Water Content Analysis of Sludge using NMR Relaxation Data and Independent Component Analysis

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Abstract

In wastewater treatment, the dewatering of sludge is one of the most important steps, because it affects largely in both the process economics and the costs of sludge disposal. To optimize the dewatering processes, it would be beneficial to be aware of the different water types present in the sludge. In addition to free water, generally there are also mechanically, physically and chemically bound water within the sludge. All these water types behave differently when the sludge is dried, and they all require a different amount of energy when being removed. In this study, the Independent Component Analysis (ICA) method has been applied to an analysis of NMR (Nuclear Magnetic Resonance) relaxation data obtained from the measurement of wastewater sludge samples with a known moisture content. The results strongly suggest that the ICA method can be used for determining the amount of different water types within the wastewater sludge without a priori knowledge on their shares.

Keywords: independent component analysis, water content, nuclear magnetic resonance, sludge, relaxation decay

1 Introduction

Sludge is a semi-solid by-product remaining after wastewater treatment, industrial or refining processes. It is a separated solid suspended in a liquid, characteristically comprising large quantities of interstitial water between its solid particles (Global Water Community, 2015). This material can be dried to reduce its volume and to remove most of the moisture content of the solids within the sludge (Global Water Community, 2015). In wastewater treatment, the dewatering of sludge is one of the most important steps, because it affects largely both the process economics and the costs of sludge disposal.

It is suggested by several authors that the moisture in activated sludge can be classified to the following four categories (Kopp & Dichtl, 2000; Vesilind 1994; Tsang & Vesiling, 1990; Vesilind & Hsu, 1997; Smith & Vesiling, 1995):

- **Free** water: water which is not bound to the particles, including void water not affected by the capillary force.
- **Interstitial** water: water bound by capillary forces inside crevices and interstitial spaces of flocs.
- **Surface** water: water bound to the surface of solid particles by adhesive forces.
- **Bound** intracellular water.

This is a widely accepted classification and can be used as the reference in determining the main water types of sludge.

Another classification of water types in sludge is to divide it in three groups, i.e. 1) free water, 2) mechanically bound water, and 3) physically or chemically bound water. The free water in sludge can be easily removed by mechanical means, whereas the bound water is held firmly within the floc, bound to the sludge or trapped between the sludge particles, and thus cannot be easily removed (Jin *et al.*, 2004). The bound water can be further divided into chemically or physically bound water which is removable only by thermal drying, and mechanically bound water which is bound by weaker capillary forces (Colin & Gazbar, 1995).

In summary, it has to be emphasized that determining the water types is not straightforward, and based on the literature it is difficult to reach an unambiguous interpretation on the distribution of water within activated sludge (Vaxelaire & Cézac, 2004). Furthermore, there seem to be no studies concentrating on the analysis of water types in sludge without a priori knowledge of the shares of different water types.

Time domain nuclear magnetic resonance method (TD- NMR) is also becoming highly attractive for industrial applications due to relatively low price, mobility, easy operating, and simple sample preparation procedure. The most successful applications of TD-NMR confirmed by international standards are solid fat content determination in food and water (ISO 8292) and oil content in oilseeds (ISO 10565). They are based on the difference of NMR parameters of water and lipids and a low exchange degree between these two fractions.

A possibility to use the same principle for analysis of lipid content in microalgae (Gao *et al.*, 2008), for analysis of oil content of olive mill wastes and municipal wastewater sludge (Willson *et al.*, 2010) was demonstrated. Effects of flocculation on the bound water in sludge as measured by the NMR spectroscopy has been studied by Carberry and Prestowitz (1985).

Moreover, the international standard for hydrogen content determination in aviation fuels (ASTM D7171 – 05, 2011) has been developed recently. Metal ions, particularly paramagnetic ions, can also change significantly relaxation times in water and biological samples (Yilmaz *et al.*, 1999; Grunin *et al.*, 2013) which can be applicable when controlling wastewater treatment. Time domain NMR data have also been used in analyzing the water contents of wood and peat based fuels (Nikolskaya *et al.*, 2011) and monitoring the precipitation of metals in mine waters (Nikolskaya *et al.*, 2015).

Independent component analysis (ICA) is a statistical method that has been successfully applied to a variety of problems in signal processing (Hyvärinen *et al.*, 2001). For example, the method has been applied to a variety of problems in several fields such as brain imaging (Pulkkinen *et al.*, 2005; Calhoun *et al.*, 2002), vision research (Zhang & Mei, 2003; Ameen & Szu, 1999), telecommunications (Ristaniemi & Joutsensalo, 1999) and financial research (Kiviluoto & Oja, 1998; Back & Weigend, 1997). ICA is a method for extracting underlying, fundamental factors or components from multivariate data. It is designed so that it searches for components that are both statistically independent and non-Gaussian (Hyvärinen *et al.*, 2001), which makes it a distinguished method among the other techniques.

The complexity of spectral information can be approached by assuming that the obtained spectra are statistically independent. Principal component analysis (PCA) is the standard approach to analyze spectral data (Hyvärinen *et al.*, 2001). PCA is based on second-order statistics, which is applicable in the analysis of Gaussian distributed data. However, spectral data can comprise interesting information having a non-Gaussian distribution that can potentially be analyzed with ICA.

In the present study, the ICA method has been applied to an analysis of NMR relaxation data obtained from the measurement of wastewater sludge samples with a known moisture content.

2 Materials and methods

2.1 NMR measurements

The seven sludge samples (See Table 1) were obtained from an industrial waste water treatment plant. The samples were gathered after the dewatering stage of the process. The water contents of samples were measured using the standard oven drying method.

Relaxation times measurements were done using a mobile NMR device with a 1H resonance frequency of 25.7 MHz (Resonance Systems Ltd). The device has been modified for online measurements in industrial conditions. The permanent magnet of 0.6 T has dimension of 140x190x150 mm weighting 19 kg. The diameter of sensor hole was 10 mm. CPMG (Carr-Parcell-Meiboom-Gill) pulse sequence for spin-spin relaxation time T2 measurements was used.

Table 1. Description of sludge samples

Sample ID	Water content [%]
Sample 1	54
Sample 2	68
Sample 3	75
Sample 4	79
Sample 5	83
Sample 6	85
Sample 7	89

2.2 Independent Component Analysis

It is assumed here that there are n observed signals (i.e., types of water), WS_1, WS_2, ..., WS_n in the data, which are linear combinations of m independent components, IC_1, IC_2, ..., IC_m. The equation for IC_i can be written as:

$$WS_i = a_{i1}IC_1 + a_{i2}IC_2 + \cdots + a_{im}IC_m$$
$$= \sum_{j=1}^{m} a_{ij}IC_j \qquad (1)$$

where $i = 1, 2, ..., n$ and the a_{ij} are real coefficients (contributions of ICs). The independent components, IC_j, and also the corresponding coefficients, a_{ij}, are unknown.

The statistical model in Eq. (1) is called the independent component analysis model (Hyvärinen *et al.*, 2001). The ICA model is a generative model that describes how the observed data are generated by a process of mixing the components IC_i. Both IC_i and a_{ij} need to be estimated using the observed data. The starting point for ICA is the assumption that the components IC_i are statistically independent, which can be concluded from non-gaussianity (Hyvärinen *et al.*, 2001). Here, a fixed-point algorithm (Fast-ICA) was used as an implementation of ICA (Hyvärinen *et al.*, 2001). The analysis was performed using the Fast-ICA toolbox under the Matlab software platform (Mathworks, Natick, MA, USA).

After the analysis, the relative shares of each component can be calculated using the following formula:

$$a_{ij,rel} = \frac{a_{ij}}{\sum_{j=1}^{m} a_{ij}} \times 100 \qquad (2)$$

3 Results

The original NMR measurement signals can be seen in Fig. 1. It can be seen that there is a clear dependency between the amplitude of the signal and the moisture content of sludge samples. The gained NMR relaxation data were then analyzed by the ICA method based on the Hyvärinen's fixed-point algorithm (Hyvärinen *et al.*, 2001). Several numbers of ICs were tested, and three ICs were eventually used, because this setting was found to yield the most consistent and stable results. The three independent components (IC) can be seen in Fig. 2. It can be seen that all three ICs have their own, independent behavior.

According to theory, the share of bound water from the total amount of water remains stable in the sludge when the water content is increased from 0 on to a certain point (See Fig. 3, above). After this point, other types of water start to accumulate. When moisture content is 100%, all water is considered to be in a free form, but when the sludge is dried, the share of free water decreases dramatically, and the share of bound water increases. The share of the so called interstitial water (bound by weaker capillary forces) reaches its highest value at around 70 – 90 % moisture content.

In Fig. 3 (below), the calculated relative shares of different water types as a function of the total water content of the samples can be seen. It can be seen that the measured and analyzed values roughly follow the theoretic values and thus support them.
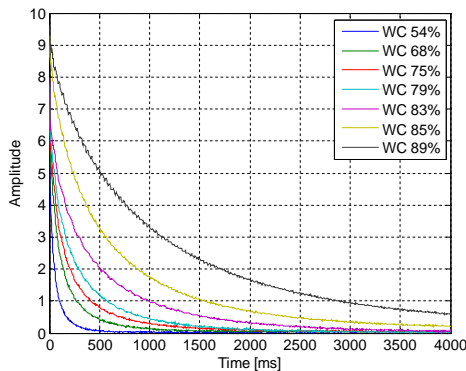

Figure 1. The observed NMR measurement signals (WS) from the 7 samples. WC = the water content of sample.
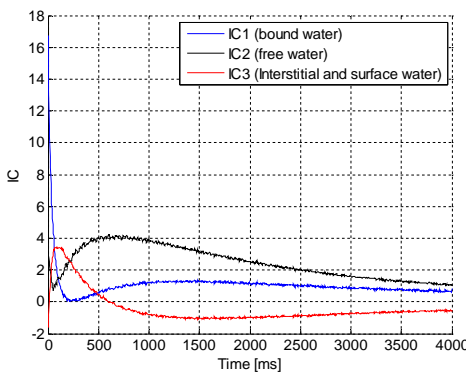

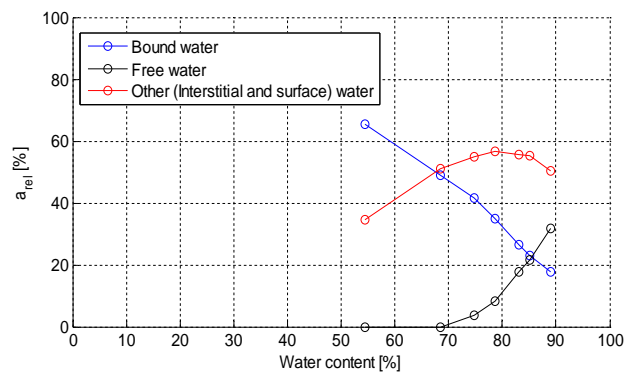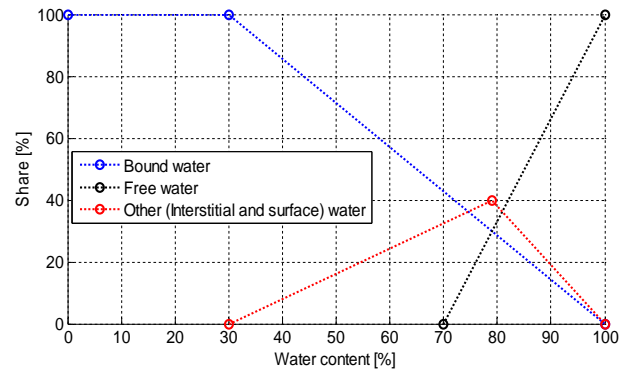**Figure 2.** The three independent components (IC) computed from the NMR relaxation data.

Figure 3. Theoretic (above) and calculated (below) relative shares of different water types as a function of the total water content of the samples.

4 Discussion

Measurement of the different water types in sludge is an exceptionally challenging problem, and to our knowledge this has not been tried before. In this respect, the results are extremely promising.

ICA is a universal statistical technique in which observed data are linearly transformed into components that are maximally independent from each other. A key issue in using the ICA method is to decide the number of ICs to be estimated. For the data set used, only the physically meaningful components were chosen. Our results support the use of three independent components in this case. This suggests that there are three types of signals in this data.

There is no universal truth on how many water types are present in sludge. The four water types including free, interstitial, surface and bound water, are widely accepted, but also other viewpoints exist. In this particular case three independent components could be most easily extracted from the NMR relaxation data. This suggests that there are three signals that are maximally and statistically independent when it comes to their spectra, but this does not mean that there could be more water types present as well.

Based on the results it seems that the combination of time-domain NMR and ICA can be used for determining the amount of different water types within the

wastewater sludge. It is also beneficial that the ICA method does not require a priori knowledge on the water types and their shares in the sludge. This makes it very specific and a promising approach to optimize the dewatering processes of sludge.

5 Conclusions

Based on the results it can be concluded that incorporating ICA into data analysis allows for decomposition of independent, systematically occurring patterns in NMR relaxation data. This new information can be used for guiding further study and may lead to a way of extracting the shares of different water types in wastewater sludge. This would help in making the sludge dewatering more economical and in reducing the costs of sludge disposal.

Acknowledgements

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