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# Bitumen-water interfacial tension modeling by using subtractive clustering method

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**ABSTRACT**

Calculation of interfacial tension during bitumen production is a crucial issue in heavy crude oil history. Upon variation in pressure, temperature and phases composition, interfacial tension between bitumen and water change. In this work a sophisticated method called subtractive clustering was utilized to predict dynamic interfacial tension between bitumen and water. The subtractive clustering method is composed of optimized fuzzy logic algorithm. A data bank which is collected from open-source literature, is used to create a reliable model. Then the prediction accuracy of the measured dynamic interfacial tension using subtractive clustering have been examined. Results state that the comparison of measured interfacial tension and predicted interfacial tension indicate acceptable accuracy of proposed model. Also more than 90 percent of data points have less than 3 percent absolute error.

**KEYWORDS**
bitumen; fuzzy modeling; interfacial tension; subtractive clustering; water-bitumen emulsion

## 1. Introduction

High volume bitumen resources are discovered all around the world. Just in Canada 1.3 trillion barrels oil sand derived bitumen are estimated. But production of bitumen is not easy as other hydrocarbon resources (Tsamantakis et al. 2005). Recently new technology thermal recovery methods like Steam Assisted Gravity Drainage (SAGD) and Cyclic Steam Stimulation (CSS) are suggested to recover more hydrocarbon from oil sands in heavy oil and tar sand resources. An interface would create because of injected steam contacted with oil sand. In this situation the steam turns to water and its heat transfers to bitumen across the interface. The transferred heat decreases bitumen viscosity and helps it to produce. This steam injection system execute periodically in Cyclic Steam Stimulation (CSS) recovery method (Rajayi and Kantzas 2011) (Speight 2015).

In the whole procedure bitumen-water emulsion is existed. Accordingly creation of bitumen-water emulsion during extraction makes serious difficulties for refining. Water droplets carry large amount of salts and leave them in surface facilities. They cause corrosion in equipment and facilities (Yan, Elliott and Masliyah 1999) (Speight and Moschopedis 1979). As a usual fact oil sand ore contains 6–13 wt% bitumen. After production or mining, because of its closure to the surface, the produced bitumen enters to the series of centrifuges. When it leaves it still have 2–3% water and 0.4% solids particles (Tsamantakis et al. 2005). However before describing the emulsion characteristics further, this kind of mixture is ran into industry application more and more commonly. Bitumen-water emulsion is complex as bitumen itself is. There is no exact prediction for bitumen properties behavior or when it is presented in water base emulsion (Schramm and Smith 1985).

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When a thermal recovery method is applying, different properties of oil sand ore and reservoir fluids change during the operation such as: fluid-fluid interfacial tension, rock-fluid interfacial tension, contact angle, fluid viscosity and wettability of the water-hydrocarbon. Study on bitumen-water interfacial is essential to evaluate the efficiency of thermal recovery method. When two phases are mixed Interfacial tension are present and the stability of the mixture dependents on the strength of tension on surface (Alvarez et al. 2008) (Drelich, Fang and White 2002) (Speight 2015). Along reservoir rock to surface, pressure, temperature and fluids composition are changing. These parameters can effect on surface tension. Also in the literature there are several cases that show variation in differential density and acid index of water change interfacial tension.

In practice, when unknown phenomena update unpredicted dynamically there is much uncertainty to model that mathematically. The main application of fuzzy logic modeling theory is its capability to handle particulate problems which conventional methods cannot adequately response. Subtractive clustering is one of the automated data-driven methods that construct the fuzzy model simply (Chiu 1994). One of its several advantages is avoiding the explosion of the rule base which is known as the curse of dimensionality problem. In this work, a unique fuzzy logic method has been utilized to model the behavior of interfacial tension between bitumen and aqueous phase and present this method as an exact study for interfacial tension modeling over bitumen-water emulsion.

## 2. Background review

Isaacs and Smolek (Isaacs and Smolek 1983) measured the interfacial tension of bitumen in contact of an aqueous phase. The variable parameters were temperature, alkalinity, surfactant type, surfactant concentration, and bitumen drop size and age interface. They evaluated the effect of each phase ones on interfacial tension of water-bitumen system. Takamura and Chaw (Takamura and Chaw 1985) found a model to explain the electric properties of the bitumen-water interfacial. Drelich and Miller (Drelich and Miller 1994) examined the effect of aqueous phase composition on interfacial tension. Variation in PH and concentration of sodium tripolyphosphate were applied in their experiments.

Yan et al. (Yan, Elliott, and Masliyah 1999) designed an experimental study to evaluate the effectiveness of various component of bitumen in bitumen-water emulsion. The various component were asphaltene, deasphaltene and fine solids. Schramm et al. (Schramm, Stasiuk, and Turner 2003) measured the interfacial tension between bitumen and aqueous phases at temperature ranged 50°C to 80°C. Also they studied the influence of interfacial tension in bitumen recovery by water base conditioning and flotation of oil sand with natural surfactant.

In 2004, Masliyah et al. (Masliyah et al. 2004) discussed bitumen extraction process and analyzed individual extraction steps. They investigated oil sand lump, size reduction, bitumen flotation and interaction among different components. Wu and Czarnechi (Wu and Czarnechi 2005) proposed a two-layer model to describe the layout of large and small surface active diluted bitumen-water interface by using a thermodynamic approach. Their model was able to reconcile contradiction found in literature. Tsamantakis et al. (Tsamantakis et al. 2005) studied interfacial properties of water in diluted bitumen emulsion by using micropipette. They proved this technic is useful in studying the interfacial micrometer-sized emulsion droplets. Also they observed bitumen concentration and different solvent mixture effect on crumpling of the interface.

Chaverot et al. (Chaverot et al. 2007) evaluated the time dependence of the interfacial tension between commercial bitumen from different origins droplets and acidic aqueous (PH 2) by pendent drop method. They observed significant changes of the interfacial tension over time scales of several thousands of seconds because of the existence of surface-active species in bitumen. They obtained quantitative data on endogenous surfactant for first time. Rajayi and Kantzas (Rajayi and Kantzas, 2011) studied the effect of temperature and pressure on contact angle and interfacial tension of quartz-water-bitumen systems by using the axisymmetric drop shape analysis (ADSA) method. Their experiment covered a temperature range from ambient to 100°C and pressure range from ambient to 1000 psi.

## 3. Material and methods

An interfacial tension data bank is collected from literature which is mentioned before. There is a series of 180/220-grade bitumen from six Western European refineries in this data bank. They represent a good panel of commercial bitumen in market and cover a wide range of acid indices technically. Dynamic interfacial tension of these bitumen samples are measured and found its dependency on temperature, bitumen acid index, differential density and also time. Thus these results cover temperature range from 90°C to 140°C and differential density range from 0.016 to 0.05 gr/cc. It is specified that the range of bitumen acid index is from 0.09 to 4 mgr of KOH/gr and experiments are continued to 6000 seconds. Also it must be noted that the bitumen dynamic interfacial tension is reported in unit of mN/m. verses time.

### *3.1. Subtractive clustering algorithm*

Choose of cluster center and their initial locations strongly effect on performance of common clustering algorithm. The fuzzy C-means algorithm or k-means and mountain clustering are good examples of this kind of clustering algorithms (Bezdek, 2013) (Jang, Sun, and Mizutani 1997). Yager and Filev (Yager and Filev 1994) proposed the mountain method which is a simple and effective method to estimate the number and initial cluster centers locations. Their algorithm was based on proper data gridding and computing a potential value for each grid based on its distances on the actual data points. A point with many data points nearby consider as a good pint with high potential value. Then the grid point with highest potential value is selected for the first cluster center.

The novelty of their work was when the first center is chosen, the other grid points potential reduced according to their distance from the center of cluster. Then grid points near the first cluster will achieve reduced potential greatly. From the remaining potential value, a grid point with highest value is selected for next center. This procedure of acquiring new cluster center and reduction of remaining potential value continues until all of the grid points have the potential value less than the threshold. This algorithm is simple and effective but maintain function must evaluate at each grid point and computation grows exponentially with dimension of the problem and this is the main weak point of mountain method.

Chiu (Chiu 1994) suggested an extension of previous algorithm, called subtractive clustering. This extension helps to solve computational problem associated with maintain method. In this new extension it consider data points as the candidate of cluster centers instead of grid points. So this technique change the dependency of computation from problem dimension to problem size. Although this method makes good approximation and reduces volume of computation but the results could not be accurate when the actual cluster centers are not essentially located at one of the data points. Also the specification of grid resolution is not needed, but instead of that tradeoffs between accuracy and computational complexity must be considered. The mountain method's criteria is extended by subtractive clustering for accepting and rejecting cluster centers.

The procedure of subtractive method is explained in following. It considers a collection of n data points $\{X_1, X_2, X_3 \ldots X_n\}$ in an M dimension space. Then it assumes the data points to have been normalized in ach dimension so it bounds them by a unit hypercube without loss of generality. After that it considers each data point as a potential center. The data point potential of $X_i$ is defined as:

$$P_i = \sum_{j=1}^{n} e^{-\frac{4\|X_i - X_j\|^2}{r_a^2}} \quad (1)$$

where, the symbol || || is the Euclidean distance, and $r_a$ denotes positive constant cluster radius in data space. Therefore distance of a data point to all other data points effect directly on the measure of the data point potential. So a high potential value data points, have many neighboring data points unconditionally. The $r_a$ represents the effective neighborhood radius this means data points which are located outside of this radius have slight influence on the potential. After the computation of every data point

**Table 1.** Domain of data bank parameters.

| Parameter | Domain | |
|---|---|---|
| | Minimum | Maximum |
| Temperature (°C) | 90 | 140 |
| Differential Density (gr/cc) | 0.016 | 0.05 |
| Time (sec) | 0 | 6000 |
| Acid Index (mgr KOH/gr) | 0.09 | 4 |

potential, the data point with highest potential is selected as the first cluster center. For example let $X_1$ be the location of first cluster center. Therefore the potential of each data point, Pi, is defined as:

$$P_i = P_i - P_1^* e^{-\frac{4\|X_i - X_1\|^2}{r_b^2}} \quad (2)$$

where $r_b$ is penalty radius in data space which is positive constant. So, amount of potential is subtracted from each data point as a function of its distance from the first cluster center. The data point near first cluster will selected as next cluster center because it has significantly reduced potential. The penalty radius defines the neighborhood effectively and has measurable reductions in potential. After revision on all data points' potential, the next data point with highest remaining potential is selected as nest cluster center. These steps repeat until enough clusters potential obtained. There are specific criteria for refusing and selecting clusters centers to end the clustering process and avoid marginal cluster centers. Also the procedure of best cluster determination to achieve clustering algorithm results are presented in Figure 1. Besides the domain of data bank parameters is shown in Table 1.

## 4. Results and discussions

To utilize the modeling method introduced in previous section to approximate the dynamic interfacial tension behavior of Bitumen-water, first the gathered data are configured as following: I = [temperature, density differences of bitumen-water system, acid index, time] and O = [interfacial tension]

Where I is the input matrix and O is the output vector. Note that for modeling purpose the input and output data are divided into two groups. First contains 80 percent of the data for training the model and parameter estimation and the second group contains 20 percent of data for model test and validation.

The prepared train data are integrated into fuzzy clustering procedure, mentioned previously. During this algorithm first potential value, corresponding to each data point is obtained. Then this potential values will be updated and points with maximum potential value are set as a fuzzy cluster or fuzzy rule, in each iteration. When stopping criteria introduced in (Chiu 1994) have been reached the obtained

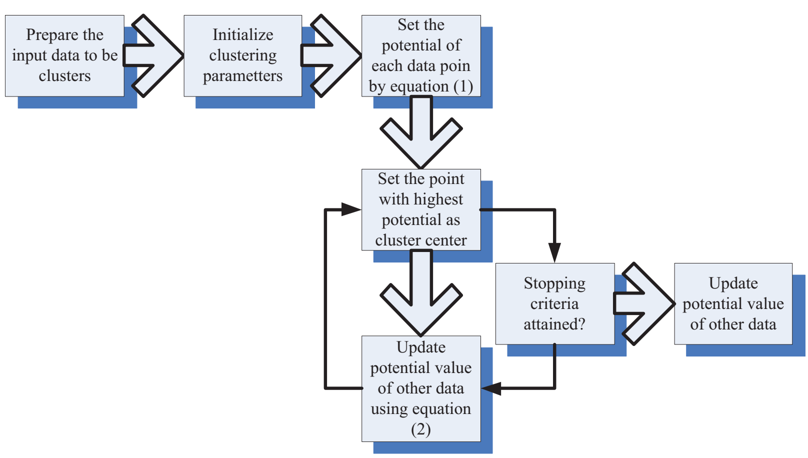


**Figure 1.** Subtractive clustering procedure.

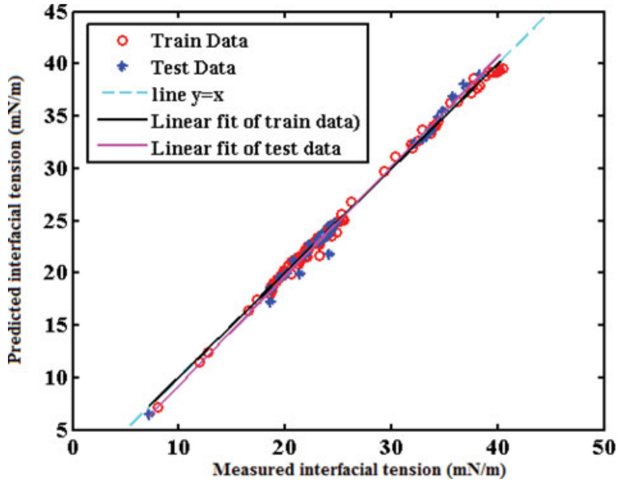


**Figure 2.** Predicted interfacial tension vs. measured interfacial tension.

fuzzy clusters are incorporated as a fuzzy rule base. Note that the clustering radius is assigned such that the distance between minimum and maximum value of each dimension of data is divided by 300. The radius are calculated as following: 0.1667, 0.0001, 0.0130, 20.1212, 0.1085

The obtained fuzzy rule base is not applicable individually. The next step is to integrate the fuzzy rule base with fuzzy inference engine, fuzzifiers, and defuzzifiers (Priyono et al. 2005). Once the overall fuzzy system have been determined the prediction performance of the model can be examined. In this respect the train input data and test input data are applied to the fuzzy system and the corresponding predicted interfacial tension will be calculated. The Figure 2 represents the measured or actual interfacial tension verses the predicted interfacial tension from fuzzy system. According to the results are illustrated in Figure 2, two linear fit can be obtained, for train data and test data respectively, which are as following: Train data linear fit: $Y_{train} = 0.9971^{*}x + 0.0730$ and Test data linear fit: $Y_{test} = 1.0470^{*}x - 1.2483$

Here x correspond to the measured interfacial tension, as input to the linear fits. Also the R2 index for train and test data examinations are calculated which are: R2 for train data: 0.9971 and R2 for test data: 0.9914.

The results indicate that the performance of the obtained modeling method is promising and the approximation and prediction dynamic interfacial tension is satisfactory. Figure 3 illustrates the relative

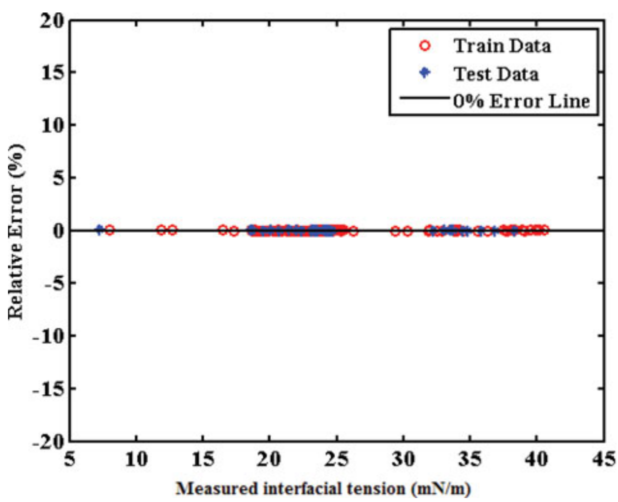


**Figure 3.** Relative error percentage of predicted interfacial tension vs. measured interfacial tension.

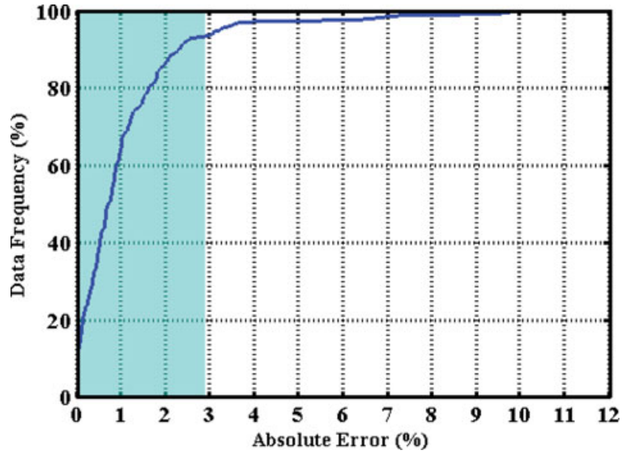


**Figure 4.** Statical representation of absolute error percentage of data frequency.

error percentage of the train data and test data which are distributed close by the zero percent relative error. For a statical examination of the modeling results based on the all data (both train and test data), data frequency and absolute error have been determined and depicted in Figure 4. The important note that can be conducted from the Figure 4 is that More than 90 percent of data points have less than 3 percent absolute error.

## 5. Conclusions

In this paper a well-known modeling concept called fuzzy clustering method is utilized to approximate the dynamic interfacial tension behavior of Bitumen-water, with respect to temperature, density differences of bitumen-water system, acid index and time. Subtractive fuzzy clustering is an advance fuzzy clustering method which possess the advantages of other clustering method besides coping with the disadvantage. The approximation and prediction of the measured dynamic interfacial tension using subtractive fuzzy clustering have been examined. Comparison of the interfacial tension from fuzzy model and the measured interfacial tension, points out that the proposed modeling method approximates the dynamic interfacial tension with acceptable accuracy. And also the prediction performance of the fuzzy model for unseen data points is promising. Results show that more than 90 percent of data points have less than 3 percent absolute error.

## References

Alvarez, G., S. Poteau, J.-F. Argillier, D. Langevin, and J.-L. Salager. 2008. Heavy oil— water interfacial properties and emulsion stability: Influence of dilution. *Energy & Fuels* 23 (1):294–9.

Bezdek, J. C. 2013. *Pattern recognition with fuzzy objective function algorithms*. New York, United States: Springer Science & Business Media.

Chaverot, P., A. Cagna, S. Glita, and F. Rondelez. 2007. Interfacial tension of bitumen— water interfaces. Part 1: Influence of endogenous surfactants at acidic pH. *Energy & Fuels* 22 (2):790–8.

Chiu, S. L. 1994. Fuzzy model identification based on cluster estimation. *Journal of Intelligent & Fuzzy Systems* 2 (3):267–78.

Drelich, J., C. Fang, and C. White. 2002. Measurement of interfacial tension in fluid-fluid systems. *Encyclopedia of Surface and Colloid Science* 3:3158–63.

Drelich, J., and J. D. Miller. 1994. Surface and interfacial tension of the Whiterocks bitumen and its relationship to bitumen release from tar sands during hot water processing. *Fuel* 73 (9):1504–10.

Isaacs, E., and K. Smolek. 1983. Interfacial tension behavior of Athabasca bitumen/aqueous surfactant systems. *The Canadian Journal of Chemical Engineering* 61 (2):233–40.

Jang, J.-S. R., C.-T. Sun, and E. Mizutani. 1997. Neuro-fuzzy and soft computing: a computational approach to learning and machine intelligence. *IEEE Transactions on automatic control* 42 (10):1482–84.

Masliyah, J., Z. J. Zhou, Z. Xu, J. Czarnecki, and H. Hamza. 2004. Understanding water-based bitumen extraction from Athabasca oil sands. *The Canadian Journal of Chemical Engineering* 82 (4):628–54.

Priyono, A., M. Ridwan, A. J. Alias, R. Atiq, O. Rahmat, A. Hassan, and M. Ali. 2005. Generation of fuzzy rules with subtractive clustering. *Jurnal Teknologi* 43 (D):143–53.

Rajayi, M., and A. Kantzas. 2011. Effect of temperature and pressure on contact angle and interfacial tension of quartz/water/bitumen systems. *Journal of Canadian Petroleum Technology* 50 (06):61–7.

Schramm, L. L., and R. G. Smith. 1985. The influence of natural surfactants on interfacial charges in the hot-water process for recovering bitumen from the Athabasca oil sands. *Colloids and Surfaces* 14 (1):67–85.

Schramm, L. L., E. N. Stasiuk, and D. Turner. 2003. The influence of interfacial tension in the recovery of bitumen by water-based conditioning and flotation of Athabasca oil sands. *Fuel Processing Technology* 80 (2):101–18.

Speight, J. G. 2015. *Handbook of petroleum product analysis*. New Jersey, United States: John Wiley & Sons.

Speight, J. G., and S. E. Moschopedis. 1979. The production of low-sulphur liquids and coke from Athabasca bitumen. *Fuel Processing Technology* 2 (4):295–302.

Takamura, K., and R. S. Chow. 1985. The electric properties of the bitumen/water interface Part II. Application of the ionizable surface-group model. *Colloids and Surfaces* 15:35–48.

Tsamantakis, C., J. Masliyah, A. Yeung, and T. Gentzis. 2005. Investigation of the interfacial properties of water-in-diluted-bitumen emulsions using micropipette techniques. *Journal of Colloid and Interface Science* 284 (1):176–83.

Wu, X., and J. Czarnecki. 2005. Modeling diluted bitumen— water interfacial compositions using a thermodynamic approach. *Energy & Fuels* 19 (4):1353–9.

Xu, Y., T. Dabros, H. Hamza, and W. Shefantook. 1999. Destabilization of water in bitumen emulsion by washing with water. *Petroleum Science and Technology* 17 (9-10):1051–70.

Yager, R. R., and D. P. Filev. 1994. Generation of fuzzy rules by mountain clustering. *Journal of Intelligent & Fuzzy Systems* 2 (3):209–19.

Yan, Z., J. A. Elliott, and J. H. Masliyah. 1999. Roles of various bitumen components in the stability of water-in-diluted-bitumen emulsions. *Journal of Colloid and Interface Science* 220 (2):329–37.