Viscosity effects in High-Gradient Magnetic Separation Technique

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ABSTRACT

High-Gradient Magnetic Separation is a method that has been used for decades to purify aqueous low viscous media with great success. The application of magnetic carrier beads for bioprocessing has many advantages; not only for selective separation of the target protein from the biomass but also it can be applied as enzyme carrier particle to catalyse a polymeric synthesis. The advantages of the magnetic process are low energy consumption, no solvent application and a very stable particle for recycling. However the viscosity of the fluid affects the process significantly and it is the aim of this work to find out where the limits are.

KEYWORDS
Magnetic Filters, Magnetic Separators, Purifiers, Simulation, Metal Meshes

1. Introduction

The production of surfactant bio-polymer by enzyme catalysis with renewable natural substrates has an important impact on oil independence and carbon footprint reduction of bulk chemicals [2]. High-viscosity substrate (polyols and fatty acids from renewable resources) are used to produce ester-based surfactants. Fixed bed processes with porous columns are not suitable as the pressure drop is too high. Hence meso-structured silica beads are applied as enzyme-carrier particles. After the bioprocess the particles are separated totally for carrier reuse and to avoid the degradation of the product. The high viscosity of the fluid is up to 1000 mPa s depending on the reaction condition (temperature, substrate nature) and it is therefore the main challenge for the treatment. Conventional separation processes involve a dilution of the fluid, in order to decrease the viscosity. As for economic and ecological reasons the usage of solvents is undesirable, a new separation pathway has to be found.
The HGMS principle (**H**igh-**G**radient **M**agnetic **S**eparation) utilizes magnetic force which is induced through the distortion of an external magnetic field by ferromagnetic metal meshes. Next to the wire, high magnetic forces occur, causing magnetizable particles to adhere. The filtration effect of a HGMS apparatus has similarities with deep bed filtration, although the principle has important benefits. The deposition of particles is mainly based on the magnetic force which is effective even at a great distance from the wire. In addition, the magnetic force ensures the separation of small magnetizable particles of micro- and submicrometer scale [1]. The pore volume of the filter matrix is therefore significantly higher than a common deep bed filter which decreases the filtration resistance significantly.

The work presents the performance of the magnetic separator regarding the impact of the viscosity on the separation efficiency as a function of the flow and the magnetic field strength. As result experimentally gained data are shown and compared to

theoretical gained values. This enables the precise engineering of new filters and eases the scale-up of existing devices.

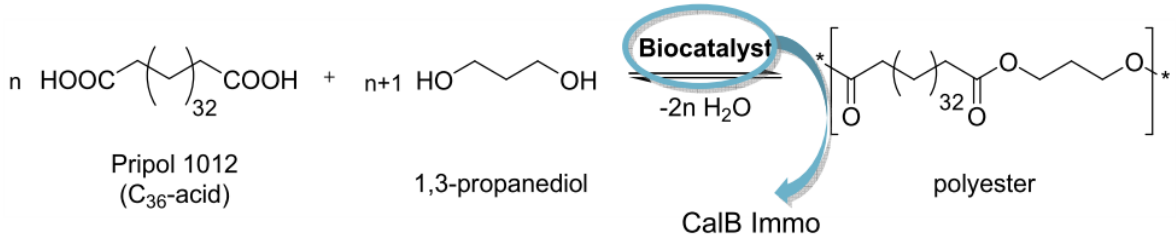
2. Materials and Method

Magnetizable particles (Type A1, B1, C1 and D1) of two different particle matrix material, morphology, particle size and magnetization were selected in order to test their separation efficiency. The characterization of the physical properties was necessary to prove their relationship to bead's separation quality. The magnetic force depends on the particle size and magnetization as well as on the viscosity of the fluid. Furthermore the solid density, primary size of the particles and surface area are important parameter influencing the separation efficiency. The particle size distribution is determined by laser diffraction HELOS, the solid density is characterized by multivolume gas pycnometer Modell MP 1305 with helium for A1 and B1 and liquid pycnometer for C1 and D1. The BET surface area is measured by Multipoint method; sample preparation included degassing of the material at 150°C for 20 min. The magnetization curves measurements were conducted at IFG, KIT Campus North. The maximal magnetization is determined at 8E6 A/m. Remanence and coercitivity characterize the degree of deviation from super-paramagnetism. The main characteristics are listed in Table 1.

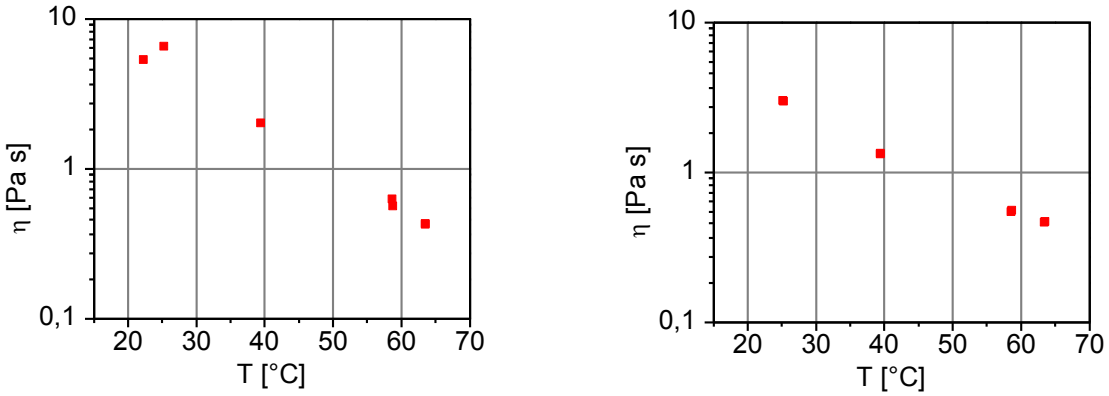
Table 1: Particle characteristics

Type:	A1	B1	C1	D1
Matrix material	Silica	Silica	Silica	PVA
particle size x_{50} [µm]	4	2,1	2,00	3,16
solid density [g/cm³]	3,46	3,91	3,3	2,1
BET [m²/g] (Multipoint)	32,07	4,662	13,78	18,536
max. magnetization [Am²/kg]	39,44	41,34	80,53	32,46
remanence [Am²/kg]	9,7	6,81	29,49	0,24
coercitivity [kA/m]	11,93	10,44	12,74	0,43
Primary particle size BET [nm]	54,1	329,2	131,9	154,1

The reaction of a polymerised fatty acid (C36-acid) with 1,3-propanediol on a lipase carrier system under dehydration is a solvent free process that produces polyester [3].



As viscosity of the fluid is also crucial for the applied magnetic force on particles, the viscosity of the substrate (Pripol 1012 from Croda) and the polyester were characterized by Applied Mechanics group (AME) at KIT.



a) b)

Figure 1: Viscosity measurements of the a) substrate and of the b) product polyester

The diagrams in Figure 1 show that viscosity at room temperature is higher for the substrate. Although at reaction temperature at about 60°C the viscosity of both fluids is the same. Thus all separation experiments were taken out with the substrate at 60°C. The density of the fluid was measured by Mohr-Westphal balance at 40, 50 and 60°C. The values give a line with the formula $\rho[g/cm^3] = 0{,}0006 \cdot T[°C] + 0{,}958$ with R^2=0,999.

As the experiments were done with magnetic particles in a viscous medium, the particle concentration of the feed and filtrate was determined by turbidity measurements with a 90° scattering light absorption for diluted suspension (NTU: Nephelometric turbidity unit). According to Beer–Lambert's law the absorption of light is proportional to the concentration if scattering of light is insignificant. For low concentration of the filtrate the 90° scattering light absorbtion is better suited to distinguish more clearly. Scattering light absorbtion is not linear for high concentration. The Hach turbidimeter is calibrated between 0 and 800 NTU, in this range the scattering intensity is proportional to the concentration. For higher concentrated suspension (for example the feed with 2 g/l) this is not true as multi-scattering occurs. The calibration is determined for every particle/fluid system as shown in Figure 2 and Figure 3.

The separation efficiency can be evaluated by measuring the turbidity value of the filtrate. The values of the filtrate concentration correlate to the concentration according to the calibration curves.

$$E = 1 - \frac{c_{filtrate}}{c_{feed}} \cdot 100\%$$

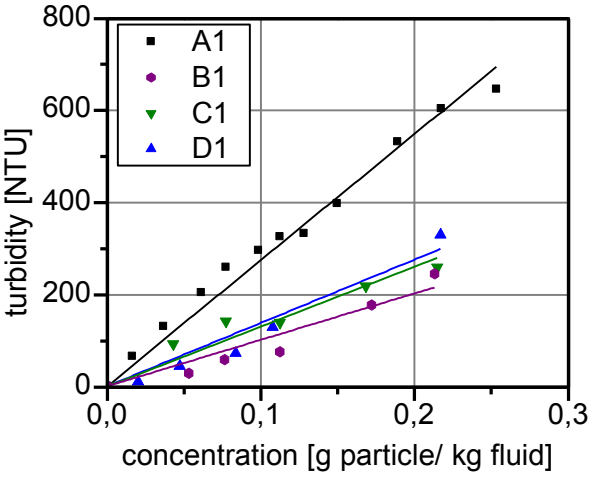


Figure 2: Calibration of the concentration of product A1 - D1 within Pripol 1012

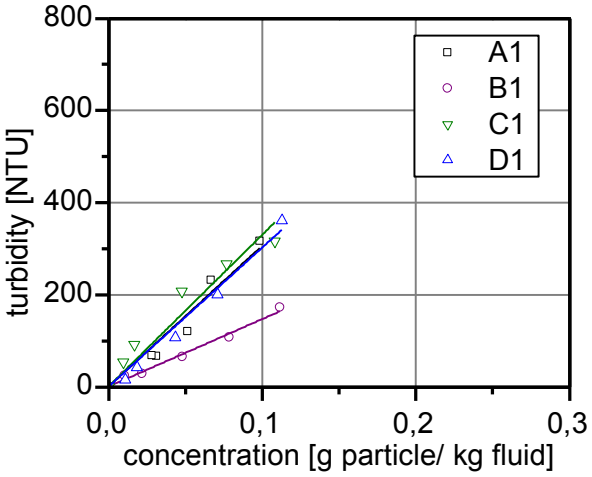


Figure 3: Calibration of the concentration of product A1 - D1 within water

3. Results

As separation equipment a small cylindrical magnetic filter with an inner diameter of 30 mm and a length of 85 mm equipped with 37 wire mesh stages of 3 different wire diameter and mesh width combinations were used. This arrangement is suitable in order to be able to take each filter stage separately from the filter for loading and backwashing experiments.

In order to supply viscosity independent results, Figure 4 shows the separation efficiency with growing Re-number of the experiments in water (half-filled symbols) and Pripol 1012 (not filled symbols). The Re-number has the characteristic length l Which is the diameter of the filter cell of 30 mm. The separation efficiency is plot at a probability scale which enables a closer look on the differences between the four products tested. The experimental results show that product D1 is best separated, product A1 is worse.

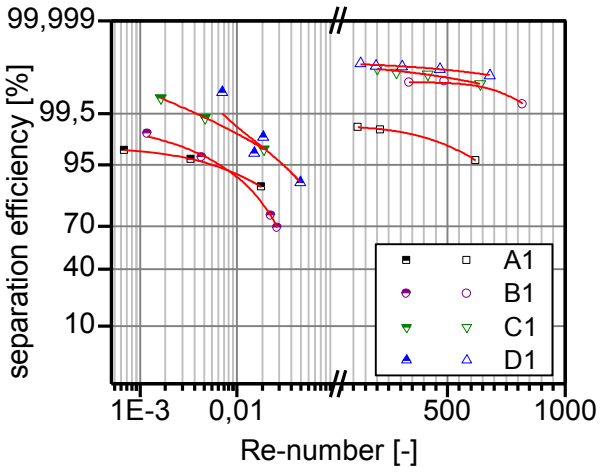


Figure 4: Experiment overview of the separation efficiency comparing the four tested products in Pripol 1012 (half filled symbols) and water (not filled symbols)

In order to be able to compare the four tested particle systems, the separation efficiency was calculated according to the method that is described in [4] and plotted against the ratio of magnetic velocity to inflow velocity. The magnetic velocity is defined according to Watson as the maximum velocity of a particle in the immediate proximity to the wire [5],

$$v_m = \frac{1}{18}\,\mu_0\,\Delta\kappa\,\mathrm{M}_D\,\mathrm{H}_0\,\frac{x^2}{a\,\eta}$$

with the permeability of vacuum μ_0, the difference of susceptibility of the particle and the fluid $\Delta\kappa$, the magnetization of the wire M_D, the magnetic background field H_0, the viscosity η, the wire radius a and the particle diameter x. The characteristic ratio of magnetic velocity to inflow velocity v_m/v_0 is an appropriate characteristic of the effectiveness of separation. It has been shown that the ratio should be bigger than 1, else separation is insufficient [1]. As particles are usually polydispers with a certain particle size distribution· $q_i(x_i)$ an integral magnetic velocity v_m has been defined in order to be able to compare different particle systems.

$$\frac{\mathrm{v_m}}{\mathrm{v_0}} = \frac{1}{\mathrm{v_0}} \int_{\mathrm{x_0}}^{\mathrm{x_n}} \mathrm{v_m}\ \mathrm{x_i}\ \cdot \mathrm{q_i}\ \mathrm{x_i}\ \mathrm{dx}$$

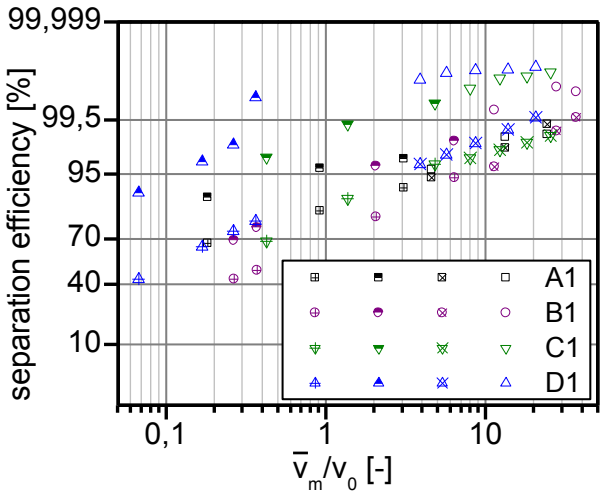


Figure 5: Comparison of calculated and experimental gained data for the four particle systems in water and Pripol 1012. Half-filled symbols are experimental data in Pripol 1012, straight crossed symbols are calculated values in Pripol 1012, not filled symbols are experimental data in water and lateral crossed symbols for data evaluated by calculation.

Figure 5 shows a comparison of the calculated and the experimental gained data. It is visible that the calculated values are lower than the experimental. The points of the itemized particle systems are lying on a line in the shown probability/log10 plot in good approximation. The difference between the calculated data is due to the fact that the particle size distribution, the magnetization and the number and size of the separation matrix varies. The difference between theoretical and test results owing to the lack of taking magnetic induced agglomeration into account within the calculation. Unexpectedly, the values of product A1 are higher than for product B1, which is probably caused by underestimation of fine particles during particle size distribution

measurement (laser diffraction). The particle size distribution measured by CPS Disc Centrifuge showed a slightly better resolution of the fine fraction. Nevertheless it has been assumed that these particles are unstable and fine particles detach from bigger agglomerates as the experiments for this product A1 showed that a large amount of very small particles could not be separated efficiently by magnetic separation.

4. Conclusion

The work shows that the separation of magnetic particles off viscous media of about 900 mPa s is possible if the fluid velocity or Re-number is sufficiently low. The calculations showed that it is possible to approximate the separation efficiency although the magnetic induced agglomeration was not included. This method enables to predict separation efficiency of new developed magnetic carrier particles in order to optimise the magnetic and morphological characteristics. The work is the basic for apparatus design development, with a given throughput the dimension of the filter cell can be sized. Further development will be done regarding back-washing of the filter to ensure that the process reaches a steady state.

5. Acknowledgements

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