (OD/IPDI). As the octadecane (OD)/IPDI ratio increased from 3:1 to 7:1, the crystallization temperature of the octadecane core raised from 17.8 °C to 20.8 °C, and the melting enthalpy increased from 151.1 J/g to 202.2 J/g. Essentially, the change in the feed ratio altered the shell confinement of the octadecane core, thereby regulating the overall phase-change enthalpy of the microcapsules. After 100 heating-cooling cycles, no significant attenuation of the phase-transition peak or enthalpy was observed, indicating excellent structural stability of the polyurea shell and sustained shell confinement of the octadecane core. This article established a controllable preparation method for phase-change microcapsules through microfluidic interfacial polymerization, and revealed the continuous action mechanism from droplet formation to confined phase-change. The findings offer a promising route toward the controlled fabrication of high-performance phase-change energy-storage microcapsules and provide a basis for their potential application in passive thermal management.

Keywords: Microfluidics; Interface Polymerization; Polyurea Microcapsules; Shell Confinement; Phase-Change Behavior

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1. Introduction

With the rapid advancement of mobile communications and artificial intelligence, mobile devices such as smartphones and laptops are becoming thinner, more highly integrated, and more computationally powerful. Meanwhile, the widespread adoption of AI-based edge computing and high-frame-rate rendering has further increased the heat flux density of chips, batteries, and power-management modules [1]. At the same time, the space available for heat dissipation continues to shrink as devices become thinner. As a result, localized heat accumulation has become an important factor limiting device stability, service life, and the sustained utilization of computational performance [2,3].

The current thermal management for terminals relies on active thermal conduction solutions such as fan cooling, metal heat sinks, and heat pipes, which remain essential for managing the overall thermal load of electronic devices. However, because they primarily rely on macroscopic heat conduction or fluid-based heat removal, they are less effective at rapidly suppressing local temperature peaks and homogenizing temperature around highly localized hotspots. Meanwhile, increasingly stringent energy-efficiency requirements and the broader transition toward low-carbon technologies are driving the electronics industry toward lower-power and more energy-efficient operation. Conventional active cooling approaches alone may therefore be insufficient to simultaneously address the demands of precise local temperature regulation and improved system-level energy efficiency. **Therefore, efficient passive thermal storage based on phase-change materials provides a potential complementary strategy for mitigating localized overheating and enhancing the overall energy efficiency of thermal management systems.**

Phase-change materials (PCMs) can absorb and release latent heat through reversible solid–liquid phase transitions within a specific temperature range, enabling thermal energy to be stored and released when needed. They therefore serve as the key thermal storage medium for passive thermal management. When a phase-change core material is encapsulated within a polymer shell to form a phase-change microcapsule, the resulting core–shell structure can spatially confine the core material. This structure can reduce leakage of the liquid phase, improve thermal cycling stability, and retain much of the latent-heat-storage capability of the core material. These characteristics make phase-change microcapsules attractive for localized hotspot control and temperature homogenization in mobile electronic devices [4,5].

The preparation of phase-change microcapsules has gradually evolved from relatively simple batch processes toward methods offering greater control over particle structure and properties. Conventional approaches mainly include stirred emulsion polymerization, in-situ polymerization, and spray drying. Among them, mechanically stirred emulsion polymerization is one of the most widely used approaches. In this process, mechanical shear generates an oil–water dispersion, after which polymerization at the droplet interface forms the shell around the core material [6]. However, the shear field generated by mechanical stirring is spatially nonuniform, often resulting in broad particle-size distributions and variations in droplet sphericity. The thickness, interfacial structure, and compactness of the resulting shell are also difficult to control precisely, which can lead to variations in thermal storage performance between batches and limit the reproducibility required for high-precision thermal management [7]. Other conventional approaches, including spray drying, likewise face challenges such as limited control over particle size and the formation of shell defects. These limitations make it difficult to deliberately tailor microcapsule structure and thermal properties.

Microfluidic technology provides a novel technological approach for the precise and controllable preparation of phase-change microcapsules. Within microscale confined channels, the dynamic balance between fluid shear force and interfacial tension can be used to generate oil–water droplets with highly uniform dimensions and excellent monodispersity. These droplets provide well-defined templates for subsequent interfacial polymerization. At the same time, microfluidic systems offer a useful platform for quantitatively investigating how interfacial tension, fluid viscosity, and shear affect droplet formation and breakup [8].

A number of studies have explored the use of microfluidics to prepare phase-change microcapsules. Hao et al. [9] used a coaxial microfluidic device to achieve continuous preparation of phase-change microcapsules and demonstrated that the droplet size could be continuously adjusted by varying the flow rates of the two phases. Anni et al. [10] further investigated how the flow-rate ratio between the core and shell phases affected the core–shell ratio and the macroscopic thermal storage properties of the resulting microcapsules. These studies have clearly demonstrated the advantages of microfluidics in improving particle-size uniformity and controlling core–shell composition. However, existing research has focused mainly on how to produce microcapsules with uniform dimensions and desirable thermal properties. The processes occurring after droplet formation-particularly interfacial

polymerization and the subsequent influence of the polymer shell on crystallization and melting of the core material—have more often been treated as separate processing or performance issues. As a result, the continuous evolution from a transient fluid interface to a stable polymer-confined interface, and how this evolution ultimately affects phase-change behavior, remains insufficiently understood.

In recent years, artificial intelligence and computer-vision methods have also begun to enter the field of microfluidics. For example, Tang et al. [11] prepared an oil-in-water-in-oil double emulsion using the flow focusing method and established a machine learning prediction model for the core droplet and double emulsion particle size, providing a quantitative basis for controlling encapsulated droplets. However, relatively few studies have used real-time visual analysis to quantitatively characterize the complete sequence of necking, breakup, and spherical stabilization of droplets used for phase-change microcapsule while directly relating these observations to the underlying interfacial forces.

From the perspective of materials chemistry, the thermal storage performance of phase-change microcapsule is not solely determined by the intrinsic characteristics of the core material. Instead, it is the result of the combined effects of key interconnected processes, including droplet interface formation, interfacial polymerization reactions, and shell confinement effects. The interfacial polymerization reactions at the oil-water interface directly determine the chemical structure and microstructure of the microcapsule shell. Furthermore, the shell confinement generated after the formation of the polymer shell further influences core thermal storage parameters such as the phase-change temperature, supercooling degree, and phase-change enthalpy.

Based on these considerations, we regard the n-octadecane@polyurea microcapsule as a system in which the interface evolves continuously from a fluid interface to a reactive interface and finally to a confined material interface. This study therefore focuses on three closely related scientific issues: (1) how the competition between viscous/shear effects and interfacial tension controls the destabilization, fracture, and stable spheroidization of precursor droplets; (2) how aqueous DETA and oily IPDI polymerize at the oil-water interface to form a continuous polyurea shell, and how the core-shell composition affects the integrity of the shell; (3) after the formation of the polyurea shell, how spatial confinement further affects the crystallization temperature, supercooling degree, and phase transition enthalpy of octadecane.

CFD simulations and real-time microscopic image analysis were combined to establish a quantitative correspondence between simulated and experimentally observed droplet-formation behavior. Fourier Transform Infrared Spectroscopy (FTIR), X-ray Diffraction (XRD), and Scanning Electron Microscopy (SEM) were used to characterize the interfacial polymerization process and core-shell structure, while Differential Scanning Calorimetry (DSC) was employed to investigate the relationship between core-shell composition, confinement, and phase-change behavior.

The main contributions of this study are twofold. First, we developed a microfluidic interfacial polymerization approach for the controlled preparation of phase-change microcapsule. By generating monodisperse droplets and thereby establishing a more uniform reaction interface than conventional mechanically stirred emulsions, the method provides improved control over the conditions under which interfacial polymerization takes place, and achieves a key transition from the conventional disordered and random state to a dispersed and quantitatively controllable state of the interfacial polymerization reaction. This creates an opportunity to further investigate and regulate the formation and structure of the polyurea shell. Second, by combining CFD simulations with real-time microscopic image analysis, we quantitatively investigated the formation mechanism of droplet precursors and established a direct connection between simulation and experiment. Together with the subsequent characterization of interfacial polymerization and phase-change behavior, this work provides an experimental basis for using interfacial chemical processes to tune the thermal properties of phase-change microcapsule.

2. Experimental design and research methods

2.1 Experimental Materials

The materials used in this experiment include octadecane (OD), diethylenetriamine (DETA), isophorone diisocyanate (IPDI), polyvinyl alcohol (PVA) and sodium dodecyl sulfate (SDS), all of which are analytical grade (AR) and provided by Shanghai McLean Biochemical Technology Co., Ltd., China; the experimental water is deionized water. This experiment requires the configuration of two liquids, the dispersed oil phase and the continuous water phase, as shown in Figure 1.

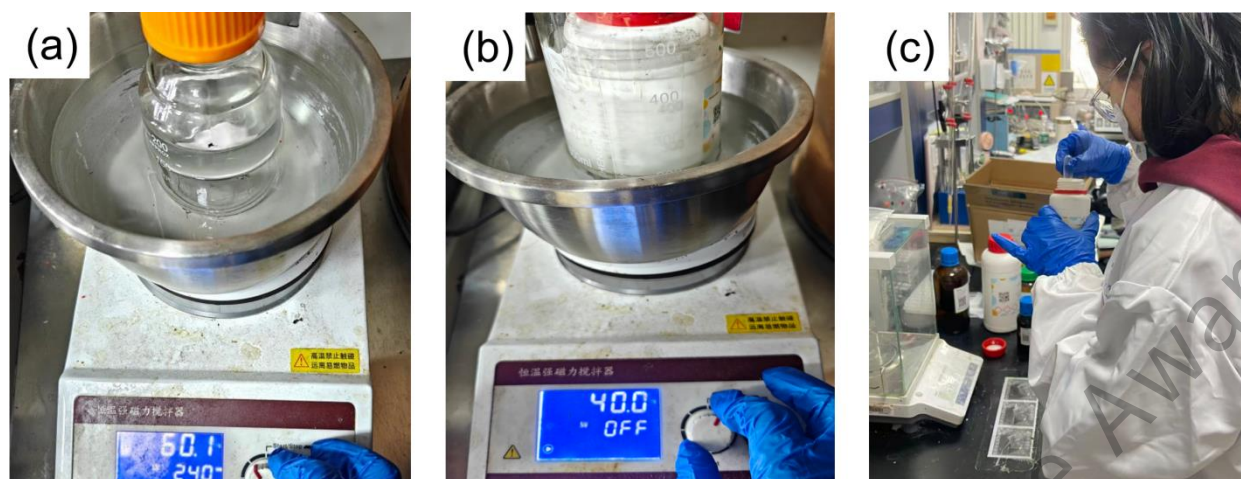


Figure 1. Experimental raw material solution preparation process: (a) PVA and SDS are dissolved in heated water; (b) Octadecane is heated and dissolved in a water bath; (c) Preparation of dispersed oil phase

Continuous water phase preparation: PVA in the water phase is mainly used to increase the viscosity of the water phase, and SDS is used to reduce the oil-water interfacial tension and promote droplet formation. The two jointly improve the stability of droplets during microfluidic spherification and subsequent solidification processes, and reduce droplet aggregation, adhesion and rupture. According to the set mass fraction, add PVA, SDS and DETA to deionized water to prepare a continuous water phase. During the preparing, first add about 80% water to a clean container, and slowly add PVA under continuous stirring to avoid powder agglomeration. The solution is then heated to 60°C until the PVA is completely dissolved, resulting in a clear solution without visible particles. After the solution is cooled to room temperature, SDS and DETA are added in sequence, and deionized water is used to make up to the set total mass. The mass fractions of DETA, PVA and SDS in the continuous aqueous phase were 4.0 wt%, 1.5 wt% and 1.0 wt% respectively.

Dispersed oil phase preparation: OD serves as the octadecane core, which is used to realize latent heat storage and release through solid-liquid phase-change; IPDI is used as an oil-soluble shell material precursor, mixed with molten OD to form a dispersed oil phase. Before the experiment, OD was fully melted in a 40°C water bath, and was used after it was transformed into a clear and uniform liquid. As a water-soluble cross-linking reaction monomer, DETA diffuses from the continuous water phase to the oil-water interface, and polymerizes with the isocyanate group of IPDI at the oil-water interface, gradually forming a polyurea shell. IPDI has strong moisture sensitivity. Try to shorten the time it is exposed to humid air during the collection

process, and seal and store it immediately after sampling. The dispersed oil phase is prepared according to the set OD/IPDI mass ratio. Taking the OD/IPDI mass ratio as 3:1 as an example, after weighing 3 g of molten OD, add 1 g of IPDI and stir at 300 r/min for 30 minutes at 40 °C until the system is homogeneous and there is no visible stratification.

Each solution is calculated based on the actual weighing results of the current experiment, and the mass method is used to determine the proportion of each component. The containers used must be kept clean and dry before weighing. The continuous water phase is allowed to stand for deaeration before use to reduce flow fluctuations and channel blockage caused by bubbles entering the chip. After the oil phase is prepared, it is covered and kept warm, and its residence time before injection is minimized to reduce the possibility of side reactions between IPDI and environmental moisture.

2.2 Instruments and devices

The microcapsule experimental equipment (customized, purchased from Shanghai Pengzan Biotechnology Co., Ltd., China) mainly consists of a pressure control system, a flow focusing microfluidic chip, a constant temperature water bath, a droplet collection and stirring device, and an optical microscope (M500C model, purchased from Shenzhen Huaxian Optical Instrument Co., Ltd., China); the post-processing uses a suction filtration device and a blast drying oven, and a connecting tube with an inner diameter of 0.6 mm PTFE is used to connect the raw material tube and the flow focusing microfluidic chip (Figure 2). The dynamic formation process of droplets is continuously recorded using an M500C optical microscope and supporting image acquisition system. Before formal acquisition, spatial calibration was completed under the same objective lens magnification, camera resolution, digital zoom ratio and image cropping range as the experimental observation. Taking a 100 μm droplet as an example, according to the software interface record, the image acquisition frame rate is 9.5 fps, and the spatial calibration coefficient is 0.80 pixel/ μm , that is, 1 pixel corresponds to 1.25 μm ; at the same time, a 100 μm ruler is added to the screen to review the calibration results. The microscope observation magnification is 10 times.

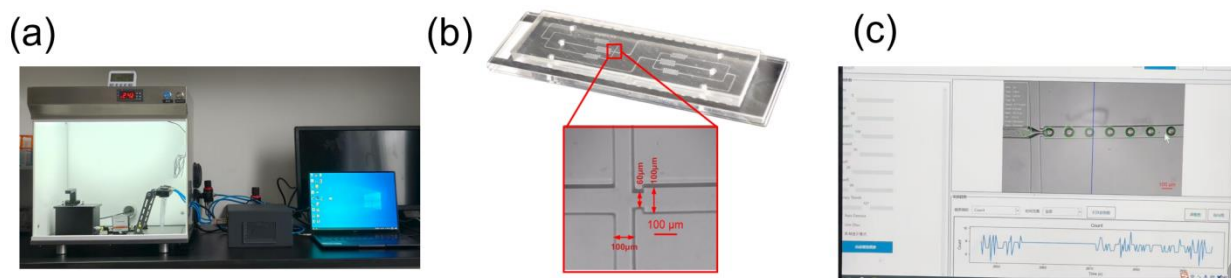


Figure 2. Experimental device and online monitoring: (a) Microfluidic experimental device; (b) Flow focusing chip; (c) Dynamic monitoring of droplet generation.

Before recording each set of operating conditions, adjust the light source intensity, focal length, and exposure parameters to ensure that the edges of the microchannel and the droplet contours are clearly distinguishable. Subsequently, fix the positions of the microscope, lens, camera, and microfluidic chip, and perform image acquisition under the conditions of maintaining the same field of view, magnification, resolution, and cropping ratio. After changing the flow rates of the two phases, wait until the droplet breakup location, generation period, and downstream motion state are basically stable, and then continuously acquire effective images. After reading the microscopic images frame by frame, convert the pixel size to actual size based on the calibration coefficient. The droplet generation frequency is calculated based on the number of effective droplets passing through a fixed observation area per unit time.

2.3 Experimental design and procedures

In this article, the n-octadecane@polyurea core-shell system is investigated, focusing on the underlying mechanisms of microcapsule spheroidization, interfacial shell formation, and phase-change behavior under shell confinement (as shown in Figure 3). Three sets of interconnected variables are designed: spheroidization, formation of the polyurea shell, and phase-change behavior under shell confinement. The first group varies the flow conditions of the dispersed and continuous phases and uses the capillary number in the dispersed phase to characterize the competition between viscosity and interfacial tension, investigating the spheroidization of droplet precursors used to form the phase-change microcapsules. The second group varies the OD/IPDI mass ratio to analyze the shell precursor ratio, leakage, shell integrity, and their influence on the characteristic peaks of urea. The third group compares the core and shell compositions as well as the crystal morphology and thermal behavior of samples at different scales, and explores the effects of shell confinement, relative shell content, and particle size.

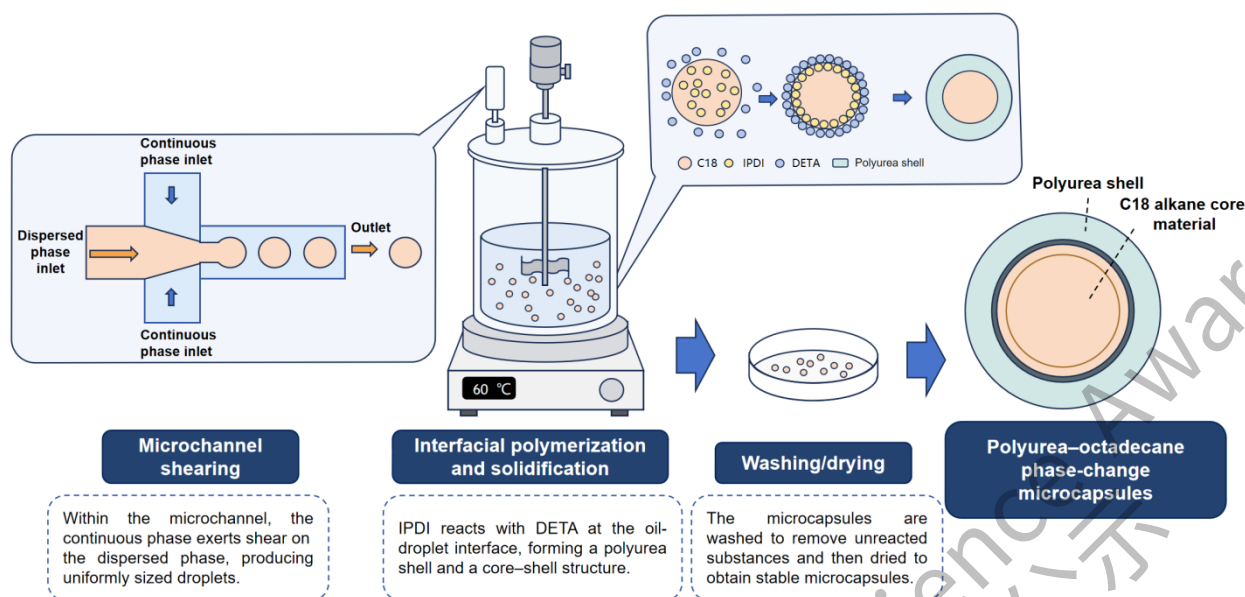


Figure 3. Schematic diagram of the preparation process of n-octadecane@polyurea phase-change microcapsules.

2.3.1 Experimental study on the spherical formation of precursor droplets of phase-change microcapsules

In order to reveal the regulation mechanism of interface force on the stable spherical formation of droplet precursors for phase-change microcapsules. Melt OD in a 40°C water bath, and add IPDI according to the set OD/IPDI mass ratio to prepare a dispersed oil phase; the continuous water phase consists of an aqueous solution of PVA, SDS and DETA, in which the DETA concentration is adjusted to 4 wt%. The two phases are introduced into the flow focusing microchannel respectively, and the interaction between viscosity, shear and interfacial tension is changed by changing the flow conditions of the two phases.

During the experiment, micrographs of droplet formation were continuously captured, and the accompanying software of the experimental equipment was used to identify and extract parameters such as droplet contour, average particle size, standard deviation, CV value, and generation frequency from the online micrographs. These parameters were then cross-verified with the interface deformation and fracture states predicted by CFD simulation, and the capillary number range corresponding to stable ball formation was determined from a dynamic experimental perspective.

2.3.2 IPDI/DETA interfacial polymerization and microcapsule shell formation experiment

This part of the experiment aims to elucidate the reaction mechanism of the interfacial polymerization of IPDI/DETA to form a polyurea shell. The droplet precursors generated by

microfluidics continuously enter the aqueous phase collection system containing DETA. The aqueous phase DETA diffuses toward the oil-water interface, and its amino group reacts with the isocyanate group in the oil phase IPDI to form a urea bond. Due to the low solubility of polyurea in the oil and water phases, the primary polymer is preferentially deposited at the interface; as the monomers continue to migrate and react, the local deposited layers gradually connect and form a continuous polyurea shell, thereby fixing the liquid OD inside the shell.

In order to investigate the impact of the shell precursor formula ratio on shell formation (as shown in Figure 4), on the basis of keeping other conditions consistent, different OD/IPDI mass ratios (3:1, 5:1, 7:1, etc.) were set, and the changes in leakage, adhesion, shell integrity, and polyurea characteristics corresponding to changes in the relative content of the shell precursor were compared. The collected liquid was then placed in a constant-temperature water bath at 60 °C for 6 h, and the particles were maintained dispersed under gentle stirring at a constant rotation speed of 60 r/min, allowing the interfacial reaction to continue and complete the solidification of the shell.

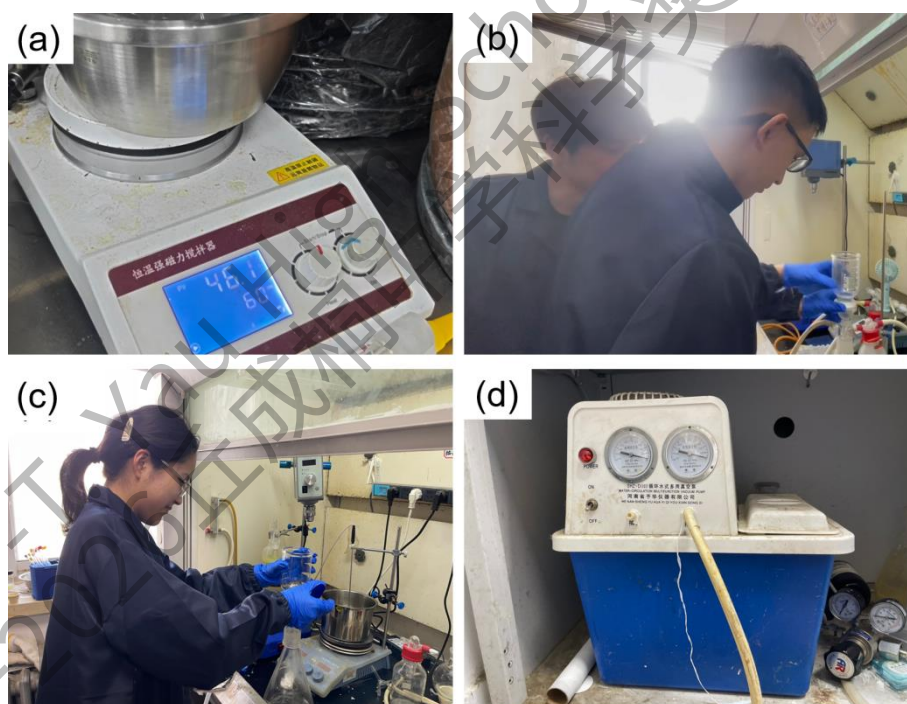


Figure 4. Curing experimental study: (a) Heating and solidification temperature rise stage; (b-c) Assembly of filtration device; (d) Vacuum pump for vacuum filtration.

After the reaction is completed and cooled to room temperature, the microcapsules are collected by suction filtration. The filter cake is first washed with deionized water to remove residues in the water phase, and then washed with n-hexane to remove uncoated OD and oil-

soluble residues on the particle surface; deionized water washing and n-hexane washing are performed alternately 3-5 times. The washed samples were dried in a blast drying oven at 60 °C for 12 h, cooled and sealed for subsequent testing and research.

2.3.3 Core-shell composition and confined phase transition experiments

This part of the experiment is used to analyze the regulation mechanism of shell confinement on the phase-change behavior of the octadecane core. Microcapsules with different OD/IPDI mass ratios and 100, 300 and 500 μm scales were selected as comparison objects, and pure OD was used as the benchmark sample. On the premise that the chemical composition of the core material remains unchanged, the characteristic functional groups of each sample are first confirmed through FTIR, and then the position and relative intensity of the characteristic diffraction peaks are compared through XRD to determine whether the shell formation and particle size changes change the basic crystal form of OD; DSC is then used to compare the melting temperature, crystallization temperature, supercooling and phase-change enthalpy.

2.4 Characterization method

2.4.1 Morphological characterization of microcapsules and their droplet precursors

The M500C optical microscope was used to record the droplet formation process, and images were collected at a fixed field of view and stable flow stage, and combined with real-time microscopic image recognition to obtain the average particle size and generation frequency. An S-4700 scanning electron microscope (SEM, Hitachi, Japan) was used to observe the overall sphericity, particle size consistency, surface continuity, adhesion and damage of the dried microcapsules to provide morphological evidence for whether interfacial polymerization forms stable core-shell particles. For SEM sample preparation, first gently sprinkle the fully dried microcapsules onto the conductive adhesive. Non-adherent particles were subsequently removed by a gentle stream of nitrogen. The specimen was then sputter-coated with a gold layer.

2.4.2 Fourier transform infrared spectroscopy test

Fourier transform infrared spectroscopy (FTIR, Agilent Technologies Cary 630, USA) was used to analyze the functional group composition of pure OD and dried microcapsules, with a spectral range of 4000-500 cm^{-1} . Focus on comparing the OD alkane characteristic absorption near 2950-2850 cm^{-1} , 1460 cm^{-1} and 720 cm^{-1} , and the polyurea-related absorption near 1650 cm^{-1} and 1550 cm^{-1} . By whether the peak position appears and how the relative peak intensity changes at

different OD/IPDI feed mass ratios, it can be judged whether the polyurea shell is formed and the changing trend of the relative content of the shell material.

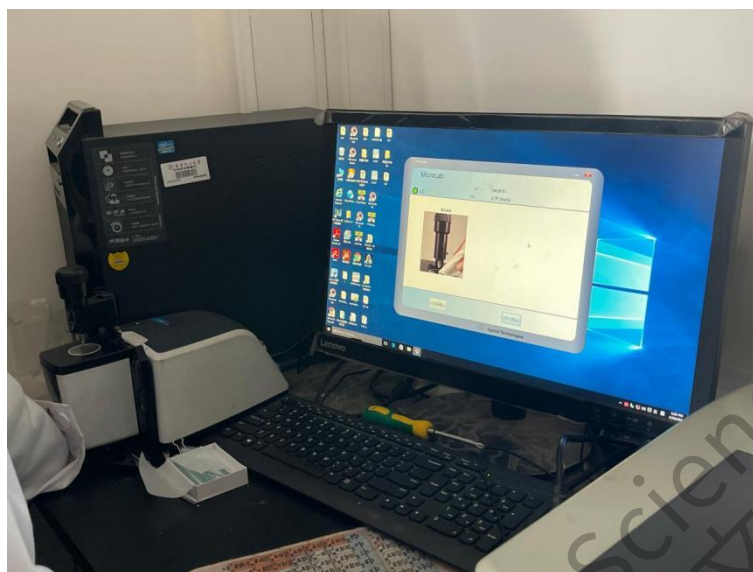


Figure 5. Fourier transform infrared spectrometer test on the sample.

2.4.3 X-ray diffraction testing

X-ray diffraction (XRD, Bruker D8 ADVANCE, Germany) was used to analyze the crystal structure of pure OD and each group of dry microcapsules, with a scanning range of $2\theta=5-90^\circ$, a scanning rate of $5^\circ/\text{min}$, $\text{Cu K}\alpha$, $\lambda=0.154 \text{ nm}$. Before the XRD test, gently grind the dried samples and evenly spread them into the sample tank. Keep the spreading state of each group of samples consistent, and compare the position, peak shape and relative intensity of the OD characteristic diffraction peaks in the range of $2\theta=10^\circ-30^\circ$. Whether the peak position is shifted is used to determine whether interfacial polymerization and spatial confinement induce new crystal form changes, and the relative intensity change is used as auxiliary evidence of changes in the degree of crystallization and the relative content of the core material.

2.4.4 Differential Scanning Calorimeter Testing

Differential scanning calorimetry (DSC, NETZSCH, Germany) was used to characterize the crystallization and melting behavior of pure OD and microcapsules. The test temperature range was $-10-70^\circ\text{C}$, the heating and cooling rates were both $10^\circ\text{C}/\text{min}$, and the test was conducted under N_2 protection. The melting peak temperature and melting enthalpy are obtained from the heating curve, and the crystallization peak temperature and crystallization enthalpy are obtained from the cooling curve; the phase-change enthalpy is obtained by integrating the corresponding absorption and exothermic peak areas, and the supercooling is calculated according to the

difference between the melting peak temperature and the crystallization peak temperature. Different samples use consistent temperature programs and data processing methods to compare the effects of OD/IPDI mass ratio and particle size changes on phase-change temperature and heat storage capacity.

For the DSC test, weigh 5-10 mg of the sample, place it in an aluminum crucible, and hermetically seal it. An empty aluminum crucible was used as reference. The nitrogen flow rate was set to 40 mL/min. The integration baseline, onset/end temperatures, and peak temperature determination method were kept consistent across all samples, and the original heat flow curve was retained. For the thermal cycling stability test, the sample was subjected to 100 heating-cooling cycles within the same temperature boundaries. The DSC peak position, temperature (T_{peak}), peak shape, and phase-change enthalpy were compared before and after the cycles.

During the DSC test, weigh 5-10 mg of the sample and place it in an aluminum crucible to seal it. Use the empty crucible as a reference; the nitrogen flow rate is: 40 mL/min. The integration baseline, start and end temperatures, and peak temperature reading methods of each sample remain consistent, and the original heat flow curve is retained. The thermal cycling stability experiment uses the same temperature rise and fall boundaries to perform 100 thermal cycle on the sample, and compares the DSC peak position, peak shape and phase-change enthalpy before and after the cycles.

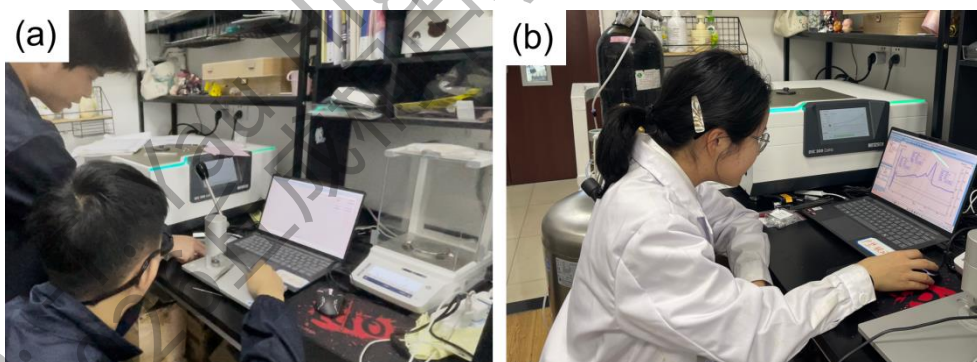


Figure 6. DSC thermal performance testing of samples

3. Results and discussion

3.1 Interfacial spheroidization behavior of phase-change microcapsule precursor droplets

The spheroidization of droplets in restricted microchannels is not a simple flow rate optimization, but a process in which the droplet interface deforms, necks, and breaks under the combined action of viscous shear and interfacial tension. This section uses CFD as an auxiliary

analysis method to observe the evolution of the interface morphology under different capillary numbers, and combines the experimental results to analyze the relationship between the interface force and the spherical state of the droplets.

3.1.1 Simulation methods and interface force parameters

In this article, flow-focused microchannels are used to study the formation process of n-octadecane@polyurea droplet precursors. As shown in Figure 7, the oil phase is the dispersed phase and the aqueous phase is the continuous phase. The continuous phase squeezes and shears the dispersed phase from the upper and lower sides; the channel width and depth are both 240 μm .

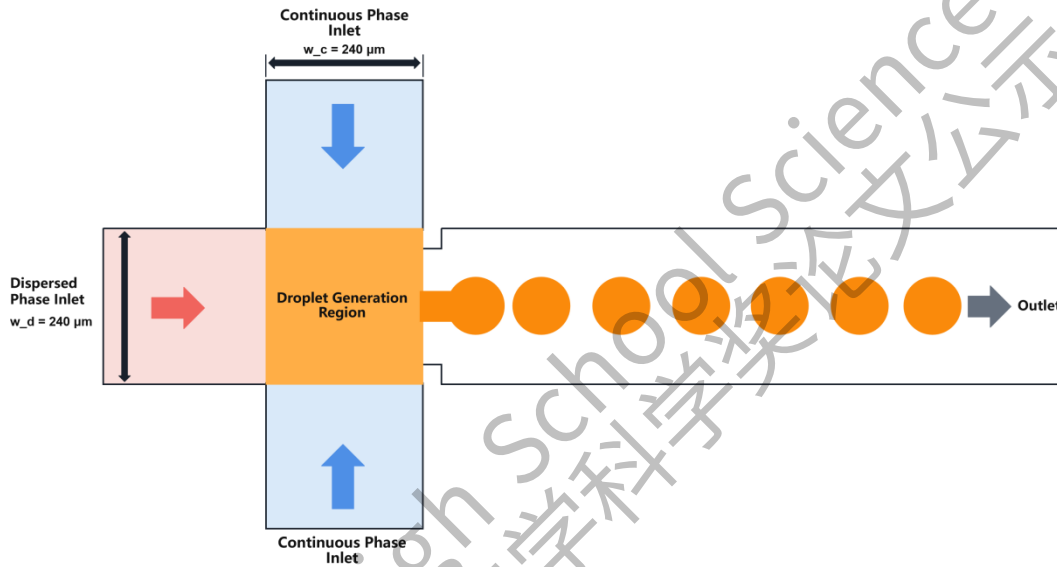


Figure 7. Schematic diagram of the spherical forming process of microfluidic channels

The COMSOL transient two-phase flow model is used, and the dynamic changes of the oil-water interface are tracked through the VOF method. The model adopts the classical mass and momentum conservation equations, assuming that the two phases are incompressible, immiscible and in a laminar flow state, ignoring the influence of gravity; the physical property parameters of the two phases are shown in Table 1. The focus of the simulation is to compare the evolution of droplet morphology under different interface stress conditions, rather than modifying the classical fluid mechanics model itself.

In order to characterize the relative relationship between the viscosity of the dispersed phase and the interfacial tension, the capillary number Ca_d of the dispersed phase is used as the key dimensionless parameter:

$$Ca_d = \frac{\mu_d v_d}{\sigma} \quad (1)$$

In the formula, μ_d is the dynamic viscosity of the dispersed phase, v_d is the flow rate of the dispersed phase, and σ is the oil-water interfacial tension.

Ca_d reflects the strength of the viscous effect relative to the interfacial tension, and its changes can be used to judge the trend of the droplet interface transitioning from stable recovery to continuous deformation, tailing, and jet flow.

3.1.2 Simulation results and analysis

Under the condition of fixed continuous phase flow velocity $v_c=1.0 \times 10^{-2}$ m/s, changing the dispersed phase flow velocity v_d causes Ca_d to change accordingly; when the cross-sectional areas of the two branches are the same, the flow ratio q can be approximately expressed as v_d/v_c . The material parameters are shown in Table 1, and the droplet shapes corresponding to different Ca_d s are shown in Table 2 and Figure 8.

Table 1 Simulation parameters of two-phase materials

project	Dispersed phase	Continuous phase
	(oil phase)	(aqueous phase)
Density $\rho/(\text{kg} \cdot \text{m}^{-3})$	890	1000
Dynamic viscosity $\mu/(\text{Pa} \cdot \text{s})$	0.117	0.072
Interfacial tension parameters $\sigma/(\text{N} \cdot \text{m}^{-1})$	0.02	0.02

When the continuous phase conditions remain unchanged, the increase in dispersed phase flow rate and Ca_d will change the force balance on the droplet interface, causing the droplets to gradually transform from periodic breakage to trailing and jet states.

Table 2. Droplet forming parameters and status under fixed continuous phase flow rate

v_c (m/s)	v_d (m/s)	Ca_d	q	Droplet forming state
0.01	0.002	0.0072	0.2	The droplets are smaller, the formation period is longer, and the number of downstream droplets is small.
	0.004	0.0144	0.4	Stable into balls, with uniform droplet size and spacing
	0.006	0.0216	0.6	The liquid tongue is significantly extended, the fracture position moves downstream and tails appear.
	0.008	0.0288	0.8	The tailing is more obvious, and droplet adhesion and local fusion appear.
	0.010	0.036	1.0	A continuous jet is formed, and no complete fracture occurs during the observation period.

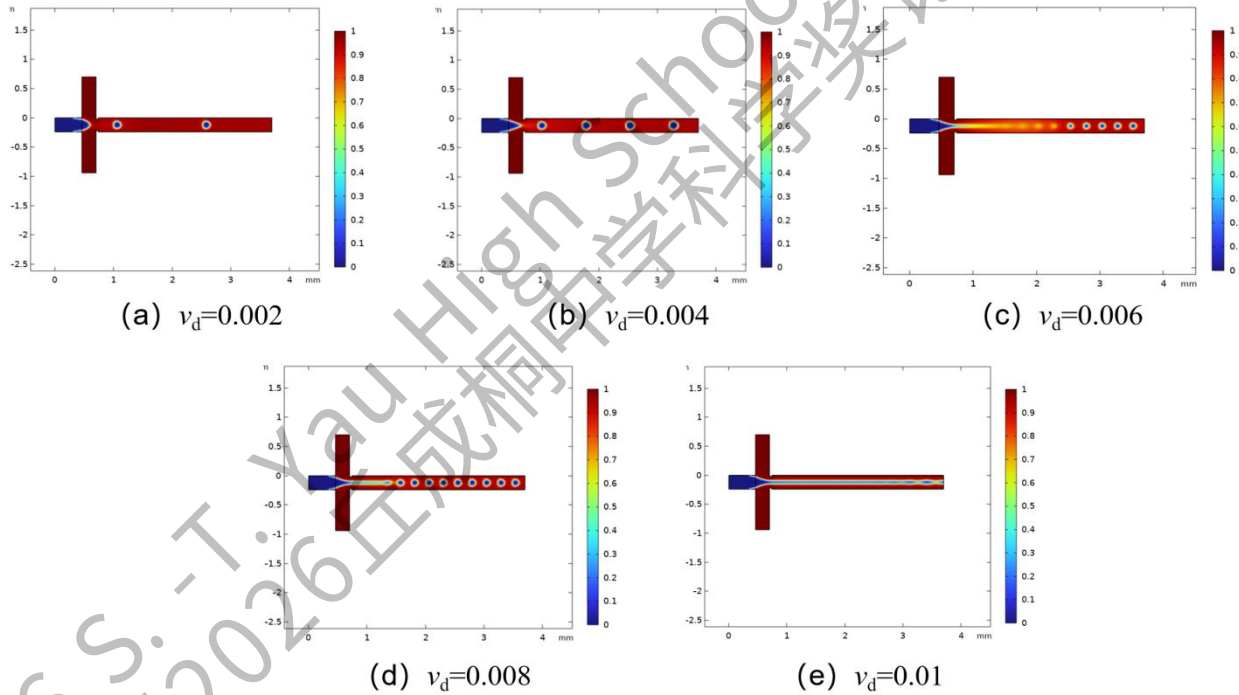


Figure 8. Simulation effect of dispersed phase flow rate on droplet balling (units in the figure are m/s).

When $Ca_d=0.0072$, the interfacial tension is relatively dominant, and the droplets can break, but the dispersed phase is replenished slowly and the formation period is longer; when Ca_d increases to 0.0144, the viscous/shearing effect and the interfacial tension reach a more coordinated dynamic balance. The droplets periodically undergo expansion-necking-fracture-

retraction to form more regular droplets; when Ca_d further increases to 0.0216 and above, the dispersed phase continues to advance and the interface deformation is enhanced, the necking position moves downstream, and tailing and jets gradually appear. It can be seen that the droplet morphology transformation comes from the competition between the interface deformation driving force and the interface restoring force, rather than the process improvement brought about by a single increase in flow rate. Figure 9 further summarizes the above force-bearing mechanism. In this system, droplets can still break into balls within the range of $Ca_d = 0.0072$ to 0.0216 , and moderate Ca_d is more conducive to the formation of regular and periodic droplets; when Ca_d continues to increase, the interfacial tension is difficult to restore the droplet shape in time, and the system gradually transforms into a tailing and jet state.

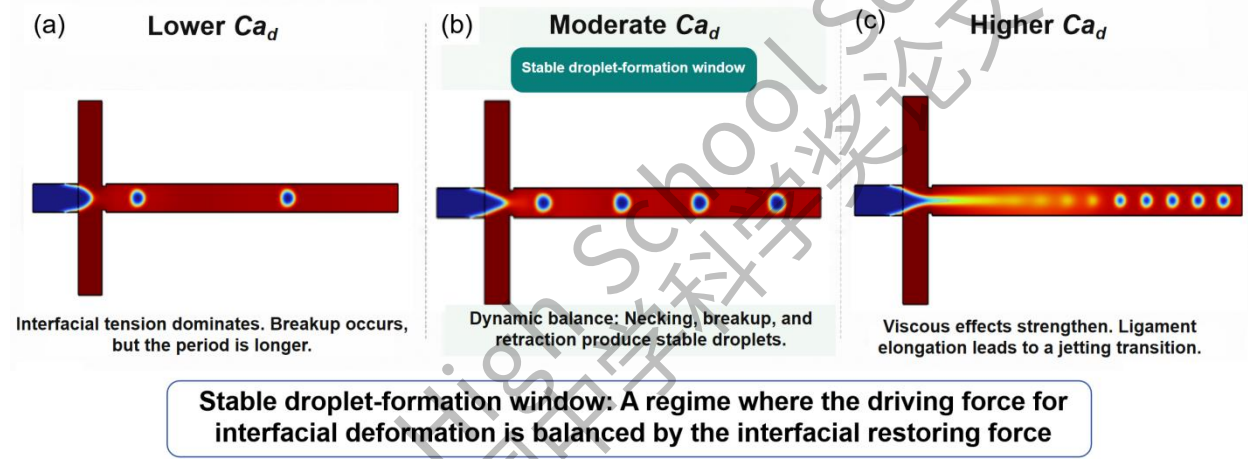


Figure 9. Mechanism of droplet ball formation driven by interfacial force competition in restricted microchannels.

3.2 Droplet spheroidization behavior assisted by online microscopic image recognition

To verify the relationship between interface force-droplet morphology revealed by Computational Fluid Dynamics (CFD), this article employs online microscopic image recognition for quantitative analysis of microfluidic microscopic images. The experimental equipment is equipped with online microscopic image recognition software to extract droplet size, CV value, and generation frequency, which are used to determine whether the droplets are in a regular, periodic state of breaking into spheres. The recognition results are shown in Figure 10. As the flow rate of the dispersed phase increases, the average droplet size increases from $77.6 \mu\text{m}$ to $136 \mu\text{m}$ and $194.9 \mu\text{m}$, while the droplet generation frequency also increases, indicating that after the

enhancement of the dispersed phase supply, the droplets undergo more pronounced interface expansion before breaking, consistent with the trend of liquid tongue extension observed in CFD.

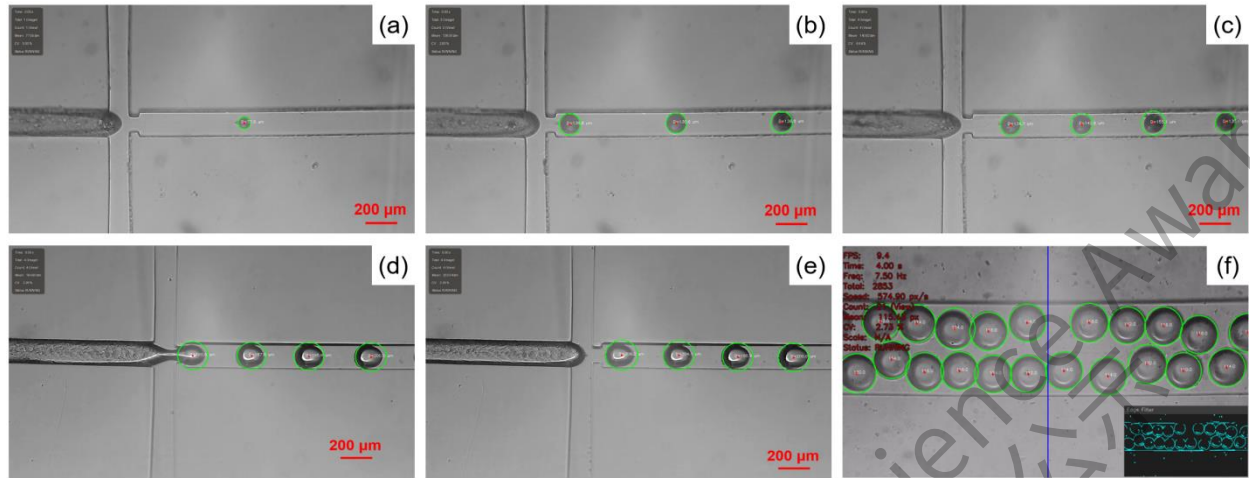


Figure 10. Microsphere formation results through online microscopic image recognition.

Further experiments were conducted with a continuous phase flow rate of 0.01 m/s and a dispersed phase flow rate of 0.005 m/s for comparison with simulations (Figure 11). Online identification of microscopic images revealed an average droplet size of 196.6 μm and a CV value of 1.88%. In the experiments, droplets were consistently and regularly generated; simulations under the same conditions also exhibited periodic breakup. The consistency between the two provides dynamic experimental evidence for the analysis of droplet spheroidization controlled by interfacial forces, rather than simply being used to screen optimal process parameters.

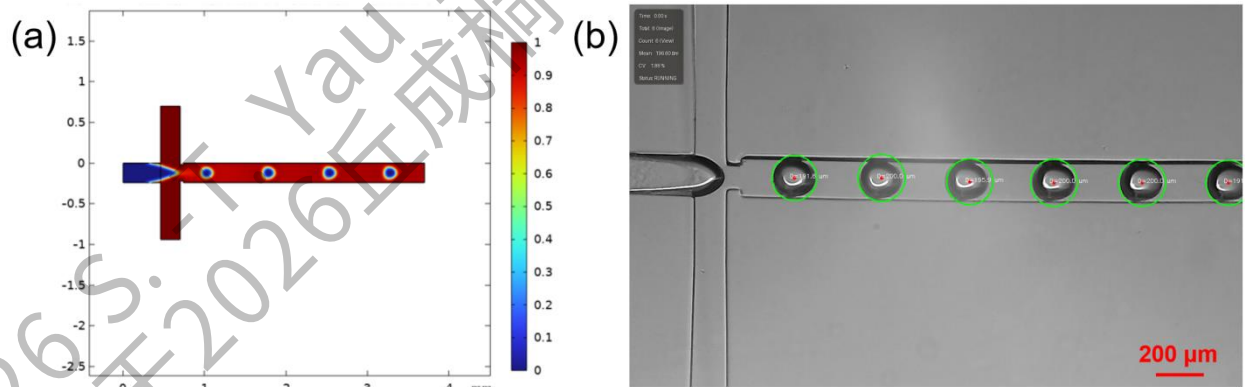


Figure 11. Microsphere molding simulation results and experimental verification.

3.3 Interfacial polymerization and structural characterization of microcapsules

In order to analyze the effect of IPDI/DETA ratio on the formation of polyurea shell and the morphology of microcapsules, SEM characterization of microcapsules prepared under different

ratios was performed. The results are shown in Figure 12. As the relative dosage of IPDI increases, the spherical state of the microcapsules gradually improves: as shown in Figure 12(a), when the dosage of IPDI is low, there are more irregular particles, damaged structures and adhesion agglomerations in the sample, and some particle surfaces have collapse or incomplete coating; as shown in the Figure 12(b), after appropriately increasing the IPDI ratio, the number of microcapsules with intact spherical contours increased markedly, and interparticle adhesion was alleviated to some extent; however, a small number of microcapsules with irregular morphology were still observed; as shown in Figure 12(c), when the IPDI dosage was further increased to an appropriate range, most microcapsules showed a more regular spherical shape, the surface coating was more continuous, the particle boundaries were clear, and the damage and agglomeration were significantly reduced. This change shows that increasing the IPDI content in the oil phase can provide more -NCO groups for interfacial polymerization, allowing them to fully react with DETA in the water phase, thereby promoting the continuous deposition of polyurea at the oil-water interface and forming a more continuous and complete shell.

When IPDI is relatively insufficient, the number of -NCO groups participating in the interface shelling reaction decreases, and the generated polyurea is difficult to completely cover the OD droplets, thereby forming a thin or locally discontinuous polyurea shell, causing molten OD to seep out from the shell defects, as shown in Figure 12(d). The leaked OD will make the microcapsule surface sticky and cause particle adhesion, bridging, and agglomeration during subsequent solidification, filtration, and drying processes, which corresponds to the irregular particles and agglomeration observed in Figure 12(e-f). Experimental results show that when the DETA concentration is maintained at 4 wt% and OD/IPDI mass ratio is no higher than 7:1, the overall sphericity and dispersion of the microcapsules obtained are better, the shell surface is relatively continuous, and no obvious core material leakage is observed; when the OD/IPDI mass ratio further increases, the amount of IPDI corresponding to the unit mass of OD decreases, resulting in a relative shortage of shell material precursors, a decrease in the shell integrity of the microcapsules, and an increased risk of core material leakage, adhesion, and agglomeration. The SEM morphology is basically consistent with the changes in the macroscopic state of the sample, which together indicate that appropriately increasing the IPDI ratio is beneficial to the formation of the polyurea shell and the improvement of the structural stability of the

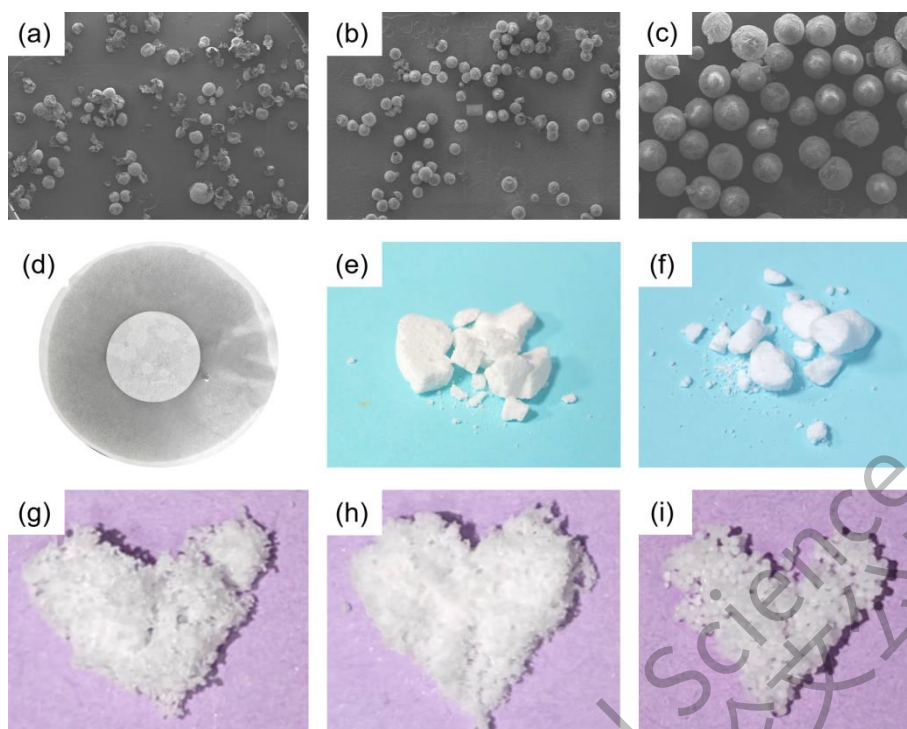


Figure 12. Morphology of n-octadecane@polyurea phase-change microcapsules: (a-c) SEM morphology of the microcapsules under different relative amounts of IPDI; (d-f) Macroscopic morphology of the microcapsules with core material leakage, particle adhesion and agglomeration under inappropriate conditions of IPDI/DETA mass ratio; (g-i) Macroscopic morphology of 100, 300 and 500 μm n-octadecane@polyurea phase-change microcapsules with intact shells and no obvious leakage.

After the aqueous phase DETA diffuses to the oil-water interface, its amino group undergoes an addition reaction with the isocyanate group of the oil phase IPDI to form a urea linkage; since the generated polyurea has low solubility in the two phases, the reaction product is preferentially enriched at the interface and gradually forms a coating layer [12]. As the reaction continues, the interface product evolves from local deposition to a continuous polyurea shell, thereby fixing the initial liquid oil droplet into a stable core-shell structure, as shown in Figure 13.

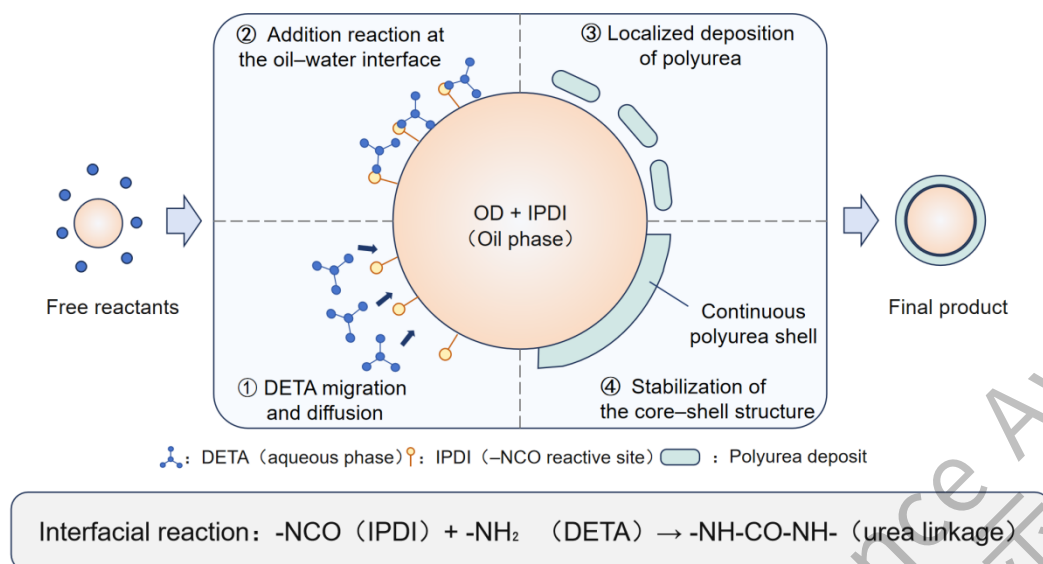


Figure 13. Schematic diagram of the polyurea shell formation mechanism of IPDI/DETA at the oil-water interface.

In order to explore the chemical composition, interface reaction behavior and core material crystallization characteristics of n-octadecane@polyurea phase-change microcapsules at different OD/IPDI feed mass ratios. The samples were characterized by Fourier transform infrared spectroscopy (FTIR) and X-ray diffraction (XRD), and the results are shown in Figure 14.

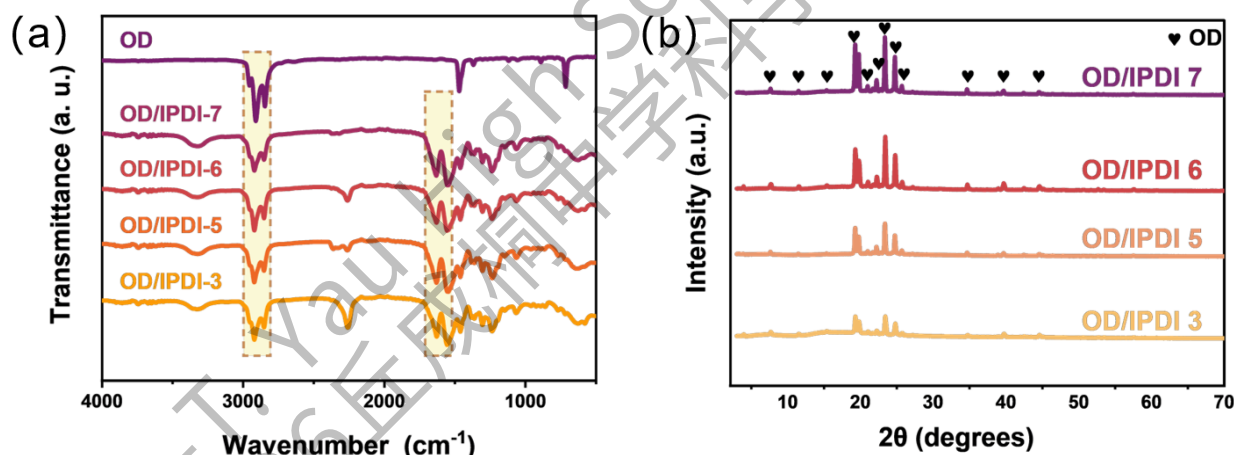


Figure 14. Chemical structure and crystal structure characterization of polyurea-octadecane microcapsules with different OD/IPDI ratios: (a) Fourier transform infrared spectra of 100 μm microcapsules under different core-shell mass ratios; (b) XRD patterns of 100 μm microcapsules under different core-shell mass ratios.

Figure 14(a) is a diagram of the infrared spectroscopy (FTIR) of microcapsules with different OD/IPDI core-shell ratios. It can be seen from the figure that at $2950\text{-}2850\text{ cm}^{-1}$ (asymmetric and symmetrical telescopic vibration of $\text{-CH}_2\text{-}/\text{-CH}_3$), 1460 cm^{-1} (bending vibration of -CH_3) and 720 cm^{-1} (in-plane swing vibration of long-chain alkanes), there are obvious characteristic peaks of

OD [13]; as the proportion of IPDI in the system increases, 1650 cm^{-1} (urea-based The characteristic peaks of polyurea at C=O stretching vibration) and 1550 cm^{-1} (N-H bending vibration) gradually increased [14], proving that the polyurethane structure was successfully prepared and generated. At the same time, the relative strength of the OD characteristic peaks weakened, which was consistent with the trend of decreasing the mass fraction of the core material and increasing the relative content of the polyurethane shell. Figure 14 (b) shows the XRD map of microcapsules with different OD/IPDI ratios. Pure OD exhibits obvious characteristic diffraction peaks in the range of 2θ of 10° - 30° . The position of the OD characteristic diffraction peaks of the microcapsule samples with different ratios has not been significantly offset, indicating that the polyurethane coating has not changed the basic crystal shape of the OD, and the core material can still maintain its crystallization and phase transition capabilities.

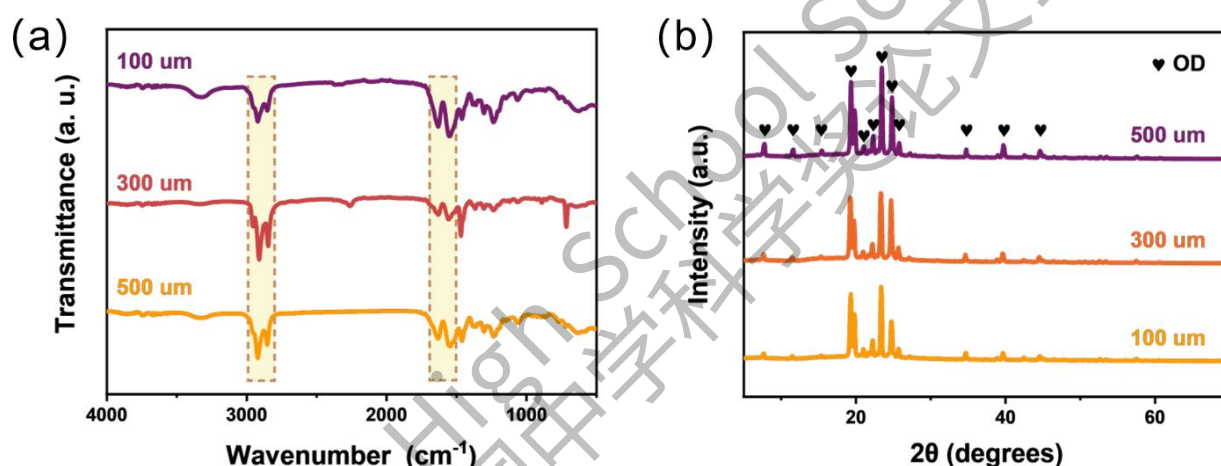


Figure 15. Characterization of chemical structure and crystal structure of n-octadecane@polyurea microcapsules of different particle sizes: (a) Fourier infrared spectra of 100, 300, and 500 μm -level microcapsules; (b) XRD spectra of 100, 300, and 500 μm -level microcapsules.

Figure 15(a) shows the FTIR spectra of microcapsules with particle sizes of 100, 300, and 500 μm . All samples simultaneously present the characteristic peaks of alkane in OD and the characteristic absorption peaks of polyurea, and the main peak positions are basically the same, indicating that changing the particle size of the microcapsules does not cause significant changes in its main chemical composition and functional group structure, and IPDI/DETA interfacial polymerization can occur under various particle size conditions. Figure 15(b) shows the XRD patterns of microcapsules with different particle sizes. Samples with different particle sizes all show characteristic diffraction peaks that completely match pure OD, indicating that the OD in

microcapsules with different particle sizes maintains the original crystal structure. The OD characteristic diffraction peak of the larger particle size sample is relatively strong, indicating that the degree of crystallization order may be improved. This may be because as the particle size of the microcapsules increases, the shell confinement of the octadecane core per unit volume is relatively weakened, which is conducive to the orderly arrangement and crystallization of OD molecules.

Larger microcapsules experience weaker shell confinement, and the crystallinity is slightly increased, which provides guarantee for the thermal energy-storage performance of the microcapsules. From the structure-activity relationship, an increase in the relative IPDI feed content was reflected in the increased intensity of the characteristic polyurea absorption bands, consistent with a higher relative shell content, while the OD crystalline peak position remains basically unchanged, indicating that changes in the core-shell composition mainly change the interface and confinement environment, rather than inducing a new crystalline transformation of octadecane; this provides a structural basis for subsequent explanations of phase transition temperature, supercooling and enthalpy changes from the perspective of shell confinement.

The morphology and structure of the successfully prepared phase-change microcapsules of different scales were characterized. The optical microscopic images in Figure 16 (a-c) show that microcapsules of different particle sizes are all regular spherical shapes with uniform particle sizes, without obvious agglomeration, adhesion or breakage. This proves that the preparation process of microchannel emulsification combined with interfacial polymerization can achieve precise control of microcapsule size. The scanning electron microscope images in Figure 16 (d-f) further confirm that the surfaces of microcapsules with different particle sizes all present a dense surface morphology without obvious cracks, pinholes or shell defects, indicating that the interfacial polymerization reaction is sufficient and a complete and continuous polyurea shell is formed, which can effectively maintain the microcapsule structure. Therefore, the complete and continuous polyurea shell is not only the morphological evidence of the core-shell structure formed by interfacial polymerization, but also the structural prerequisite for subsequent stable shell confinement, limiting the migration of the liquid core material, and maintaining reversible phase-change.

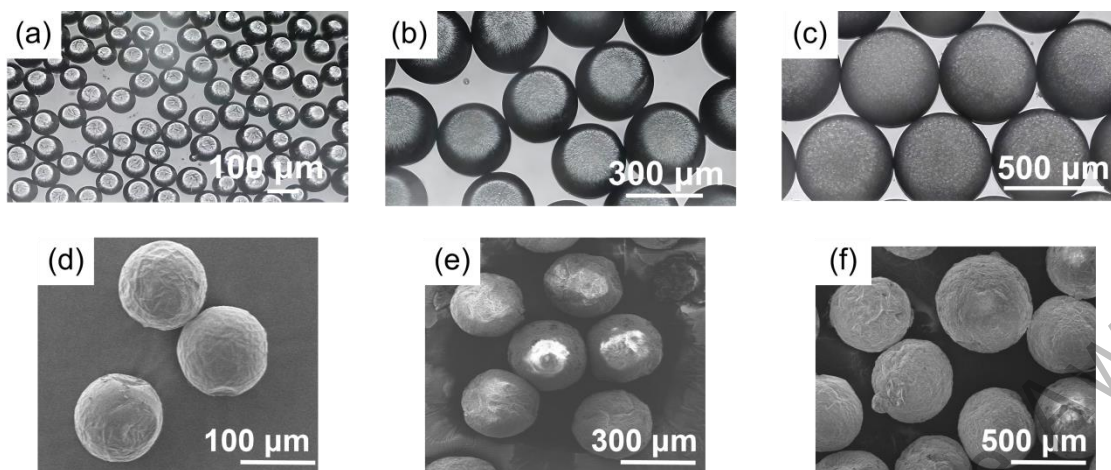


Figure 16. Microcapsule images of different scales: (a-c) Optical microscope images; (d-f) SEM images of microcapsules of different scales.

3.4 Microcapsule phase transition behavior and shell confinement

In order to explore the thermal energy-storage performance of phase-change microcapsules, the samples were characterized by differential scanning calorimetry (DSC) (Figure 17). The DSC heating curve (Fig. 17a) and cooling curve (Fig. 17b) show that the microcapsule samples prepared at all tested OD/IPDI feed mass ratios show obvious endothermic peaks during the heating process and symmetrical exothermic peaks during the cooling process, indicating that the coating of the polyurea shell does not inhibit its crystallization-melting behavior and has a phase-change thermal-storage capability. The overall melting temperature T_m of the microcapsules is distributed in the range of 32.3–34.1°C (as shown in Figure 17c), which is slightly higher than the melting point of pure octadecane (28.2°C). The crystallization temperature T_c showed a monotonous upward trend as the OD/IPDI feed mass ratio increased, from 17.8 °C in 3:1 to 20.8 °C in 7:1, and the corresponding supercooling dropped from 16.0 °C to 11.5 °C, indicating that the higher the proportion of core material, the smaller the supercooling. This is because at high core material content, shell confinement of the octadecane core is weakened, and the steric hindrance of crystallization nucleation is reduced [15]. The nucleation process is more likely to occur, which is more conducive to practical heat storage applications. Figure 17d shows that as the OD/IPDI feed mass ratio increases, the melting enthalpy ΔH_m of the microcapsules significantly increases from 151.1 J/g to 202.2 J/g, and the solidification enthalpy ΔH_c increases from 151.1 J/g to 203 J/g, proving that the phase-change process is reversible and there is no obvious heat loss or irreversible structural change. The enthalpy value increases with the increase of core material

proportion, which is consistent with the theoretical proportional relationship between phase-change enthalpy and core material mass fraction, indicating that the heat storage density of microcapsules can be effectively controlled by adjusting the OD/IPDI ratio.

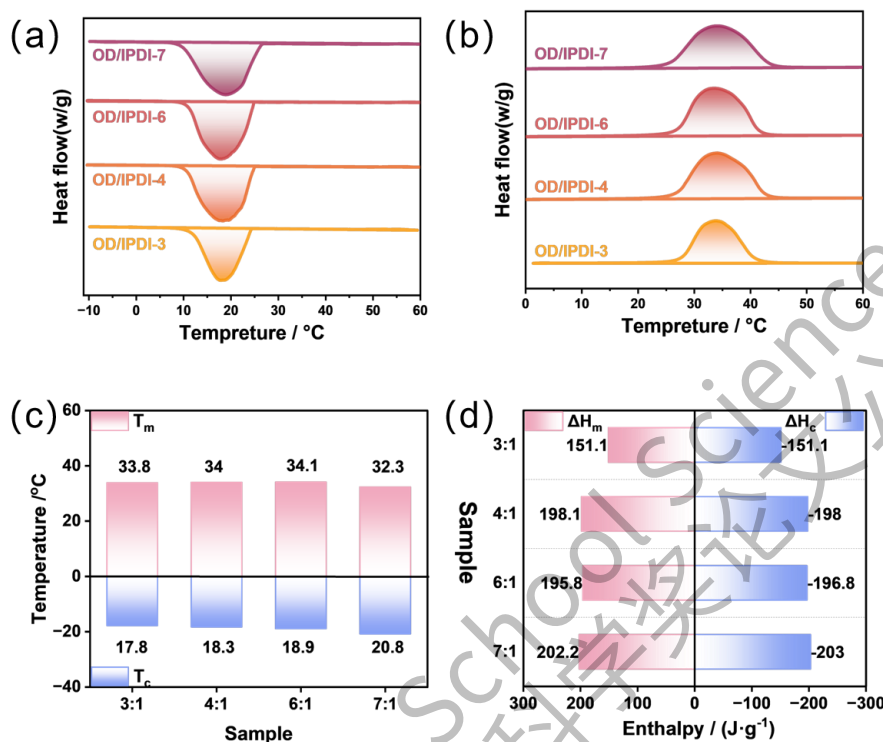


Figure 17 Thermal properties of 100μm microcapsules with different core-shell ratios: (a) DSC heating and melting curve; (b) Differential scanning calorimetry cooling and crystallization curve; (c) Melting temperature and crystallization temperature; (d) Melting enthalpy and crystallization enthalpy.

Figure 18 further reveals the reversible phase-change process of octadecane under shell confinement conditions. When the temperature rises, the ordered structure of octadecane crystals gradually destroys and absorbs latent heat during the melting process; the melted octadecane is still encapsulated in the polyurea microcavity. When cooling, the octadecane molecules rearrange themselves and undergo nucleation and crystal growth processes while releasing latent heat. Combining the DSC results at different OD/IPDI feed mass ratios, it can be seen that when the relative content of the polyurea shell increases, shell confinement is enhanced and the crystallization rearrangement is more obviously constrained; when the OD/IPDI feed mass ratio is increased, shell confinement is relatively weakened, the crystallization temperature increases, and the supercooling decreases. This forms a structure-activity mapping of core-shell composition, degree of shell confinement, crystallization behavior and heat storage response.

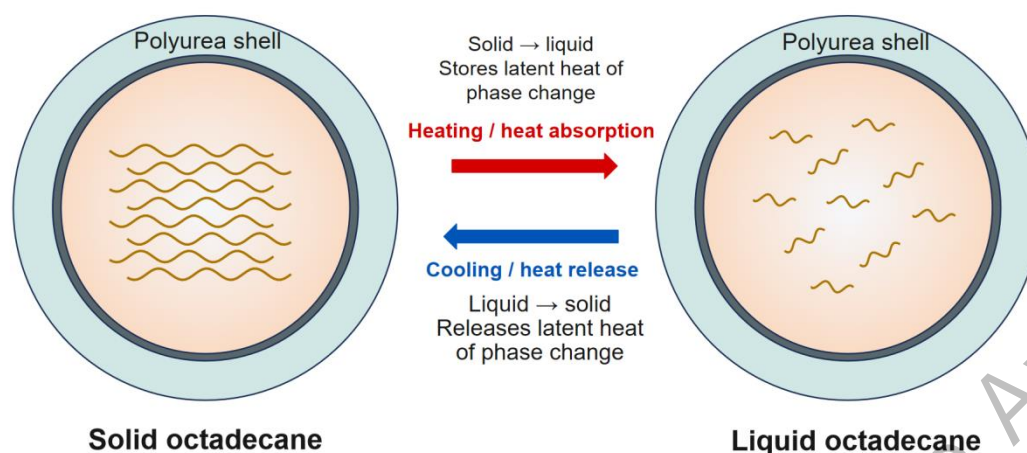


Figure 18 Schematic illustration of heat absorption and release by the octadecane core under polyurea-shell confinement.

In order to evaluate the thermal cycling stability and durability of the heat-storage performance of n-octadecane@polyurea phase-change microcapsules, 100 thermal cycles were conducted on microcapsules with particle sizes of 100, 300 and 500 μm respectively. As can be seen from Figure 19, the melting endothermic peak and crystallization exothermic peak of microcapsules of three particle sizes are still clear and complete after cycling, and the peak positions do not shift significantly. The melting temperature and crystallization temperature only show small changes, indicating that the phase-change process of microcapsules has good reversibility. At the same time, the phase-change enthalpy value did not decrease significantly, indicating that its latent heat storage capacity remained basically stable during the cycle. The above results show that the polyurea shell can withstand the volume changes caused by repeated solid-liquid phase transitions of octadecane, and its coating effect and shell confinement effect are not significantly weakened. The DSC curve remains stable before and after cycling, further indicating that there is a corresponding relationship between the stability of the microcapsule core-shell structure and the stability of the thermal energy-storage performance.

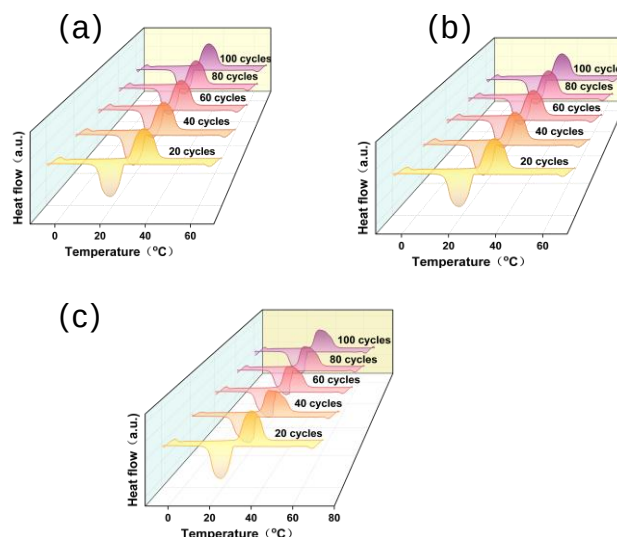


Figure 19. change curves of phase-change microcapsules after different numbers of thermal cycles: (a)100 μm ; (b)300 μm ; (c)500 μm .

4. Conclusion and Outlook

4.1 Conclusion

In response to the growing need for phase-change microcapsules with well-controlled dimensions for more efficient thermal management in electronic devices, this study developed n-octadecane@polyurea phase-change microcapsules using a microfluidic interfacial polymerization approach. The work systematically examined three connected stages of microcapsule formation and thermal storage: the stable formation of spherical droplet precursors, the formation of the polyurea shell at the oil–water interface, and the regulation of the phase-change behavior of the core material by shell confinement. The interfacial interaction mechanism underlying the formation and thermal energy storage processes of phase-change microcapsules was thereby revealed. The conclusions are summarized as follows:

(1) In confined microchannels, the capillary number is a key dimensionless parameter governing the formation of spherical droplet precursors. When the dispersed phase capillary number Ca_d is within the range of 0.0072 to 0.0216, stable droplets with uniform size and good sphericity could be obtained. Under the optimized conditions, the resulting droplets had an average diameter of 196.6 μm and a CV of only 1.88%, demonstrating a high degree of monodispersity.

(2) The formation of the polyurea shell proceeds through interfacial polymerization at the oil–water boundary. DETA in the aqueous phase migrates toward the oil–water interface and reacts with IPDI in the oil phase through step-growth addition polymerization. The resulting

polyurea gradually accumulates and deposits at the interface, eventually forming a continuous and relatively dense polymer shell around the octadecane core.

(3) Changes in the OD/IPDI feed mass ratio alter the shell confinement of the octadecane core, thereby affecting its crystallization behavior and phase-change enthalpy. As the OD/IPDI mass ratio increases from 3:1 to 7:1, the crystallization temperature of the octadecane core rises, the degree of supercooling decreases significantly, and the phase-change enthalpy increases from 151.1 J/g to 202.2 J/g. After 100 heating–cooling cycles, the DSC peak profiles and phase-change enthalpy showed no significant decrease, indicating good thermal cycling stability of the microcapsules.

Overall, this study established a controllable microfluidic interfacial polymerization method for preparing phase-change microcapsules, achieving a key transformation of interfacial polymerization reactions from the conventional disordered and random state to a dispersed and controllable state. Compared with conventional preparation methods in which droplet formation and interfacial reactions are difficult to control independently, the microfluidic approach provides a more defined reaction environment and allows the formation process to be studied and adjusted at the interface. The results demonstrate that controlling the droplet template and the interfacial polymerization conditions can influence the resulting shell structure and, in turn, the thermal behavior of the encapsulated phase-change material. These findings provide an experimental basis for further chemical design of phase-change microcapsules with tunable thermal storage properties.

4.2 Outlook

(1) The microfluidic preparation of phase-change microcapsules offers precise control over droplet size and stable performance. In the future, AI-assisted image recognition could be further incorporated into the fabrication process to provide real-time feedback on droplet formation and help optimize operating parameters. In parallel, multichannel microfluidic systems could be explored to increase production throughput while maintaining control over particle size and uniformity.

(2) Owing to the unique advantages of microfluidic-prepared phase-change microcapsules, particularly their uniform and controllable sizes, these microcapsules have broad application prospects in diverse fields, such as thermal management of AI chips and 5G/6G equipment, thermal safety of power batteries in new energy vehicles, temperature regulation in photovoltaic

and energy storage systems, low-energy buildings, and smart textiles. With further development of environmentally friendly shell materials, these microcapsules may also contribute to reducing the energy required for temperature control by storing and releasing thermal energy more efficiently. Such applications could support the broader development of low-carbon thermal-management technologies and contribute to efforts toward carbon peaking and carbon neutrality.

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Acknowledgements

This project was jointly proposed and independently completed by Zhenyu Feng and Xingyou Ai, students in the International Division of Beijing No. 80 High School. The research was conducted over a ten-month period, from November 2025 to August 2026.

The project began with a personal experience that deeply affected Zhenyu. Her grandmother became bedridden and unable to communicate verbally, making it difficult to recognize fever in time. This experience led Zhenyu, while still in middle school, to wonder whether it would be possible to develop a material that could indicate a fever visually without requiring electricity. During a group meeting at Professor Kai Pan's research laboratory at Beijing University of Chemical Technology, she was introduced to phase-change microcapsule technology for the first time. This marked the beginning of her research in the field. She subsequently developed thermally responsive phase-change microcapsules for fever indication, a project that received First Prize at the 45th Beijing Youth Science Creation Competition and First Prize at the 2025-2026 Invention Convention China National Finals.

Rather than stopping there, Zhenyu continued to explore how to overcome the limitations of conventional fabrication methods and how the technology could be applied in broader contexts. During a chemistry class taught by Ms. Heng Wang, she learned about the practical challenges of localized overheating in mobile electronic devices, such as smartphones and laptops. This prompted a new question: could phase-change materials be used not only to indicate temperature changes, but also to absorb heat from localized hotspots and mitigate overheating in electronic chips? Based on this idea, she invited Xingyou Ai, president of the Chemistry Club, to join the project. Together, we gradually extended the research toward more fundamental questions involving the precise control of interfacial polymerization and the phase-change mechanisms governed by shell confinement.

Ms. Heng Wang is Zhenyu's AP Chemistry teacher, Xingyou's A-Level Chemistry teacher, and also our grade-level coordinator. At the early stage of the project, she helped us evaluate potential research directions in light of the laboratory resources available at the two schools and worked with us to identify a feasible research pathway. After the research direction was established, we joined her Honors Research Mentorship Program, where we used our after-school time to systematically develop our skills in experimental operation, research design, and data analysis. During the manuscript-writing stage, she provided detailed feedback and guidance

through her IB academic writing course. She followed our progress throughout the entire research process and consistently helped us rethink problems when we encountered experimental bottlenecks.

We are deeply grateful to Professor Kai Pan of Beijing University of Chemical Technology for his academic guidance throughout the project. He provided systematic advice on the research rationale, mechanistic interpretation, experimental design, and overall structure of the paper. Through our discussions with him, we ultimately established microfluidic interfacial polymerization as the core technical approach of this study.

During the experiment, we regularly discussed with Dr. Ke Chen at the Beijing University of Chemical Technology laboratory both the preparation of n-octadecane@polyurea microcapsules and core technical knowledge including thermal properties and cyclic stability. In addition, we conducted self-directed learning and practice through literature research. In accordance with the laboratory's safety protocols, Yi Li, a Master student, waited with us until each experiment had been completed and all equipment had been cleaned and properly stored before leaving the laboratory. He personally demonstrated the operation and measurement procedures for major analytical instruments, including scanning electron microscopy, Fourier-transform infrared spectroscopy, X-ray diffraction, and differential scanning calorimetry. During the 100-cycle heating-and-cooling stability tests, when the instruments operated continuously day and night, he remained available and accompanied us during experiments that extended late into the night.

All of the guidance and assistance described above was provided voluntarily and without compensation. The experimental procedures, collection and recording of raw data, and manuscript preparation were independently completed by us.

We had a clear division of responsibilities, which allowed us to complement each other's strengths and work in a coordinated manner. Zhenyu was responsible for topic selection and the preliminary literature review, as well as the CFD simulations used to investigate the factors governing the stable formation of spherical droplets and the microcapsule formation process at different OD/IPDI feed mass ratios. Xingyou was responsible for validating droplet formation using a machine-vision system and for investigating the thermal properties and cyclic thermal stability of the microcapsules. The initial manuscript was drafted by Zhenyu and then supplemented and revised by Xingyou. Finally, we jointly revised and finalized the paper.

Three major challenges emerged during the experimental process. At the beginning of the study, the precursor droplets were difficult to maintain as stable spheres. After extensive literature review and repeated optimization of flow rates and interfacial conditions, we precisely controlled the capillary number of the dispersed phase within the range of 0.0072–0.0216, enabling the stable generation of spherical precursor droplets. We also found that manually counting microcapsules was inefficient and prone to measurement error. To address this problem, we introduced a real-time visual analysis system for microfluidic microspheres, which enabled real-time acquisition and automated analysis of particle-size data. Finally, because the original preparation procedure was relatively complicated and difficult to operate continuously and consistently, we repeatedly optimized the process parameters. Through these iterations, we successfully obtained monodisperse microcapsules with an average particle diameter of 196.6 μm and a particle-size coefficient of variation (CV) of only 1.88%.

Statement on the Use of AI: Any AI-related technologies used in this project were discussed with and approved by our supervising teachers in advance, and all AI-assisted results were individually reviewed and verified by us. The microfluidic microsphere real-time visual analysis system used in this study (V1.0; Software Copyright Registration No. 2026SR0625514) was provided by Beijing University of Chemical Technology. Developed using OpenCV-based image-processing algorithms, the system can identify the size, morphology, and number of droplets and microcapsules in real time and automatically calculate parameters including formation frequency, volume distribution, and coefficient of variation (CV). It was used solely as an experimental monitoring and data-analysis tool and does not constitute generative AI.

Finally, we sincerely thank the Beijing Youth Talent Program for providing the research platform, and all the teachers from both schools for their careful guidance. For us, this project is not an endpoint, but a beginning. In future work, we hope to further optimize the structure and composition of polyurea shell materials to achieve more precise control over phase-change properties. At the process level, we plan to integrate online microscopic image recognition and process monitoring to improve droplet-formation control and explore multichannel approaches for scalable and continuous production. At the application level, we hope to extend this technology to areas such as low-energy building temperature regulation and thermal management of power batteries, contributing to more energy-efficient and low-carbon technologies.