

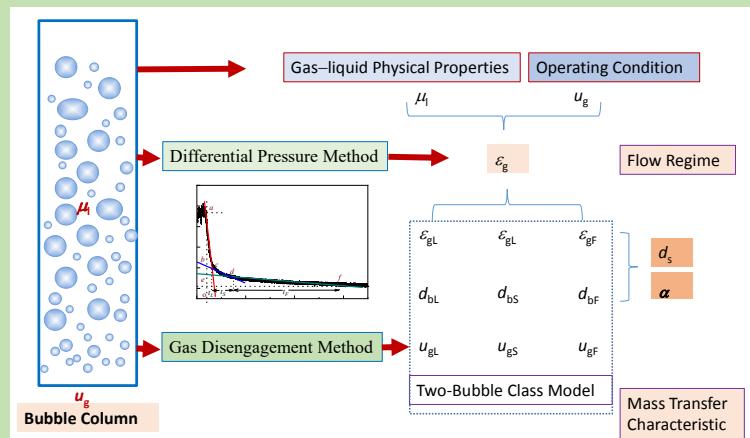
## Hydrodynamic study on a gas–liquid bubble column with high viscosity SEBS solution

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**Abstract:** For development and design of SBS hydrogenation reactor, the hydrodynamic behavior of high viscosity solution in gas–liquid bubble column using SEBS-1650 hexane solution as liquid phase was studied by differential pressure method and gas disengagement method. The effects of viscosity on gas holdup of solution with low surface tension, gas holdup of large and small bubbles, rise velocity of large and small bubbles and specific surface area were investigated. With the increase of viscosity, gas holdup decreased significantly, and flow pattern directly transited turbulent regime. From the curve of gas disengagement method, three types of bubbles were identified in bubble column: large bubbles, small bubbles and fine small bubbles. The gas holdup of small bubbles and fine small bubbles gradually decreased with the increase of viscosity. Viscosity has a slight influence on the rise velocity of large and small bubbles, and the specific surface area decreased significantly with the increase of viscosity. Based on the experimental results, the formulas for calculating gas holdup of large and small bubbles and average gas holdup were listed. These results provided some necessary and reliable basic data for designing and developing a SBS hydrogenation reactor.

**Key words:** gas holdup; large and small bubble; bubble column; bubble rising velocity; SEBS



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# SEBS 高黏度溶液气液鼓泡塔的流体力学研究

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**摘要:** 针对 SBS 加氢反应器开发与设计, 以 SEBS-1650 己烷溶液为液相, 采用差压法和床层塌落法研究了气液鼓泡塔中高黏度溶液的流体力学行为, 考察了黏度对低表面张力溶液的气含率、大小气泡气含率、大小气泡上升速度和比表面积等因素的影响。结果表明, 随黏度增加, 大气泡增多, 气含率明显降低, 塔内流型处于湍流区; 由床层塌落曲线确定鼓泡塔内存在三种类型的气泡: 大气泡、小气泡及细小气泡, 随黏度增加, 小气泡与细小气泡逐渐减少; 黏度对大小气泡的上升速度略有影响, 比表面积随黏度增加而明显降低。根据实验结果给出了大小气泡气含率与平均气含率的计算公式。

**关键词:** 气含率; 大小气泡; 鼓泡塔; 气泡上升速度; 改性热塑性弹性体

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## 1 前言

丁二烯和苯乙烯嵌段共聚物为 SBS 热塑性弹性体, 兼有橡胶和塑料的特性。针对 SBS 橡胶段聚丁二烯不饱和双键造成的易老化、耐热性差等缺点, 国内外开发了 SBS 选择加氢制得的新型改性热塑性弹性体(SEBS)工艺, 与 SBS 产品性能相比, SEBS 具有优异的耐老化性、耐臭氧、耐热性和稳定性等, 可广泛用于高档弹性体、树脂改性、胶黏剂、润滑油增黏剂、绝缘材料、航空部件等<sup>[1-3]</sup>。SBS 溶液黏度较大, 与已有气液研究体系性质有明显区别, 其流体力学性质也有较大差异。因此, 研究气液鼓泡塔中高黏度溶液的流体力学行为, 对设计和开发合理的 SBS 加氢反应器非常重要。

近几十年来, 国内外开展了广泛的气液鼓泡塔内的实验研究与模拟, 考察了鼓泡反应器流体力学和传质特性<sup>[4-10]</sup>。黏度是液相重要的物理性质之一, 影响各相流动与传质性能。关于黏度对鼓泡塔内流体力学参数的影响, 国内外学者进行大量研究。一般采用油酸钠或表面活性剂改变水的表面张力, 用甘醇或甘油改变水的黏度, 或采用醇类的水溶液。Xing 等<sup>[11]</sup>通过实验研究和数值模拟研究了黏度对鼓泡塔内流体力学行为的影响, 研究对象为甘油溶液, 表面张力较大。Ruzicka 等<sup>[12]</sup>用不同浓度和黏度的甘油水溶液考察了黏度对鼓泡塔内均匀状态到非均匀流状态的影响。Losi 等<sup>[13]</sup>研究了不同浓度的石蜡油体系, 黏度达  $0.804 \text{ Pa}\cdot\text{s}$ , 表面张力仅为  $0.026 \text{ N/m}$ 。Kajero 等<sup>[14]</sup>研究了黏度  $0.005\sim5 \text{ Pa}\cdot\text{s}$  的硅油在小直径鼓泡塔中对流型和气泡行为的影响, 随液体黏度增加, 栓塞流的发生率呈扩大趋势; 随液体黏度增加, 液

体团块中的液体空隙率降低。Sharaf 等<sup>[15]</sup>研究了黏度高达  $1.7 \text{ Pa}\cdot\text{s}$  的静态液体中气泡动力学的影响, 发现气泡动力学高度依赖于密度和黏度比, 通过模拟和实验进行了验证。以上文献中大部分实验体系为改性水的溶液体系, 与实际有机溶液差别很大。研究表明液相黏度的影响有双重效应<sup>[16,17]</sup>: 随液相黏度增加, 一方面气相聚并加强, 气泡平均直径增加, 气含率降低; 另一方面, 黏度增加使气泡上升曳力系数增加, 上升速度减小, 气含率增加, 随黏度增加气含率先增加后减小, 存在一个极大值。可将黏度影响分为 3 个范围: 低黏度( $\mu < 3 \text{ mPa}\cdot\text{s}$ )时, 气含率随黏度增加而增加<sup>[18-20]</sup>; 中黏度( $3 \text{ mPa}\cdot\text{s} < \mu < 12 \text{ mPa}\cdot\text{s}$ )时, 气含率基本为定值<sup>[18-20]</sup>; 高黏度( $\mu > 12 \text{ mPa}\cdot\text{s}$ )时, 随黏度增加, 气含率不断降低<sup>[21-27]</sup>。Ruzicka 等<sup>[12]</sup>发现  $0\sim3 \text{ mPa}\cdot\text{s}$  时气含率随黏度增加而增加, 但在  $3\sim22 \text{ mPa}\cdot\text{s}$  范围内气含率开始降低; Olivieri 等<sup>[28]</sup>在黏度范围  $1\sim117 \text{ mPa}\cdot\text{s}$  内使用海藻酸钠水溶液研究了液体黏度的影响, 结果表明在  $\mu=4.25 \text{ mPa}\cdot\text{s}$  时气含率最大, 然后随黏度增加, 气含率开始下降。现有文献大部分实验体系通过改性水进行, 这与实际过程的有机溶液有很大差别, 特别是在高黏度低表面张力溶液相关的研究工作较少。

本工作采用 SEBS 溶液研究高黏度溶液在鼓泡塔内的流体力学行为, 研究体系中的 SEBS 己烷溶液具有高黏度、低表面张力的性质, 与已有研究体系有很大差别。通过差压法和动态气体逸出法测量反应器内气含率、大小气泡的气含率及上升速率, 考察了反应器内流体力学行为, 为该反应器的开发和应用提供了基本化工数据。

## 2 实验

### 2.1 实验装置与物料

实验装置如图1所示,鼓泡塔为内径165 mm,外径175 mm,高3000 mm的玻璃塔。玻璃塔外有加热带,由温控仪进行控温。塔底部有分布器,实验采用两种分布器,分别是盘管式气体分布器,分布器中盘管的直径为6 mm,盘面上均匀分布37个孔( $\Phi 1.5$  mm),开孔率为0.31%,开口向上,中间进口直接接入进气口,远端封闭。盘面直径约130 mm,安装在塔底的封头内,居底部120 mm;单管分布器( $\Phi 18$  mm),开孔率1.19%。塔下半部分间隔0.85 m的位置设有两个测压口,测压口分别接差压变送器的高低两端,测量不同条件下测压口间的差压值。实验初始静液位高度为1075 mm,表观气速为0.03~0.25 m/s,采用氮气为气相,气体由储气罐供给,经调节阀、流量计、气体分布器进入鼓泡塔,从塔顶流出。

采用SEBS-1650和己烷混合溶剂(20%正己烷,80%环己烷)的混合液为液相,具体参数见表1。文献[29]给出了SBS与环己烷溶液和苯溶液的黏度,含SEBS8%以下的溶液呈现牛顿流体性质,黏度基本不随剪切速率改变。

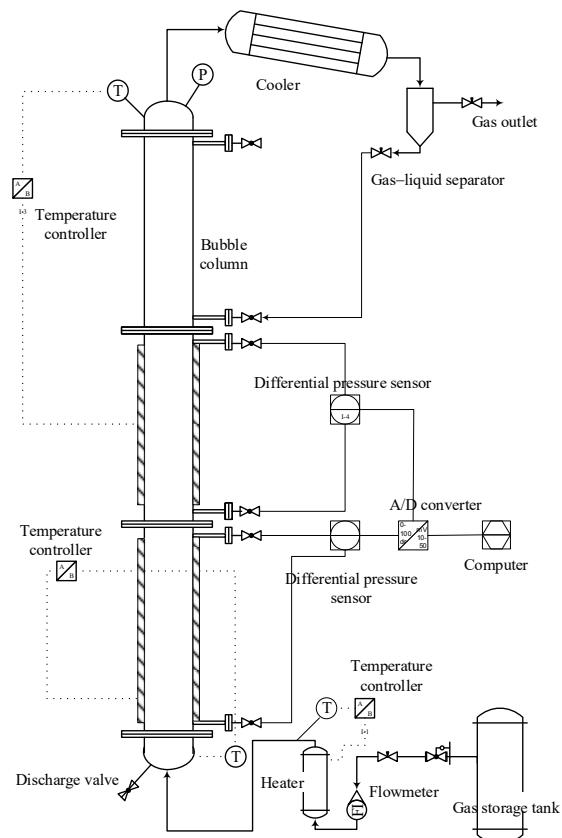


图1 实验装置图  
Fig.1 Diagram of experimental setup

表1 溶液的物理性质  
Table 1 Physical properties of solutions

No.	Solution	Density/(kg/m <sup>3</sup> )	Viscosity/(Pa·s)	Surface tension/(N/m)
H <sub>2</sub> O	Water	998	0.001	0.072
SEBS1	0.50% SEBS-1650 and cyclohexane	767	0.021	0.014
SEBS2	1.50% SEBS-1650 and cyclohexane	778	0.034	0.015
SEBS3	1.90% SEBS-1650 and cyclohexane	773	0.039	0.015
SEBS4	4.20% SEBS-1650 and cyclohexane	773	0.075	0.015

### 2.2 实验数据采集与处理

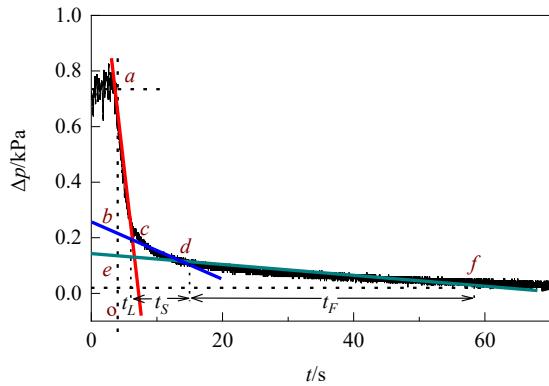
气含率的测量<sup>[30]</sup>:对气液两相体系气含率采用双引脚测量方法,传感器测得为AB两点间(距离0.85 m)的压差,ε<sub>g</sub>为AD间的平均气含率,忽略气体项,气含率为

$$\varepsilon_g = \frac{p_B - p_A}{\rho_l g \Delta H} = \frac{\Delta p}{\rho_l g \Delta H} \quad (1)$$

式中,Δp为两点间压差(kPa),ρ<sub>l</sub>为液相密度(kg/m<sup>3</sup>),g为重力加速度(m/s<sup>2</sup>),ΔH为测压点间距离(m)。Δp由差压传感器测得,为一段时间(60 s)测量的平均值,压差计采集频率为106 Hz。

大小气泡的气含率采用床层塌落法<sup>[30]</sup>(Dynamic Gas Disengagement, DGD)测得。正常运行的鼓泡床反

器中,瞬间切断气源,记录床层塌落过程中采压点间的压力信号随时间的变化曲线,根据压力信号变化曲线对鼓泡床中各流体力学特性(气含率、气泡尺寸及分布、气泡上升速度等)进行估计,大气泡先释放,然后是小气泡,大小气泡气含率从差压曲线中得出,相对误差为±5%。图2为SEBS己烷溶液在表观气速0.036 m/s时差压与时间的变化关系塌落曲线。ac段为大气泡逸出过程,c点时大气泡全部逸出,此时小气泡均匀通过差压计两引脚,ab段为大气泡产生的压差,根据式(1)可计算大气泡气含率ε<sub>gL</sub>;cd段为小气泡下端面经过两引脚之间的过程,be段为小气泡产生的压差,由式(1)计算小气泡气含率ε<sub>gS</sub>;df段为细小气泡均匀通过的过程,eo段对应细小气泡的差压,可计算出细小气泡气含率ε<sub>gF</sub>。

图 2  $N_2$ -SEBS 溶液塔落曲线(SEBS1)Fig.2 Curve of gas disengagement in  $N_2$ -SEBS solution

床层开始塌落, 两测压点间的压力开始下降, *a* 点大气泡开始释放, 当其下端面到达上测压点时, 两测点

间的大气泡气含率为零, 反映在曲线 *c* 点, *ac* 两点间的时间差  $t_L$  即为大气泡走过两测压点间的距离所需的时间, 两测压点距离  $\Delta H$  与时间  $t_L$  之比即为大气泡上升速度  $u_{bL}$ 。*ad* 两点间的时间差  $t_S$  和 *df* 两点间的时间差为  $t_F$ , 即为小气泡和细小气泡走过两测压点所需的时间, 两测压点间距离  $\Delta H$  与时间  $t_S$  或  $t_F$  之比即为小气泡(细小气泡)上升速度  $u_{bS}(u_{bF})$ , 大小气泡的上升速度相对误差为  $\pm 15\%$ 。

**比表面积<sup>[31,32]</sup>:** 气液比表面积  $\alpha$  与气泡尺寸密切相关, 利用气泡上升速度与气泡尺寸的关系式, 可由气泡终端上升速度估计气泡尺寸。文献[33]对气泡的形成和特征进行了综述, 归纳了几个关系式关联/预估气泡尺寸, 如表 2 所示。

表 2 气泡大小与气泡上升速度之间的关系式  
Table 2 Correlations for estimating bubble size from bubble rise velocity

Reference	Correlation	Range of applicability	Bubble size
Mendelson <sup>[34]</sup>	$u_b = \sqrt{\frac{2\sigma}{d_b \rho_l} + \frac{gd_b}{2}}$	$782 \text{ kg/m}^3 < \rho < 1480 \text{ kg/m}^3$ $0.02 \text{ N/m} < \sigma < 0.72 \text{ N/m}$ $0.00052 \text{ Pa}\cdot\text{s} < \mu < 18 \text{ Pa}\cdot\text{s}$	$d_{bL}, d_{bS}$
Jamialahmadi, et al <sup>[35]</sup>	$V_{sp} = \frac{\Delta\rho g d_b^2}{18\mu} \left( \frac{3\mu_l + 3\mu_g}{2\mu_l + 3\mu_g} \right)$ $V_w = \sqrt{\frac{2\sigma}{d_b(\rho_l + \rho_g)} + \frac{gd_b}{2}}$ $u_b = \frac{V_{sp} V_w}{\sqrt{V_{sp}^2 + V_w^2}}$	$626 \text{ kg/m}^3 < \rho < 1071 \text{ kg/m}^3$ $0.016 \text{ N/m} < \sigma < 0.72 \text{ N/m}$ $0.00022 \text{ Pa}\cdot\text{s} < \mu < 0.031 \text{ Pa}\cdot\text{s}$	$d_{bL}, d_{bS}$
Lehrer <sup>[36]</sup>	$u_b = \sqrt{\frac{3\sigma}{d_b \rho_l} + \frac{gd_b(\rho_l - \rho_g)}{2\rho_l}}$	$782 \text{ kg/m}^3 < \rho < 1480 \text{ kg/m}^3$ $0.02 \text{ N/m} < \sigma < 0.72 \text{ N/m}$ $0.00052 \text{ Pa}\cdot\text{s} < \mu < 18 \text{ Pa}\cdot\text{s}$	$d_{bL}$
Talaia <sup>[37]</sup>	$u_b = 0.694 \left[ \frac{gd_b(\rho_l - \rho_g)}{\rho_l} \right]^{0.5}$	$3425 < Re < 7490$ $25.5 \text{ cm/s} < u_b < 32.9 \text{ cm/s}$ $803 \text{ kg/m}^3 < \rho < 1420 \text{ kg/m}^3$	$d_{bL}$
Nickens, et al <sup>[38]</sup>	$u_b = 0.361 \sqrt{gd_b} (1 + 4.89/E_0)^{0.25}$	$0.022 \text{ N/m} < \sigma < 0.77 \text{ N/m}$ $0.0008 \text{ Pa}\cdot\text{s} < \mu < 10.9 \text{ Pa}\cdot\text{s}$	$d_{bS}, d_{bF}$
Krishna, et al <sup>[39]</sup>	$u_b = 0.62 \sqrt{gd_b} (\text{SF})$	$SF = 1, d_b/D_c < 0.07$ $SF = 1.1e^{-1.55d_b/D_c}$ $0.07 < d_b/D_c < 0.4$ $SF = 0.38(d_b/D_c)^{1/2}, d_b/D_c > 0.4$	$d_{bF}$

通过 DGD 获得气泡上升速度, 可根据表 2 中的关联式估计出气泡尺寸, 取平均值, 用于比表面积的计算。表 2 中的关联式具有一定的适用范围, 部分关联式下限范围接近本实验的范围, 可用于定性研究黏度对传质的

影响。气液比表面积为

$$\alpha = \sum_{i=1}^{i=N} \varepsilon_{gi} / d_{bi} \quad (2)$$

式中,  $\varepsilon_{gi}$  为每一级气泡的气含率,  $d_{bi}$  为气泡尺寸。

### 3 结果与讨论

#### 3.1 黏度对气含率的影响

图3为SEBS己烷溶液与水溶液平均气含率随表观气速的变化。与水-空气体系相比, N<sub>2</sub>-SEBS环己烷溶液体系的平均气含率有明显下降趋势, 因为黏度增加, 气泡聚并机会增加, 不易破碎, 引起气含率下降。许多研究和方法涉及流型识别, 其中漂移通量模型是一种经典的分析流型方法<sup>[31]</sup>:

$$j = \varepsilon_g (1 - \varepsilon_g) \left( \frac{u_g}{\varepsilon_g} \pm \frac{u_l}{\varepsilon_l} \right) \quad (3)$$

式中,  $j$ 为漂移通量,  $\varepsilon_g$ 为气含率,  $\varepsilon_l$ 为液含率,  $u_g$ 为表观气速(m/s),  $u_l$ 为表观液速(m/s)。本实验液体为间歇操作,  $u_l=0$ 。

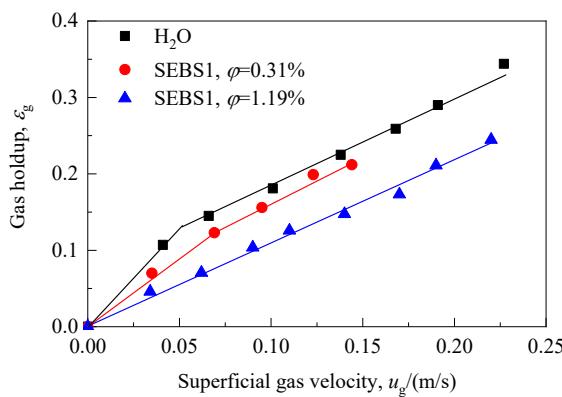


图3 SEBS溶液与水溶液的气含率随表观气速的变化  
Fig.3 Variations of gas holdup of SEBS solution and water solution with superficial gas velocity

图3实验结果用流量漂移方法表示, 如图4所示。

一般体系下, 塔内存在三种流型: 均匀流、过渡流和湍流, 从图4可看出, N<sub>2</sub>-SEBS环己烷溶液体系在实验表观气速下仅存在均匀流和湍流, 低气速的范围内已有大量大气泡生成, 出现了气泡聚并与破碎。Olivieri等<sup>[28]</sup>和Rabha等<sup>[40]</sup>指出中等黏度的体系中均匀鼓泡区已经存在不稳定现象, 高黏度溶液中均匀鼓泡区已经监测不到, 大气泡的出现可将湍流区提前。实验中气泡照片如图5所示, 从图可看出, 溶液中出现大量直径大于20 mm的大气泡, 伴随聚并与破碎现象, 除了小气泡外, 还存在大量直径小于1 mm的细小气泡, 与图2塌落曲线一致。双气泡模型中部分学者根据实验现象划分气泡当量直径, 如细小气泡  $d_b < 1$  mm 和大气泡  $d_b > 20$  mm<sup>[41]</sup>, 小气泡  $1 \text{ mm} < d_b < 10$  mm 和大气泡  $10 \text{ mm} < d_b < 150$  mm<sup>[20]</sup>, 小气泡  $0.7 \text{ mm} < d_b < 10$  mm 和大气泡  $d_b > 10$  mm<sup>[27]</sup>及大气泡  $d_b = 40 \sim 45$  mm<sup>[40]</sup>。高黏度低表面张力溶液中, 根据实验结果将气泡归为3种类型: 大于10 mm的大气泡, 1~10 mm的小气泡及小于1 mm的细小气泡。

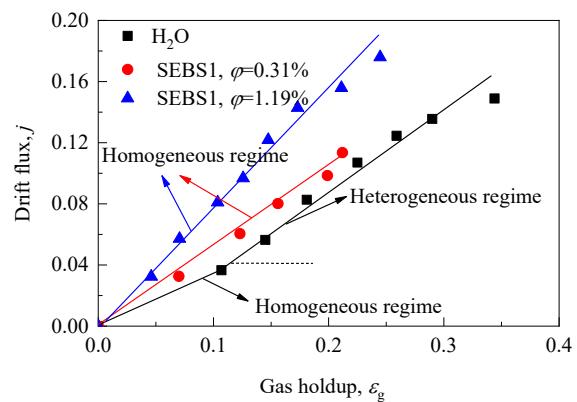


图4 漂移通量与气含率的关系  
Fig.4 Relationship between drift flux and gas holdup

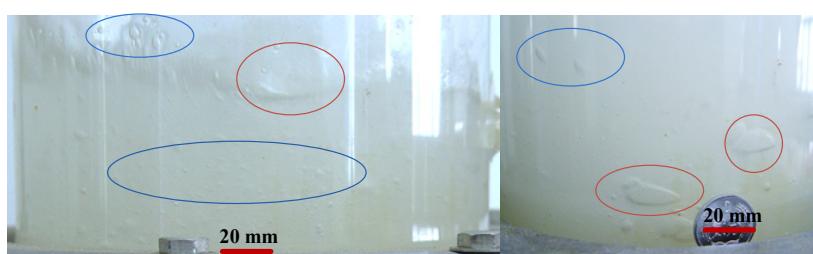


图5 实验中气泡照片( $u_g=0.036$  m/s, SEBS1)  
Fig.5 Bubble photographs in the experiment ( $u_g=0.036$  m/s, SEBS1)

图6为不同黏度下SEBS-1650己烷溶液气含率随表观气速的变化。从图可看出, 随表观气速增加, 气含率增加。随液相黏度增加, 曲线斜率明显减小, 表明相

同表观气速下随液相黏度增加, 气含率变小, 这是因为黏度增加促进了气泡聚并, 使气泡尺寸增加, 气含率降低。根据实验现象, 黏度较大的体系中, 大气泡的数量

增加,小气泡的数量较少,但细小气泡(直径小于1 mm)有增多趋势,这部分细小气泡相界面较大,对反应体系传质速率有很大影响。

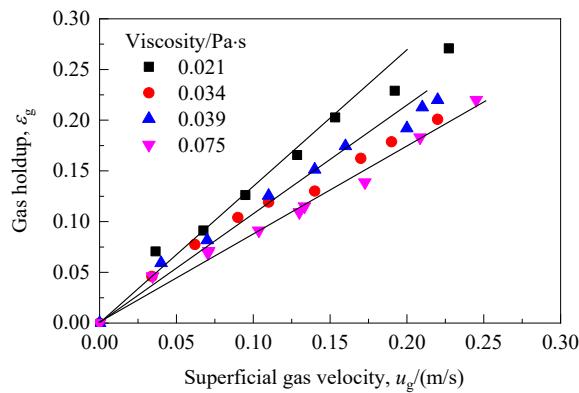


图6 不同黏度下SEBS-1650己烷溶液气含率随表观气速的变化

Fig.6 Variations of gas holdup with superficial gas velocity in SEBS-1650 cyclohexane solution under different viscosities

### 3.2 大小气泡的气含率

SEBS 溶液中气泡可分为大气泡、小气泡和细小气泡。图 7 为黏度 0.021 Pa·s 溶液中的大小气泡气含率随表观气速的变化, 随气速增加, 大气泡气含率增加, 小气泡和细小气泡气含率略有增加。

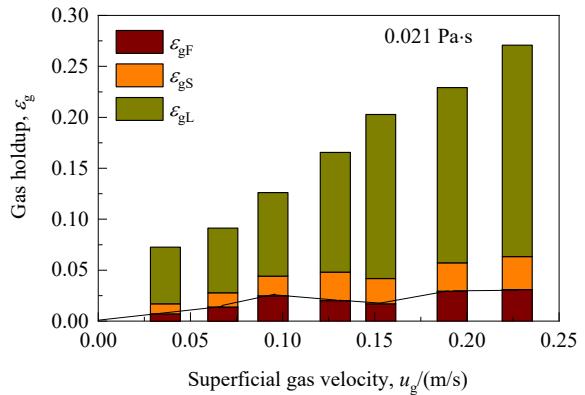


图7 大小气泡气含率随表观气速的变化

Fig.7 Variations of gas holdup of large and small bubbles with superficial gas velocity

图 8 为大小气泡气含率随黏度的变化。从图可看出,  $u_g=0.10$  m/s 时, 随黏度增加, 大气泡气含率略有增加, 小气泡和细小气泡气含率降低。这是因为大气泡以活塞流的形式上升, 小气泡螺旋式上升, 且其螺线半径随黏度增加而增加, 小气泡聚并成大气泡的机率增加, 而大气泡受液相物性影响较小, 发生聚并和破裂的概率不会随黏度改变而改变, 所以床层内大气泡气含率随黏度增加略有增加, 而小气泡分率则明显降低。根据文献 [9,16,42] 可筛选出能较准确预测高黏度下大小气泡气含率的计算式。

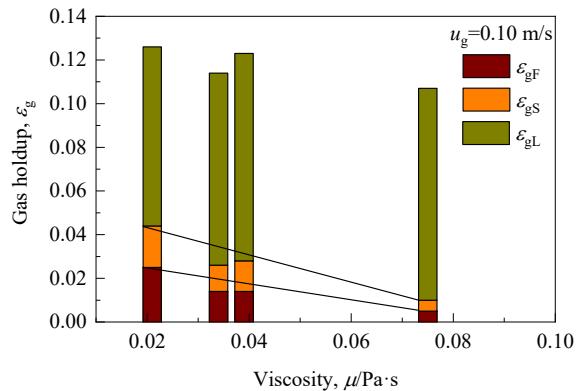
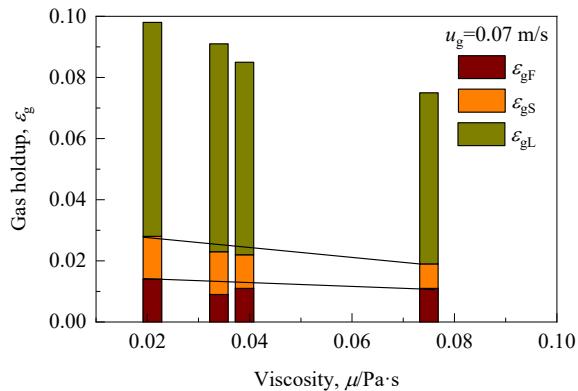


图8 大小气泡气含率随黏度的变化  
Fig.8 Variations of gas holdup of large and small bubbles with viscosity

$$\varepsilon_{gL} = 0.785 u_g^{0.642} \left( \frac{\mu_l}{\mu_0} \right)^{-0.125} \quad (4)$$

$$\varepsilon_{gS} = 0.184 u_g^{0.153} \left( \frac{\mu_l}{\mu_0} \right)^{-0.640} \quad (5)$$

$$\varepsilon_g = 0.26 u_g^{0.54} \mu_l^{-0.147} \quad (6)$$

$$\varepsilon_g = 0.171 u_g^{0.60} \mu_l^{-0.22} D_c^{-0.15} \quad (7)$$

实验范围内采用上述公式计算大小气泡气含率和平均气含率, 计算值与实验值的误差基本在±20%以内, 个别点在+30%以内, 结果如图 9 所示。式(6)和(7)实验体系为水改性体系, 即用油酸钠表面活性剂改性表面张力, 用三甘醇或甘油改性水的黏度, 这与实际体系存在

的差别,从图 9 可看出,均匀鼓泡区计算值比实验值大,表明水改性体系存在均匀鼓泡区,气含率随气速增加而线性增加,本研究的黏度体系大小气泡处于非均匀鼓泡

区,气含率降低。在湍流区(右侧),相比于文献[9,42]水改性体系,黏度大的溶液形成气泡尺寸较大,气含率实验值比经验式的计算值小。

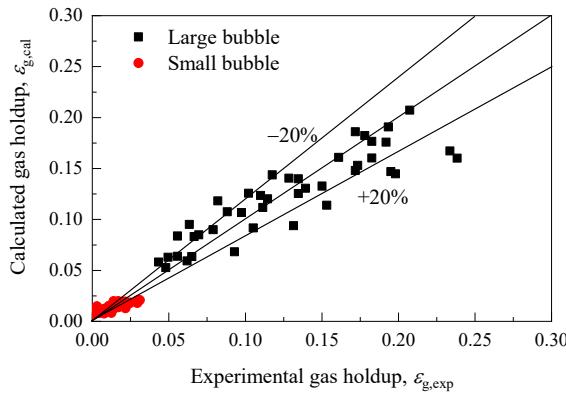


图 9 大小气泡气含率计算值与实验值的对比

Fig.9 Comparisons between calculated and experimental values of gas holdup of large and small bubbles

### 3.3 大小气泡上升速度

图 10 为大小气泡上升速度随表观气速的变化。从图可看出,随黏度增加,大小气泡速度均有不同幅度增加,这是因为黏度增加,平均气泡上升速度增加,平均气泡尺寸增加,床层内生成更多气泡,大气泡气含率增加,小气泡气含率略有降低。相同黏度下,随表观气速升高,大小气泡速度略有增大,平均气泡速度略有增加,在 SEBS 溶液中大小气泡尺寸未发生明显变化。

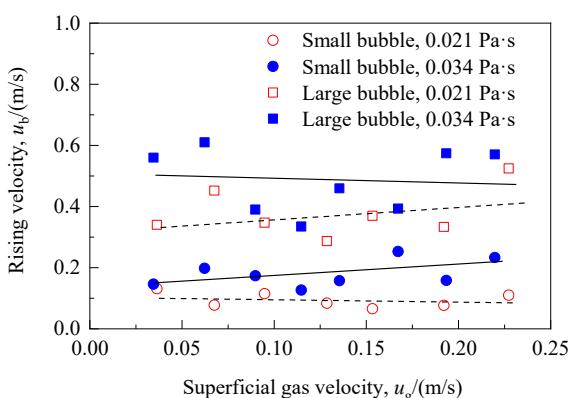


图 10 大小气泡上升速度随表观气速的变化

Fig.10 Variations of rising velocity of large and small bubbles with superficial gas velocity

根据实验得到的大小气泡上升速度,由表 2 中的计算式可计算出相应的气泡尺寸,得到该条件下的比表面积。大气泡对气液传质的贡献较小,而小气泡和细小气泡对传质的贡献大。不同黏度下比表面积随表观气速的

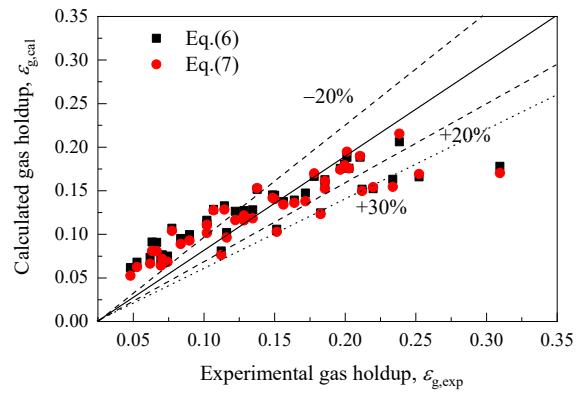


图 11 不同黏度下比表面积随表观气速的变化

Fig.11 Variations of specific surface area with superficial gas velocity under different viscosities

## 4 结 论

采用 SEBS 溶液研究高黏度溶液在鼓泡塔内的流体力学行为,得到如下结论:

(1) 在本实验操作范围内,采用雷诺数进行判定,发现鼓泡塔内提前处于湍流区,随黏度增加,气泡聚并

加剧, 气含率呈降低趋势, 给出了计算气含率的适宜关联式。

(2) 由床层塌落曲线, 确定鼓泡塔内存在3种气泡: 大气泡(大于10 mm)、小气泡(1~10 mm)和细小气泡(小于1 mm)。随表观气速增大, 碰撞和破碎加剧, 小气泡和细小气泡的气含率略有增加; 随黏度增加, 小气泡与细小气泡逐渐减少。

(3) 黏度对大小气泡的上升速度略有影响, 大小气泡的平均直径未发生明显变化, 气液比表面积随黏度增加明显降低。

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