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Limitations of resistance-in-series model for fouling analysis in membrane bioreactors: A cautionary note

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ABSTRACT

The resistance-in-series (RIS) model has been used frequently to analyze membrane fouling phenomenon encountered in membrane bioreactors (MBRs) applied to wastewater treatment. Although it is easy to apply, there is a need to be cautious in the use of the RIS model, particularly when it is used to determine the relative values of the main membrane fouling components of an activated sludge suspension. The complex living suspension is not easily represented by simple addition of resistances; when researchers have checked for additivity of components, it has *not* been found. Most of the published work assumes that additivity and often two of the three individual resistances are measured and the third simply inferred. This is not justified. Better insights into the fouling in MBRs will be dependent upon the adoption of a standardised approach to fractionation and a commitment to measure the resistances of all three components considered.

Keywords: Activated sludge; Colloidal solids; Membrane fouling; Resistance; Suspended solids; Wastewater

1. Introduction

Membrane bioreactors (MBRs) have been increasingly applied to domestic and industrial wastewater treatment over the last two decades because MBRs offer many advantages over the conventional activated sludge processes. For example, there are greater removal of influent organics, reduced sludge production and a smaller footprint. Notwithstanding the great advantages of MBRs, membrane fouling remains a primary constraint impacting upon frequency of membrane cleaning or replacement, and thereby increasing operation and maintenance costs [1,2].

Although numerous studies have been focused on membrane fouling mechanisms in MBRs, it has not yet been fully understood because many factors act simultaneously. To investigate which components of an activated sludge give the main contribution to the overall resistance, numerous studies have fractionated the activated sludge suspension into three components – suspended solids, colloidal solids and solutes; and then determined the individual resistances of the components. However, microbial complexity and possible mutual interactions complicate the analysis. Whilst an explicable and reliable understanding of membrane fouling should still be pursued to solve the problems encountered in MBRs, the relevance of the resistance-in-series (RIS) model should be questioned and its limitations noted. The aim of this paper is to discuss proper use of the RIS model, to explain why it has a limitation and to provide a further insight into the membrane fouling behaviour in MBRs.

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2. Brief summary of the RIS model

The RIS model is derived from the analysis of one dimensional Darcian flow through a filter cake. As indicated in Eq. (1), the basic idea of the RIS model is that the permeate flux, J is proportional to the driving force for filtration and inversely proportional to the sum of all the resistances.

$$J = \frac{\text{driving force}}{\sum \text{resistances}} \tag{1}$$

This equation resembles the familiar Ohm's law as indicated in Eq. (2); the current, I, in an electric circuit is proportional to the potential difference, V, but inversely proportional to the overall resistance (R_{overall}).

$$I = \frac{\text{driving force}}{\sum \text{resistances}} = \frac{V}{R_{\text{overall}}} \tag{2}$$

Now the overall resistance depends, of course, upon whether the individual resistances are in parallel or in series. This alone should provoke caution with regard to the assumption of universal acceptance of the RIS model for complex filtrations. The simplest way to disaggregate the overall resistance of a membrane filter is to divide it into a cake resistance, R_c, which is in series with a fouled membrane resistance that can be written as $R_m + R_f$, where R_m is the intrinsic membrane resistance and R_f is the internal fouling resistance produced by adsorption of dissolved matter and/or pore blockage within the membrane. This results in the simple equation:

$$J = \frac{\text{driving force}}{\sum \text{resistances}} = \frac{\text{TMP}}{\mu \cdot R_T} = \frac{\text{TMP}}{\mu \cdot (R_m + R_c + R_f)} \tag{3}$$

where TMP is the transmembrane pressure, μ is viscosity of the permeate and R_T is the total resistance.

For MBRs an alternative that is being used comes from a separation of resistances according, not to position, but to a division based on constituent components of the activated sludge suspension. Both are now considered.

2.1. Division into cake and internal fouling resistances

The total resistance, R_T, is taken as the sum of each resistance ($R_T = R_m + R_c + R_f$). This summation is only possible if each resistance is additive. For an ideal case as illustrated in Fig. 1, individual resistances act independently of each other, so that three different resistances can be added. The cake resistance is certainly in series whilst R_f is defined as the difference between the fouled membrane resistance (R_m + R_f) and the clean membrane resistance, R_m, so additivity of these terms results from their definition. Interestingly (see for example the appendix of Field et al. [3] or Hermia [4]), the resistance R_c is proportional to its mass but R_f is not. This is mentioned because a change in concentration of foulants will rarely give a directly proportionate range in resistance.

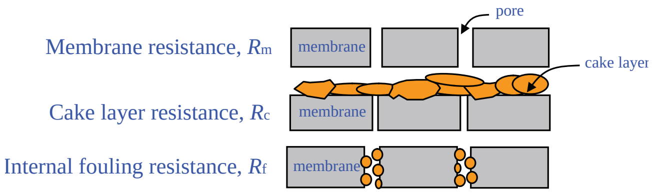

Fig. 1. Schematic of resistances in series model

For the case of membrane fouling phenomenon in MBR, Chang et al. [5] showed that R_c and R_f obtained from the RIS model were statistically reliable; they quantified separately pure-water filtration, activated sludge filtration, and pure-water filtration after removal of the cake layer from the membrane surface. Thus they had three measurements for each experiment and were able to qualify the three resistances but this does not confirm additivity. They found that the cake layer resistance (R_c) made up most of the total resistance and that the key factors controlling the R_c were the shape and size of the activated sludge flocs and the porosity of the cake layer accumulated on the membrane surface.

As the validity of differentiating each resistance on the basis of arbitrary physical tests is questionable. Some authors prefer to quote a single fouling resistance value, R_F, including both resistances of R_c and R_f (i.e. $R_T - R_m$) [6–8]. The values of R_c and R_f are interdependent; the amount of smaller particles contributing to R_f will be dependent upon the properties of the cake which inhibits passage of the smaller particles. There may be either a *filter-aid* situation or *over-clogging*, these being terms used by Hughes and Field [9] who gave a brief review of the filtration of complex suspensions containing both cells and soluble components. The difference between the two situations is the fouling potency of the smaller sized fouling components (i.e. the dissolved matter and small particles that cause pore blockage). If the fouling potency of these components is high with respect to the membrane then the cake acts as a filter-aid. The detrimental effects of allowing the soluble components unhindered access to the membrane outweigh any clogging of the cake which may occur. If the fouling potency of the smaller components is low with respect to the membrane then the cake may become over-clogged i.e., the extra resistance caused by the blocking of the interstices of the cake is, in this situation, more detrimental than any fouling the soluble components may cause to the membrane. If neither the *filter-aid* situation nor *over-clogging* occur, then it is reasonable to assume that the smaller components neither block nor adhere to the cake but pass through to the membrane filter as if the cake were not there.

Further comment on the interaction will be made later. At this stage it is noted that some might conclude that although there is a little difficulty in differentiating each resistance precisely and that the summation ($R_m + R_c + R_f$) generally matches well to the total resistance (R_T). However, such a match is a result of the definitions! Experiments are generally not performed to determine the values of R_c (in the absence of R_f) and R_f (in the absence of R_c). Furthermore there is no clear relationship between biomass concentration and R_c. Thus the consensus in the literature concerning the acceptability of the RIS model (with R_c as a true cake resistance) for the analysis of membrane fouling studies in MBR systems is ill founded, if by the RIS model one understands the summing of two independent resistances.

2.2. Division by components

The RIS model has been used in a different manner, namely to link the resistances according to the components of an activated sludge suspension. For example, in order to investigate what components of the activated sludge suspension dominate the total resistance, Meng and Yang [10] reported that resistance of the activated sludge suspension in a MBR was considered to be equal to the sum of the resistances of the suspended solids, colloids and solutes [10]:

$$R_{AS} = R_{SS}+R_{COL}+R_{SOL} \tag{4}$$

where R_{AS} is a resistance of the activated sludge, R_{SS} is a resistance of the suspended solids, R_{COL} is a resistance of the colloids and R_{SOL} is a resistance of the solutes.

Many other studies have also attempted to investigate the relative contribution of resistances of each component [11-15]. All of these studies assumed that the activated sludge consisted of solutes, colloids and suspended solids, and used an equation similar to Eq. (4) for their analysis.

Eq. (4) seems to be feasible considering that the activated sludge suspension is comprised of three different components, i.e., suspended solids, non-settleable fine colloids and solutes. But the assumption, ''total resistance of the activated sludge suspension is a sum of individual resistances of each component'', is very questionable. Particularly, the following two issues should be verified prior to the use of the Eq. (4):

(1) Is the fractionation of activated sludge into three different components practically achievable?

(2) If yes, has it to be confirmed that the sum of individual resistances ($R_{SS} + R_{COL} + R_{SOL}$) is equal to R_{AS} or not?

3. Current use of the RIS model

In the next two sections current usage of the RIS model in MBR for wastewater treatment is examined and the above questions are addressed.

3.1. Fractionation of activated sludge into components

Precise fractionation of the activated sludge suspension to the three main components seems to be difficult because a boundary between each component is not clearly defined. For example, the soluble fraction was usually acquired through a filtration of activated sludge suspension with a 0.45 μm pore size filter [11]. Thus particles that were less than 0.45 μm were classified as soluble solutes. Obviously, a clear cut-off between the solids and solutes does not exist. Another study [15] used a 0.05 μm filter to obtain a soluble fraction, indicating that there is no accepted standard or unified method for obtaining solutes from activated sludge suspensions. Also adsorption of the solutes to the filter medium during the filtration with a 0.45 (or 0.05) μm filter should be considered as significant amounts of the solutes fraction could be adsorbed onto the filter medium itself. There is an unknown loss of solutes during the fractionation procedure.

Moreover, a boundary between suspended solids and colloids is more obscure. After gravitational settling of a complete activated sludge suspension, two fractions – settled solids and supernatant – can be obtained [14,15]. The settled solids represent the suspended solids whilst the supernatant is a mixture of non-settling colloidal solids and solutes. However, it is well known that the settling property of activated sludge is very dependent upon microbial physiology, flocs structure and sludge concentration. Therefore, settling could not guarantee a reliable reproducible fractionation of the activated sludge into suspended solids and a mixture of non-settling colloidal solids and solutes.To overcome this limitation, some studies have centrifuged the activated sludge suspension and then the supernatant was classified further into colloidal solids and solutes [13]. But this has a limitation as well because extra-cellular polymeric substances (EPS) and soluble microbial products (SMP), which existed in the flocs, could be released to the supernatant during centrifugation. This would lead to increased levels of colloids and solutes in the supernatant. Consequently, the experimental procedures for fractionation are problematic.

3.2. Additivity of individual resistances

More importantly the validity of summation of individual resistances must be addressed. Even if the fractionation could be successful, additivity of each resistance should be confirmed first. To do this, it is essential to compare the resistances of the complete activated sludge (R_{AS}) and the sum of individual resistances of the components ($R_{SS} + R_{COL} + R_{SOL}$). However, most studies that have reported the contribution of each fraction to membrane fouling did not confirm the additivity. This might have been due to difficulties in fractionation; there may have been confidence that two of the three fractions were purer than the third.

In their studies to evaluate the relative contribution of individual resistances, they have evaluated only two of three fractions and then the remaining resistance of the third component was inferred by subtracting the resistances of two components from the complete activated sludge resistance (R_{AS}) [10–15]. Thus, there was no check to confirm the additivity of the resistances. For example, Bae and Tak [11] used the following procedure to obtain each resistance.

The resistance of the activated sludge (R_{AS}) was measured from the activated sludge filtration, and calculated by the following equation:

$$R_{AS} = R_{\mathrm{T}} - R_{\mathrm{m}} = \frac{\mathrm{TMP}}{\mu \cdot J_{AS}} \tag{5}$$

where J_{AS} is flux of activated sludge suspension before fractionation. Colloids and solutes were obtained from the supernatants after gravitational settling of the complete activated sludge suspension. The sum of both resistances of colloids and solutes was determined by a membrane filtration of the supernatant:

$$R_{COL} + R_{SOL} - R_{\mathrm{m}} = \frac{\mathrm{TMP}}{\mu \cdot J_{Sup}} \tag{6}$$

where J_{Sup} is the permeate flux of the supernatant. The soluble fraction that was acquired by filtration of the supernatant through a 0.45 μm filter and its resistance was measured by the filtration:

$$R_{SOL} = \frac{\mathrm{TMP}}{\mu \cdot J_{SOL}} \tag{7}$$

where J_{SOL} is the permeate flux of the soluble fraction. The remaining resistance, R_{COL} and R_{SS} were calculated from Eqs. (4) to (7). Note that R_{SS} was calculated by subtracting the sum of R_{SOL} and R_{SS} from the R_{AS} rather than by experimental determination. Thus there was no check upon the additivity of individual resistances. This is a common fault. Table 1 indicates other data and the inferred contribution is shown in italics.

Based on this procedure and calculations, the sum of each resistance ($R_{COL} + R_{SOL} + R_{SS}$) is certainly equal to the R_{AS} as shown in Table 1. If the percentage of individual resistances is added, it must be 100% because of the procedure used! This is because the R_{SS} was calculated by subtracting sum of R_{SOL} and R_{SS} from the R_{AS} rather than experimental determination.

Defrance et al. [14] tried to verify the additivity of resistances [14]. They carried out separate experiments which were different with the one depicted in Table 1. They divided the activated sludge suspension into two fractions - suspended solids and mixture of colloids and solutes – instead of three fractions. They compared the resistance of activated sludge (R_{AS}) with the sum of resistances for both components (R_{calc}). They concluded that the R_{calc} values were 48.9–53.3% higher than the R_{AS}, which is quite a large discrepancy. About 50% overestimation of resistance is not negligible for quantitative evaluation of membrane fouling. Furthermore Chang and Lee [16] also reported that the sum of individual resistances – solids and solutes fraction – were greater than R_{AS}. In their study the difference was very large with R_{calc} being 3.6–4.5 times greater than the resistance value of the undivided activated sludge suspension [16]. Both studies indicated that the additivity of individual resistances could not be justified. Consequently the determination of individual resistances to evaluate the relative contribution of each component to the overall resistances is of little merit and should be used with caution.

4. Discussion on the non-additive nature of resistances of individual components

As the sum of resistances of each fraction is different from that of a complete activated sludge suspension, it is interesting to discuss why the sum of individual resistances does not match well the overall resistance.

If a complete activated sludge suspension is filtered, the suspended and colloidal solids are deposited on the membrane surface and thus form a cake layer. Particularly, parts of solutes are entrapped or adsorbed to the cakes as well as to membrane pores and walls during

Table 1
Relative contributions of sludge fractions to membrane fouling in the literatures

Fraction of activated sludge	Contribution of individual resistances to total resistance (%)							
	Meng and Yang [10]			Bae and Tak [11]	Lee et al. [12]	Bouhabila et al. [13]	Defrance et al. [14]	Wisniewski and Grasmik [15]
	*a	*b	*c					
Solutes	13	26	31	13–14	63–71	26	5	52
Colloids	11	22	47	4–14		50	30	24
Suspended solids	*76*	*52*	*22*	*72–83*	*28–37*	*24*	*65*	*24*

a, b and c corresponded to the different physiological states of activated sludge.
Note that all the resistances were not individually measured. The inferred resistances are shown in italics.

the membrane filtration [17]. On the other hand, when the fractionated components are filtered separately, characteristics of the cakes formed by the individual components are significantly different from the cakes formed by the complete activated sludge. In particular the porosity and compressibility of the cakes formed by suspended solids and colloidal-only solids will be very different from those found in the cakes formed by the complete activated sludge. Referring back to Section 2.1, the complete activated sludge provides a filter aid for some of the components.

Many studies on the membrane filtration of proteins, bio-cells and their mixture have already reported that the sum of individual resistances of proteins and cells are different with that of the mixture solution. For example, Guell et al. [18] studied membrane filtration with yeast cells, proteins (BSA, lysozyme and ovalbumin) and their mixtures [18]. They concluded that the sum of the individual resistances of proteins and yeast cells suspension is greater than the resistance of the mixture solution. Hughes and Field [9] also reported that the sum of individual resistances of BSA and ovalbumin is over two times greater than the resistance of their protein mixture [19]. That is, the sum of resistances of individual components is usually greater than the resistance of a simple mixture of proteins and cells.

Moreover, the activated sludge suspension is not a simple mixture of individual components but a conjugate closely correlated with each component. When the fractionation of activated sludge suspension is carried out, the unique characteristics of individual components could be lost. One of the most important things to be considered during fractionation is EPS which is considered by many to be the primary membrane foulant in MBR processes. The bound-EPS is embedded in the sludge matrix and plays a key role in membrane fouling by working as a hydraulic barrier to permeation [20]. However, it may be even more potent foulant if it is released during fractionation. Such release might be expected during the course of fractionation by centrifugation due to shearing forces. This obviously makes the free-EPS (or soluble-EPS) level in the solutes higher than the level before fractionation. This could be one of the reasons for the discrepancy between the sum of resistances of individual components and the resistance of the activated sludge without fractionation. On the other hand, if the separation of solutes from activated sludge is done by filtration, adsorption of free-EPS to filter medium would be unavoidable. Consequently, the released or adsorbed EPS during the fractionation procedure is an additional important reason why the resistance of the complete activated sludge is not identical to the sum of individual resistances of the fractionated components.

5. Concluding remarks

Convenience does not imply applicability and the RIS model should be used cautiously, particularly when used to analyze the individual resistances of the sludge components. First of all, effective fractionation of the activated sludge suspension into three components (suspended solids, colloidal solids and solutes) by the experimental procedures found in literatures is likely to be difficult. Even if a fractionation were accomplished successfully, additivity of individual resistances is questionable. Results to date and the complex nature of the sludge suggest that additivity is not to be expected and should never be assumed. Nevertheless, one of the important advantages in the use of the RIS model is to characterize the relative significance of each resistance component in heterogeneous systems.

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