

입상여과에서 입자물질의 부착

Particle Attachment in Granular Media Filtration

김진근*

Kim, Jinkeun*

한국수자원공사 수도관리2처

(2004년 9월 3일 논문 접수; 2004년 10월 5일 최종 수정논문 채택)

Abstract

Granular media filtration is used almost universally as the last particle removal process in conventional water treatment plants. Therefore, superb particle removal efficiency is needed during this process to ensure a high quality of drinking water. However, every particle can not be removed during granular media filtration. Besides the pattern of particle attachment is different depending on physicochemical aspects of particles and suspension. Filtration experiments were performed in a laboratory-scale filter using spherical glass beads with a diameter of 0.55 mm as collectors. A single type of particle suspension (Min-U-Sil 5) and pH control was used to destabilize particles. The operating conditions were similar to those of standard media filtration practice: a filtration velocity of 5 m/h. More favorable particles, i.e., particles with smaller surface charge, were well attached to the collectors especially during the early stage of filtration. This selective attachment of the lower charged particles caused the zeta potential distribution (ZPD) of the effluent to move to a more negative range. On the other hand, the ZPD of the effluent did not keep moving from less negative to more negative during the later stages of filtration, and this result was thought to be caused by two reasons: ripening effects and the detachment of flocs.

Key words: filtration, particle attachment, zeta potential distribution, particle size distribution

주제어: 여과, 입자물질부착, 제타전위분포, 입자크기분포

1. INTRODUCTION

A wide variety of particles including microorganisms, other organic materials, and inorganics are present in

water as a result of either natural events or human activities. As higher quality drinking water is demanded, the need to remove those particles in drinking water treatment processes is increased.

Filtered water with a turbidity of 0.1 NTU is

*Corresponding author Tel: +82-42-629-2818, FAX: +82-42-629-3699, E-mail: k.jinkeun@kowaco.or.kr (Kim, J.K.)

considered safe for drinking purposes (Hatukai et al., 1997). Even at this low turbidity, the water can contain hundreds of particles per milliliter (McTigue et al., 1998). With proper pretreatment, most of the particles can be efficiently removed. However, some of the particles pass through the filters, or detach after being captured. The patterns of particle attachment vary with the particle size, solution chemistry, stage of filtration, and operating conditions.

O'Melia and Stumm (1967) were the first to view the water filtration process as having two distinct steps: transport and attachment. However, this distinction between transport and attachment, or physics and chemistry, is not clearly defined (O'Melia, 1987). Transport involves the long range forces or mechanisms that bring a particle near the surface of the collector or the previously retained particles. Several transport mechanisms have been postulated on a single filter grain, and the principal mechanisms to be considered are interception, sedimentation, and Brownian motion (diffusion). On the other hand, the fundamental theories of attachment are based on consideration of the forces acting on a particle with a collector of defined geometry. These forces, which control the attachment of a particle to a collector, include forces due to fluid flow, gravity, and surface interaction at close approach. The two most important forces are the van der Waals (VDW) force and the electric double layer (EDL) interaction force (Elimelech, 1994; Tobiason and O'Melia, 1988).

The total interaction energy profile can be obtained by summation of VDW and EDL forces. Fig. 1 represents typical net interaction energy profiles. In this graph, ΔE_a and ΔE_d represent activation energy for attachment and detachment, respectively (Hirtzel and Rajagopalan, 1985).

The VDW force is virtually independent of the composition of the aqueous phase, while EDL potential can be controlled by characteristics of both the solid and aqueous phases. The following methods can be commonly used to modify EDL interactions (Elimelech et al., 1995; O'Melia and Stumm, 1967); change in pH, change in ionic strength, and addition of counter-ions or

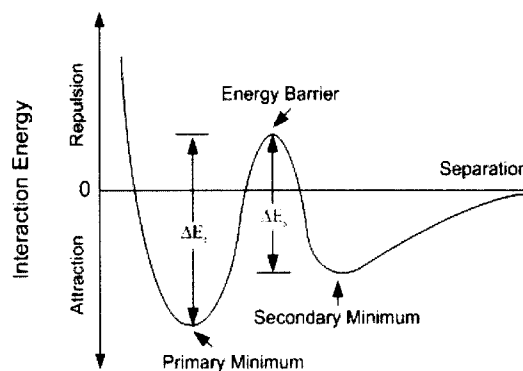


Fig. 1. Typical net interaction potential profiles.

change in solute type.

The primary focus of this research was to investigate particle attachment during the clean bed removal stage for dilute suspensions. During the research, the particle size distribution (PSD) and zeta potential distribution (ZPD) of particles, pH, solids concentration, ionic strength, and other significant parameters were measured or controlled to investigate why some particles penetrate granular media filters while others attach to the filter media.

2. EXPERIMENTAL METHODS

A schematic diagram of the experimental filtration system utilized in this research is presented in Fig. 2. A clear acrylic laboratory-scale filter column was used. This column has an inner diameter of 3.8 cm and a maximum media depth of 14.2 cm. During this research, the filter media depth was fixed at 10 cm. A more detailed explanation on the filter system can be found elsewhere (Kim, 2004).

Min-U-Sil 5 (U.S. Silica Company, WV) was used for the particle material during this research, and it was dispersed in Millipore water in order to make the particle suspension. Solid glass spheres (Potters Industries, Inc., NJ) in a size range of 0.5 to 0.6 mm were chosen for filter media. The filter media were cleaned to obtain uniform surface chemistry conditions as documented by Tobiason and O'Melia (1988). Particle and collector surface chemistry can be modified with many techniques, while

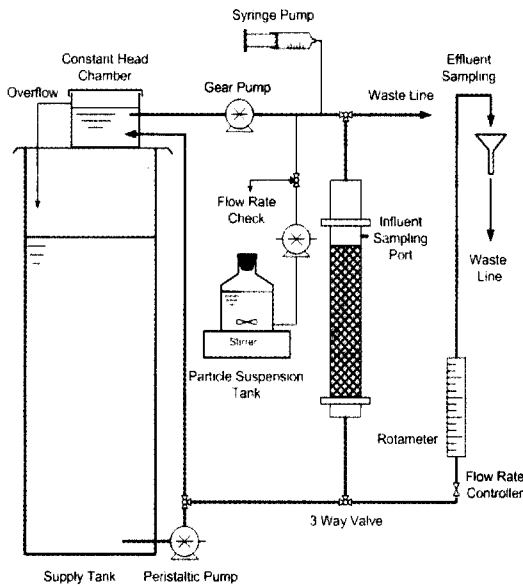


Fig. 2. Schematic of experimental filtration system (not to scale).

pH control was used to alter surface chemistry conditions during this research. Particle size and zeta potential of particles were measured by Coulter Counter (Coulter Multisizer, Coulter Electronics Inc.) and Zetaphoremeter IV (CAD, France), respectively.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1 Zeta potential distribution (ZPD)

The mean zeta potential of Min-U-Sil 5 particles at different pHs is shown in Fig. 3. Detailed test results on the zeta potential of Min-U-Sil 5 at a few selected pH values are presented in Table 1. Based on the zeta potential measurements at different pHs, three different pH conditions were chosen at a given ionic strength; 3.0, 4.0 and 5.0. During the pH control experiments, chemical

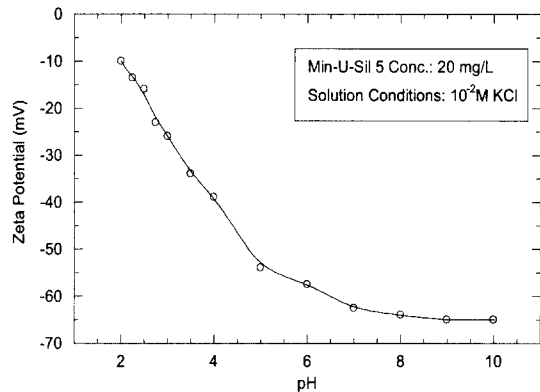


Fig. 3. Mean zeta potential of Min-U-Sil 5.

conditions of the glass beads were assumed to be the same as or at least similar to those of Min-U-Sil 5, because the main component of both materials is silica.

The ZPDs of Min-U-Sil 5 particles during the early stage of filtration at pH 3.0 are shown in Fig. 4(A), where the ZPDs of effluents are moving from a region of less negative to more negative surface charge during these first few minutes. This movement of the ZPDs can be attributed to the fact that more destabilized (less negative) particles can be easily attached to the collectors, while less destabilized particles appear in the effluent at the beginning of filtration. The t-test result confirms that these three mean zeta potential values in Fig. 4(A) are statistically different.

The ZPDs during the later stages of filtration are presented in Fig. 4(B); the zeta potential of the effluent was still more negative than that of the influent. However, the ZPDs of the effluent did not become more negative with the increase of time; the mean values stayed nearly constant between 10 and 120 minutes, but showed a small tendency to rise to less negative values.

Several factors that influenced the small decrease of the

Table 1. Summary of zeta potential measurements at three different pHs

pH	Mean zeta potential (mV)	Number of particles measured	Standard deviation (mV)	Coefficient of variation (%)
3.0	-26.4	83	7.1	26.9
4.0	-38.7	138	9.2	23.8
5.0	-54.7	116	9.3	17.0

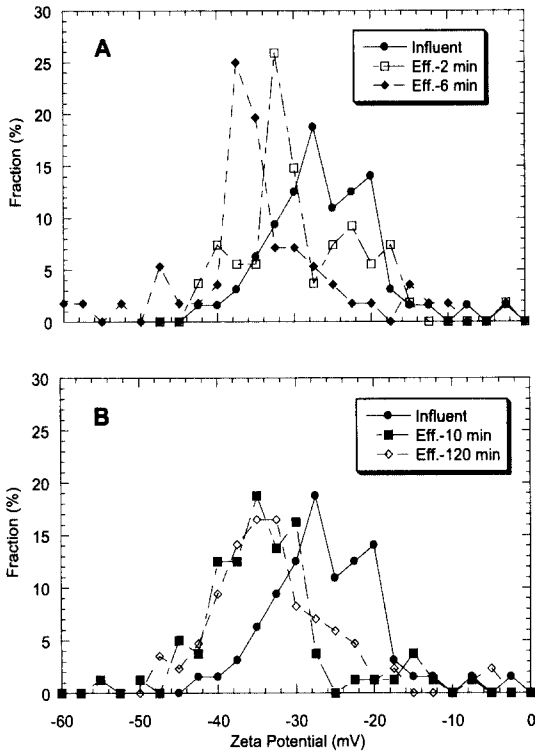


Fig. 4. ZPDs of Min-U-Sil 5 at pH 3.0 during the initial stage (A) and later stage (B).

zeta potential during the later filtration stage can be assumed. First, there was an influence of particle accumulation, i.e., the ripening effect. The surface area of the collector can be increased with the increase of solids accumulation, which leads to an improvement of transport efficiency. Second, there was a possibility of break-off flocs from the filter media. Flocs that were once attached to the filter media but were broken off can be assumed to have less negative surface charge than particles in the suspension that were never caught (non-attachment particles). Therefore, detachment of flocs can decrease the mean zeta potential.

The values of solids concentration remaining (C/C_0) and mean zeta potential changes with error bars at pH 3.0 as a function of cumulative hydraulic loading (CHL) are also shown in Fig. 5. Error bars show one standard deviation. The solids concentration remaining increased during the early stage of filtration and then kept

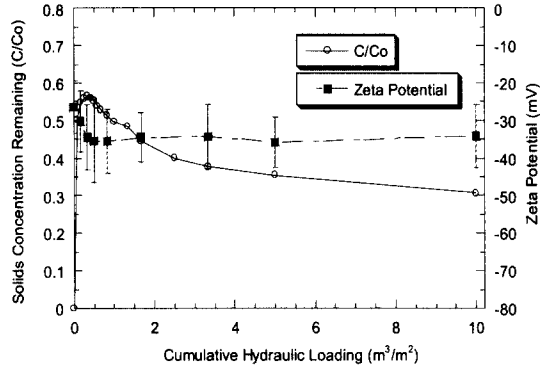


Fig. 5. Solids concentration remaining (C/C_0) and mean zeta potential values of Min-U-Sil 5 as a function of CHL at pH 3.0.

decreasing due to the ripening effect. On the other hand, the mean zeta potential values of the effluent moved quickly from less negative to more negative value during the clean bed removal stage and early ripening stage.

It can be assumed that the shape of effluent ZPDs during the initial stage of filtration is influenced mainly by surface charge interactions between particles and collectors. On the other hand, the ZPDs of the later stage of filtration, i.e., approximately after $0.83 m^3/m^2$ CHL, can be determined by surface charge interactions between particles and collectors as well as by other factors.

The ZPDs during the initial stage of filtration at pH 4.0 are shown in Fig. 6(A). Under this condition, a trend similar to that observed at pH 3.0 was noticed. On the other hand, Fig. 6(B) represents the ZPDs during the later stage of filtration at pH 4.0. During this period, the ZPDs of the effluent moved from more negative to less negative, i.e., closer to that of the influent. It can be thought that during this period the attraction force was not dominant for particle attachment. At the same time, deposited particles could increase the surface area of collectors, which in turn, increased the chance of particle transport from the bulk solution to collectors, regardless of the surface charges of particles.

The relative increase of the effluent mean zeta potential (absolute value) as a function of CHL at all three pH values is shown in Fig. 7, where the zeta potential of the

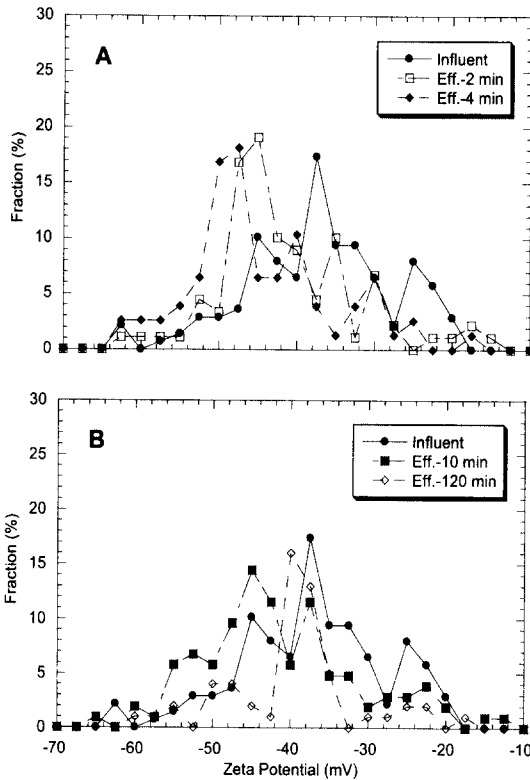


Fig. 6. ZPDs of Min-U-Sil 5 at pH 4.0 during the initial stage (A) and later stage (B).

effluent was normalized by the mean value of each influent zeta potential. During the initial stage of filtration, the absolute value of the mean zeta potential rapidly increased (i.e., became more negative) at all three pHs, which means less negative particles were well attached during this stage. The magnitude of the zeta potential increase was more significant when the absolute value of influent zeta potential was small. This means the influence of zeta potential on filtration efficiency was higher in the attraction dominant region: the net maximum changes of the effluent mean zeta potential from that of the influent were 9.4 mV (pH 3.0), 8.2 mV (pH 4.0), and 5.2 mV (pH 5.0).

3.2 Solids Concentration Removal and Particle Size Distribution

Better solids removal was confirmed under more

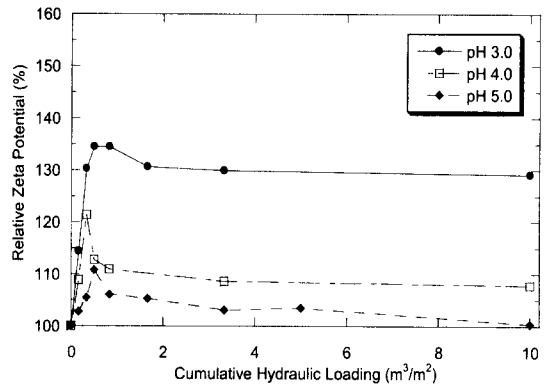


Fig. 7. Increase of mean zeta potential (absolute value) of Min-U-Sil 5 as a function of CHL at different pHs (normalized by each influent mean zeta potential).

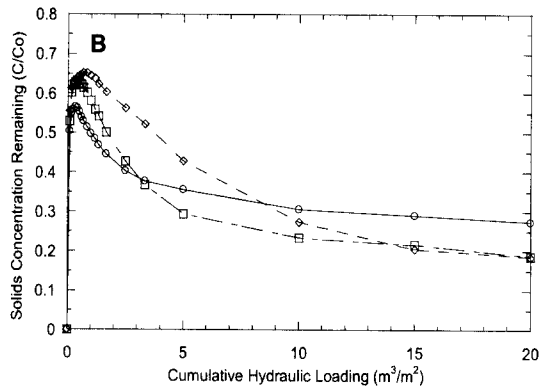
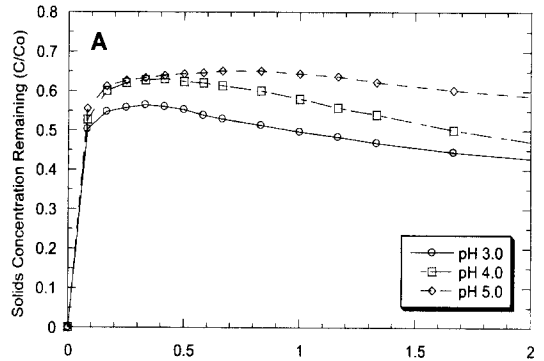


Fig. 8. Solids concentration remaining (C/C_0) at different pHs during the initial stage (A) and overall stage (B).

destabilized chemical conditions, i.e., smaller zeta potential of particles, especially at the initial stage of filtration. Less negative conditions resulted in better solids

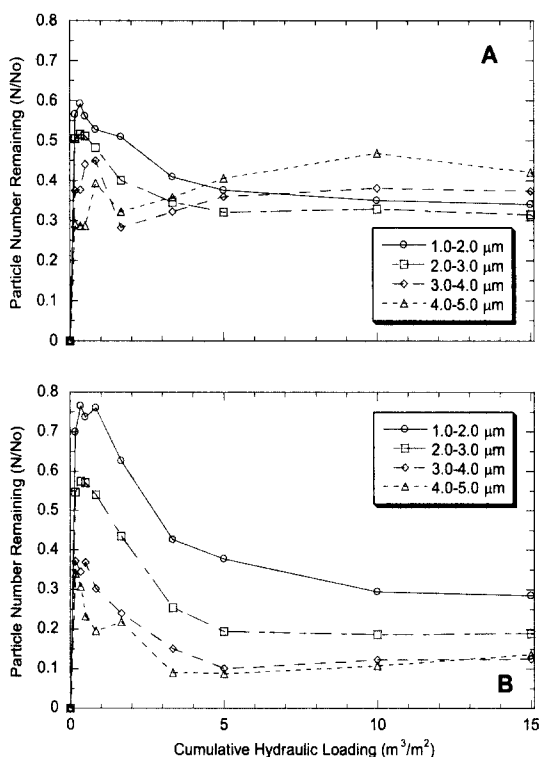


Fig. 9. Particle number remaining (N/N_0) vs. CHL at pH 3.0 (A) and 4.0 (B).

removal, at least during the clean bed removal period as shown in Fig. 8(A). On the other hand, the overall solids concentration remaining during 4 hours of filtration is shown in Fig. 8(B). The results suggest that better clean bed removal does not necessarily mean better overall particle removal. Although better clean bed removal was achieved at pH 3.0 as expected, the filters operated at pH 4.0 and pH 5.0 achieved better solids removal after a certain amount of CHL.

The particle number remaining (N/N_0) at pH 3.0 for different particle sizes is presented in Fig. 9(A). At the beginning of the run, bigger particles were well attached to the collectors, while smaller particles were not; as larger particles have better transport efficiency. Particle removal efficiency was improved after 6 minutes ($0.5 \text{ m}^3/\text{m}^2$ CHL) for smaller particles, but for larger particles ($4.0\text{-}5.0 \mu\text{m}$) removal efficiency did not improve. As larger particles were not well removed relatively at pH 3.0, this poor

removal may be one of the main reasons for the slow ripening.

Two possible reasons can explain the poorer particle removal of larger particles after the first few minutes. First, surface charge conditions of bigger particles were not as favorable for attachment as they were for smaller particles. Second, there was a possibility of the break-off of flocs, which could increase the number of larger particles; that is, some of the $4.0\text{-}5.0 \mu\text{m}$ particles in the effluent at the later stages were flocs of smaller particles that attached individually earlier but broke off together as a floc.

The particle number remaining at pH 4.0 is shown in Fig. 9(B). When these results were compared to those of pH 3, the $1.0\text{-}2.0 \mu\text{m}$ particles improved continuously; the $2.0\text{-}3.0 \mu\text{m}$ particles improved for a while and then were just about flat in both cases. However, unlike the PSD at pH 3.0, at the beginning of the filtration, better particle removal efficiency was noticed for the larger size particles (e.g., $4.0\text{-}5.0 \mu\text{m}$), and the particle removal efficiency improved from 0.33 to $5.0 \text{ m}^3/\text{m}^2$ CHL for all particle size ranges. Therefore, it can be assumed that, in this case, ripening was faster than it was at pH 3.0, because accumulation of larger particles can accelerate the ripening process.

4. SUMMARY AND CONCLUSIONS

The major observations based on these filtration experiments are stated below.

1. During the initial stage of filtration, the mean zeta potential of the effluent was more negative than that of the influent, when the zeta potential of influent particles and collectors were both negative. It can be assumed that, during this stage, less stable (more destabilized) particles are better attached to collectors than more stable particles.

2. After the initial stage of filtration, the absolute value of the mean zeta potential of the effluent decreased slightly or remained constant with time for 4 hours of filtration when the zeta potentials of the influent particles and collectors were both negative; nevertheless, it still

remained higher than that of the influent. Two reasons can be assumed to explain this trend; the ripening effect and the possibility of detachment of attached particles.

3. When the influent zeta potential is small (favorable conditions), the magnitude of the zeta potential change between the influent and effluent was large during the early stage of filtration, which means that, under favorable chemical conditions, the influent zeta potential is important for attachment.

4. Better initial particle removal does not necessarily mean better overall particle removal. It can be assumed that during the initial stage, chemical parameters such as zeta potential are important, but after a certain amount of time their importance is decreased, depending on the specific chemical conditions.

5. The influent PSD and the removal of certain size particles during the initial stage can significantly influence ripening, which in turn can influence the overall particle removal efficiency. When the same numbers of particles are removed during filtration, removal of larger particles leads to fast ripening because of their large surface area (i.e., large volume).

References

- Elimelech, M. (1994) Effect of Particle Size on the Kinetics of Particle Deposition Under Attractive Double Layer Interactions. *Journal of Colloid and Interface Science*, **164**(1), pp. 190-199.
- Elimelech, M. Gregory, J. Jia, X. and Williams, R. (1995) *Particle Deposition and Aggregation*. Butterworth Heinemann, Oxford, U.K., pp. 120-128.
- Hatukai, S. Ben-Tzur, Y. and Rebhun, M. (1997) Particle Counts and Size Distribution in System Design for Removal of Turbidity by Granular Deep Bed Filtration. *Water Science and Technology*, **36**(4), pp. 225-230.
- Hirtzel, C.S. and Rajagopalan, R. (1985) *Colloidal Phenomena Advanced Topics*. Noyes Publications, New Jersey, pp. 140-146.
- Kim, J. (2004) *Physicochemical Aspects of Particle Breakthrough in Granular Media Filtration*. Ph.D. Dissertation. The University of Texas at Austin, pp. 71-79.
- McTigue, N.E. LeChevallier, M. Arora, H. and Clancy, J. (1998) *National Assessment of Particle Removal by Filtration*. AWWARF and AWWA, Denver, CO., pp. 169-175
- O'Melia, C.R. (1987) Particle-Particle Interactions. In *Aquatic Surface Chemistry*. John Wiley and Sons, New York, pp. 385-403.
- O'Melia, C.R. and Stumm, W. (1967) Theory of Water Filtration. *J. AWWA*, **59**(11), pp. 1393-1412.
- Tobiason, J.E. and O'Melia, C.R. (1988) Physicochemical Aspects of Particle Removal in Depth Filtration. *J. AWWA*, **80**(12), pp. 54-64.