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Research Article

Correlations between SARA Fractions, Density, and RI to Investigate the Stability of Asphaltene

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Asphaltene precipitation is one of the most common problems in both oil recovery and refinery processes. Its deposition causes many problems mainly because of the fuzzy nature of asphaltene and the large number of parameters affecting precipitation. Unfortunately there is not a predictive technique for screening it. Refractive index (RI) was used as a stability test for asphaltene which makes a quantitative judgment for asphaltene stability. In our study, we first represent a novel correlation between SARA fractions and density and then demonstrate the relation between density and RI; second we developed a relation for SARA and RI, by utilizing RI to investigate the stability of asphaltene.

1. Introduction

Asphaltene deposition in oil reservoirs and/or production facilities causes a remarkable reduction of formation productivity/injectivity. Many literatures [1–3] gave the description of asphaltene problems and remedies throughout the world. Although asphaltene precipitation is a worldwide problem, its main cause has not been completely under stood [4]. The mechanism of asphaltene precipitation is very complex, despite a wealth of research on the topic. Controversy remains as to the nature of solving this problem [5]. According to the field experience [6, 7] and experimental observations [8–11], asphaltene stability depends on a number of factors, including the composition of the surrounding fluid, pressure, and temperature, in which asphaltene solubility is highly dependent on the composition of the crude, less dependent on the pressure, and hardly dependent on temperature.

2. Asphaltene Deposition Models

Effort has been done to present a universal model for asphaltene deposition. The existing models for asphaltene precipitation fall into three classes: (I) molecular thermodynamic models in which asphaltenes are dissolved in crude oil and crude oil forms a real solution [12–15]; (II) colloidal models in which asphaltene is suspended in crude oil and peptized by resins. The asphaltene precipitation is irreversible in such models [16–18]. Reversibility experiments are strongly against this type of models; (III) models based on scaling equation in which the properties of complex asphaltenes are not involved [19, 20].

3. Screening Technique for Asphaltene Stability

SARA analysis began with the work of Jewell et al. [21]. The saturate fraction consists of nonpolar material including linear, branched, and cyclic saturated hydrocarbons. Aromatics, which contain one or more aromatic rings, are more polarizable. The remaining two fractions, resins and asphaltenes, have polar substituents. The distinction between the two is that asphaltenes are insoluble in an excess of heptane (or pentane) whereas resins are miscible with heptane (or pentane) [22–24]. Resins are structural similar to asphaltenes, but smaller in molecular weight (<1000 g/mole) [24].

The proportions of each of the SARA fractions in a crude oil are related to the stability of asphaltenes in that oil. Resins have a strong tendency to associate with asphaltenes. Such association determines, to a large extent, their solubility in crude oil [25] in which resin molecules react at the addition of the light paraffin components by desorbing from the asphaltenes in an attempt to reestablish thermodynamic equilibrium, thus increasing the probability of asphaltene self-aggregation. Carbognani and Espidel [26] demonstrated that reservoirs with asphaltene problems were not primarily those with large amounts of asphaltene in the oil, but those with high saturate fractions. Leontaritis and Mansoori [17] recommended using the ratio of resins to asphaltenes as an indicator of asphaltene stability, based on the hypothesis that resins confer asphaltene stability by peptizing (or coating) asphaltenes. Fan et al. [27] observed that the proportions of each of the SARA fractions in a crude oil are related to the stability of asphaltenes in that oil. Alkafeef et al. [28] define the stability of a colloid dispersion as its resistance to flocculation, and the degree of "resistance" is used as a measure of the dispersion stability. Alkafeef et al. [29] point out that the destabilization (i.e., flocculation) of colloidal asphaltenes in oil-production flowing systems depends principally on breaking up the balances of attraction forces between the absorbed resin molecules and asphaltenes particles. De Boer and Leeriooyer [30] proposed a plot that can be used as a first screening tool to identify the potential for the oil to exhibit solid formation problems with having initial pressure, bubble point pressure, reservoir fluid density, and plotting pressure difference versus oil density. Jamaluddin et al. [31] proposed a screening criterion, based on the asphaltene weight percent versus resin weight percent. The colloidal instability index (CII) is another screening criterion, suggested by Yen et al. [32] that can be used to identify crude oil systems with deposit problems. The colloidal instability index is expressed as the ratio of the sum of asphaltenes and saturates to the sum of aromatics and resins. An asphaltene stability index was developed by Jamaluddin et al. [31] that utilized oil density at initial reservoir pressure and at bubble point pressure. Vargas et al. [33] developed a general method for modeling asphaltene stability as making relationships and obtaining a curve between the solubility parameters, cohesive energy, pressure, and composition along the bubble point and onset of asphaltene precipitation.

4. Refractive Index

The refractive index (RI) is the degree to which light bends (refraction) when passing through a medium. Values of RI can be measured very accurately and are used to correlate density and other properties of hydrocarbons with high reliability [34]. Information obtained from RI measurements can be applied for various reservoir engineering calculations. The examples are PVT behavior and surface tension of reservoir fluids [34], wetting alterations in reservoirs [35, 36], and asphaltene precipitation [37–39]. The refractive index of light crude oils can be directly measured using conventional refractometer [35, 36, 38]. However, direct measurements of the refractive index of many crudes, natural bitumen, and heavy fuels are unattainable since these liquids are too opaque so RI is only measured for fairly dilute solution; in these cases RI is determined for a series of oil/solvent mixtures and the results are extrapolated (in an assumption of a certain mixing rule) to determine the value for the crude oil [35, 37, 40]. It is usually assumed that a solution of a crude oil (bitumen) behaves as an ideal binary mixture of the components [37–39].

Solubility and RI have been related by the following formula [39]:

$$\delta = \left(\frac{\sqrt{3}\pi}{384}\frac{h\nu_e}{\sigma^3}\right)^{1/2}\frac{\sigma^3}{V/N_o}\frac{n^2-1}{(n^2+2)^{3/4}}, \tag{1}$$

where δ is the solubility; h Planck's constant; V the molar volume; σ hard sphere diameter; ν_e absorption frequency in the UV; N_o Avogadro's number; n refractive index.

The solubility parameter mapping of Wiehe [41] shows that asphaltene insolubility is dominated by aromaticity and molecular weight, not by polar or hydrogen bonding interactions. Studies at ambient conditions by Wand et al. [42] have shown that the refractive index at the onset of precipitation (PRI) is an important characteristic of oil/precipitant mixtures.

The aromatic fraction has little or no influence on RI_{oil}, whereas saturates correlate negatively and the resins and asphaltenes are positively correlated with RI_{oil} [27]. Generally speaking, anything that decreases the maltene RI also decreases asphaltene stability. An exception is the effect of increasing temperature, which causes thermal disaggregation even though RI decreases.

The refractive index is expressed as a function of composition and density through the Clausius-Mossotti or Lorenz-Lorentz equation [43, 44] in which the validity of the Lorenz-Lorentz equation to describe the density dependence of RI was investigated by Vedam and Limsuwan [44].

As reported previously [45], addition of aromatic or other hydrocarbon solvents has minimal effect on PRI. The fractions of various components in the mixture vary, concentration of the asphaltene fraction, and ratio of precipitant to solvent all vary, but the RI at the onset of precipitation is constant. In mixtures containing precipitate, do not deviate appreciably from the linear relationship between RI and volume fraction of crude oil in the oil-heptane mixture. Deviations of experimental data from "ideal" values are due to mixture containing precipitate component.

5. Fan et al.'s Method [27]

Fan and his coworkers related the refractive index of oil, $(RI)_{oil}$, to the SARA fraction by the following empirical equation:

$$(RI)_{oil} = 0.001452 \times S + 0.0014982 \times A + 0.0016624 \times (R + As), \tag{2}$$

where S is Saturate, A is aromatic, R is resin and AS is asphaltene percent. Fan et al. [27] defined PRI as the refractive index at the onset asphaltene precipitation and proposed that the difference between the refractive index of the oil $(RI)_{oil}$

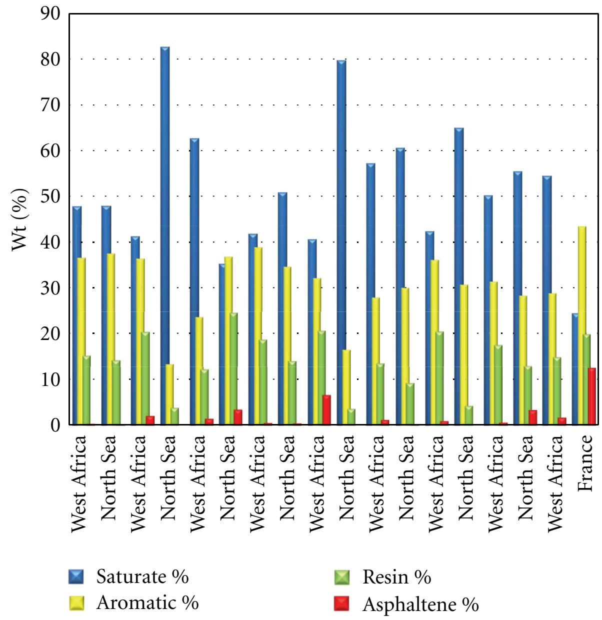

Figure 1: SARA percent weight for different fields.

and PRI can be used as a measure of the asphaltene stability, defined

$$\Delta(\mathrm{RI}) = (\mathrm{RI})_{\mathrm{oil}} \times \mathrm{PRI}. \tag{3}$$

And proposed the following stability criteria:

(i) crude oil with $\Delta(\mathrm{RI}) > 0.060$ are more likely to have stable asphaltenes;

(ii) crude oil with $\Delta(\mathrm{RI}) < 0.045$ are more likely to have asphaltene deposit problems;

(iii) crude oil with $0.045 < \Delta(\mathrm{RI}) < 0.060$ are in the border region.

6. Model Performance

For evaluation of proposed models, statistical indicators are usually applied to analyze models which are expressed as:

$$R^2 = 1 - \frac{\sum_{i=1}^{n}\left(y_{i_{\mathrm{observed}}} - y_{i_{\mathrm{calculated}}}\right)^2}{\sum_{i=1}^{n}\left(y_{i_{\mathrm{observed}}} - \overline{y}\right)^2}. \tag{4}$$

Average relative error:

$$\mathrm{ARE} = \frac{1}{n}\sum_{i=1}^{n}\left(\frac{y_{i_{\mathrm{calculated}}} - y_{i_{\mathrm{observed}}}}{y_{i_{\mathrm{observed}}}}\right)_i. \tag{5}$$

Average absolute relative error:

$$\mathrm{AARE} = \frac{1}{n}\sum_{i=1}^{n}\left|\frac{y_{i_{\mathrm{calculated}}} - y_{i_{\mathrm{observed}}}}{y_{i_{\mathrm{observed}}}}\right|_i. \tag{6}$$

Standard deviation:

$$\mathrm{SD} = \sqrt{\left(\frac{1}{n-1}\right)\sum_{i=1}^{n}\left(\frac{y_{i_{\mathrm{calculated}}} - y_{i_{\mathrm{observed}}}}{y_{i_{\mathrm{observed}}}}\right)_i^2}, \tag{7}$$

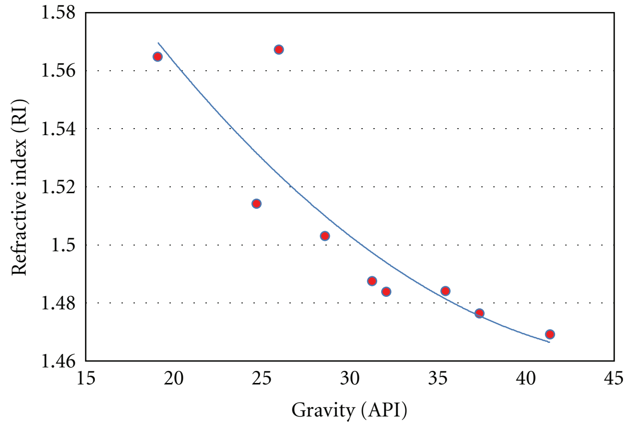

Figure 2: As API increases, the refractive index decreases.

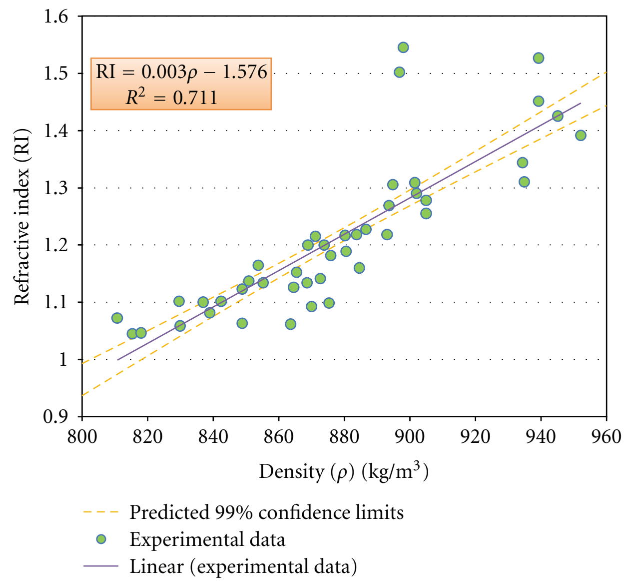

Figure 3: The linear relationship between RI and density with confidence limit of 99%.

where $y_{i_{\mathrm{observed}}}$ is the actual value, $y_{i_{\mathrm{calculated}}}$ is the predicted value of y, and $\overline{y}$ is the mean of the data values as in the above-presented formula.

7. Results and Discussion

SARA and density data for SARA-density relation are derived from previous works [27, 46]. SARA fractions are plotted in Figure 1, and the relation between SARA fraction and density was derived as follows:

$$\rho = 0.1402635 \times \mathrm{S} + 0.1390173 \times \mathrm{A} + 0.1655588 \times R^{0.9731603} + 0.1014975 \times \mathrm{AS}^{1.121967} - 13.26466, \tag{8}$$

where S is saturate, A is aromatic, R is resin, and AS is asphaltene percent. Related statistical indicators are shown in Table 1 that represents the precision of correlation.

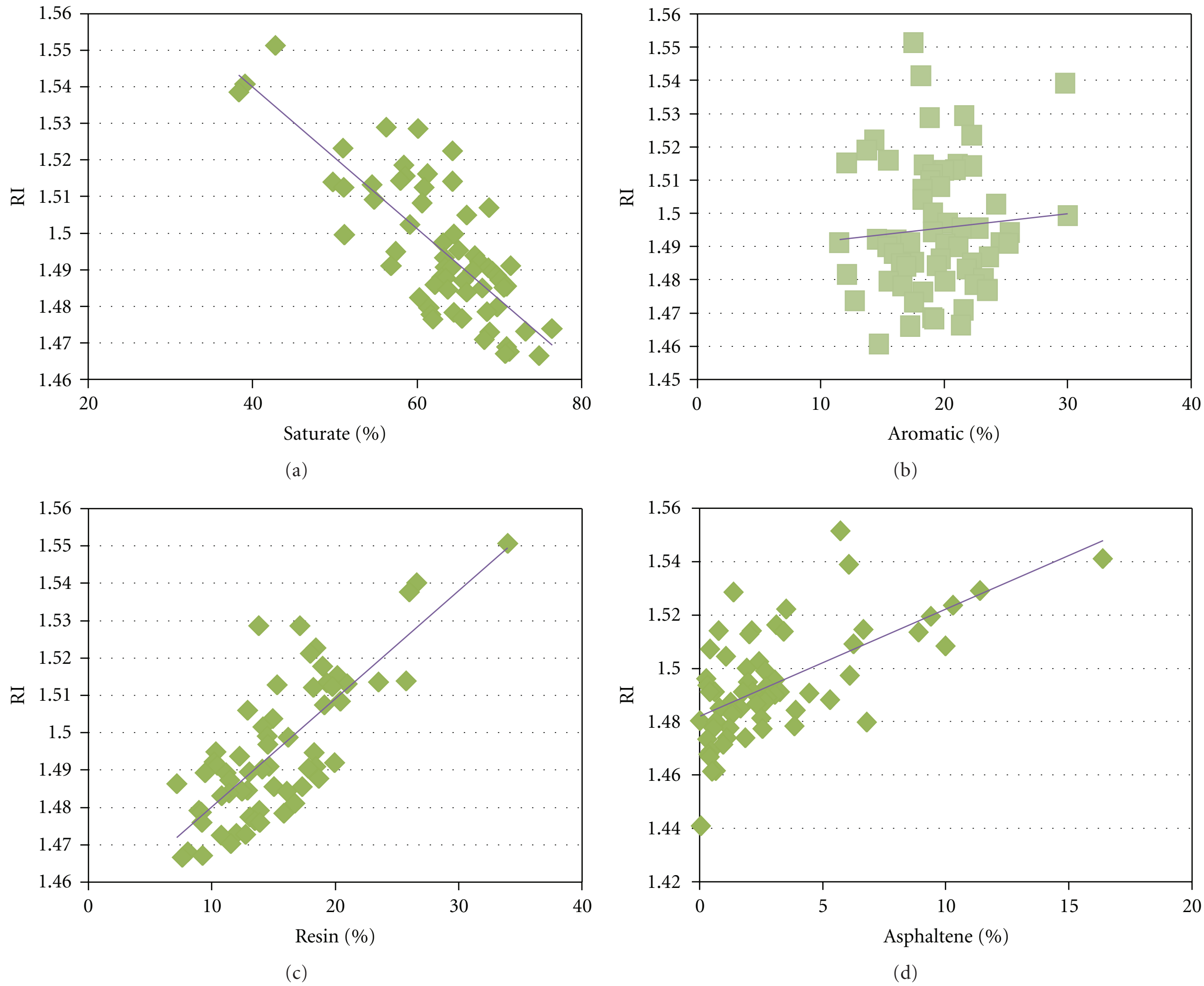

Figure 4: The behavior of RI with change of each fraction of SARA, the negative effect of saturate is apparent.

Table 1: Statistical indicators for considering the performance and precision of correlations.

Correlation	R^2	ARE	AARE	SD
SARA-density	0.951	0.000125	0.009055	0.010999
RI-density	0.771	−0.125557	0.125557	0.133987
RI-SARA (Fan et al.)	0.686	−0.000139	0.006419	0.007698
RI-SARA (proposed)	0.727	−0.000047	0.005752	0.006843

In this paper, it is also demonstrated that, according to relation between density and API°, whenever API° increases, the RI decreases (Figure 2), then the data for density and RI were plotted versus each other with a confidence limit of 99%, the present experiment shows that this relationship is linear (Figure 3) which with having density RI could be predicted:

$$\mathrm{RI} = 0.003\rho - 1.576. \quad (9)$$

The related correlation precision indicators are pointed in Table 1. Finally we presented a relation between SARA fraction and RI development and obtained a new correlation which, in addition to demonstrating the negative effect of saturate (Figure 4), shows better precision to obtain more accurate RI than what Fan et al. proposed. The proposed correlation by this paper is

$$\mathrm{RI} = -0.0008515 \times \mathrm{S} - 0.0002524 \times \mathrm{A} + 0.0016341 \times R + 0.0013928 \times \mathrm{AS} + 1.524412. \quad (10)$$

In Figures 5 and 6, obtained RI from Fan et al. and our correlation have been plotted versus observed RI; their accuracy comparison is presented in Table 1. By using Δ(RI) asphaltene stability could be investigated.

8. Conclusion

In this paper, we investigate relations between SARA fractions and density, between density and RI, and finally

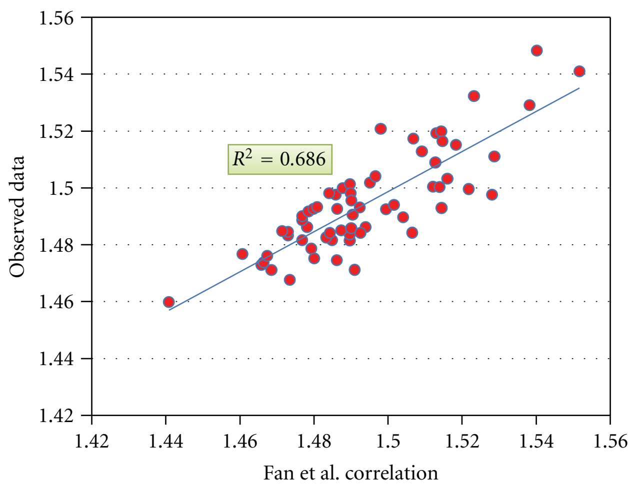

Figure 5: The calculated RI with Fan et al.'s correlation versus observed RI.

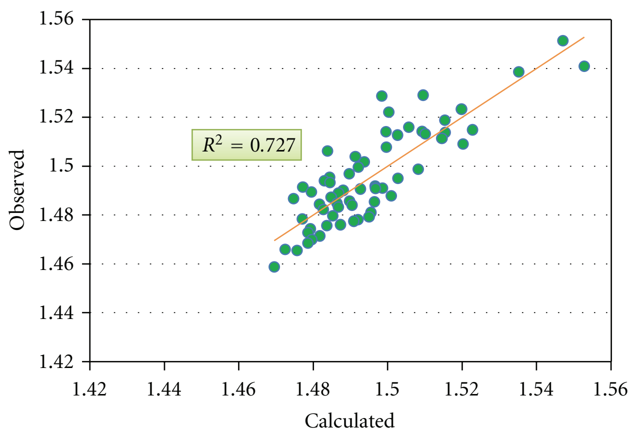

Figure 6: The calculated RI from proposed correlation versus observed RI.

between SARA fractions and RI. As an effort for screening asphaltene deposition is done, the obtained relations show satisfactory results and open new ways to determine RI from density and subsequently diagnosing asphaltene instability for causing problem. Also a comparison between current and previous works for SARA-RI relation has been performed which shows the better accuracy of our model.

Abbreviations

δ: Solubility parameter
ν_e: Absorption frequency in the UV
N_o: Avogadro's number
σ: Hard sphere diameter
A: Aromatic
AS: Asphaltene
CII: Colloidal stability index
g/mole: Gram per mole
API°: American Petroleum Institute
h: Planck's constant
n: Refractive index
PRI: Refractive index at onset point
R: Resin
RI: Refractive index
RI_{oil}: Refractive index of oil
S: Saturate
SARA: Saturate, aromatic, resin, asphaltene
V: Molar volume
ρ: Density.

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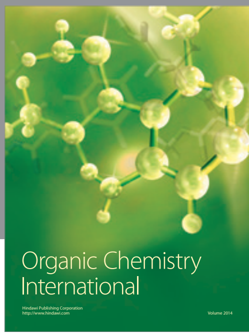
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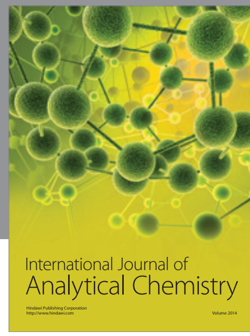
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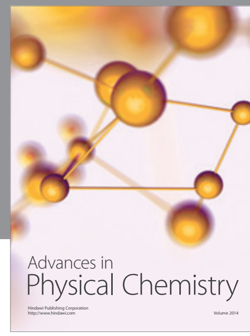
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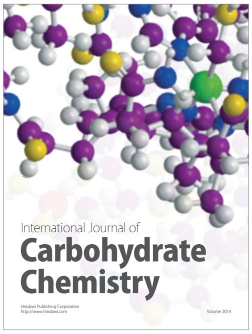
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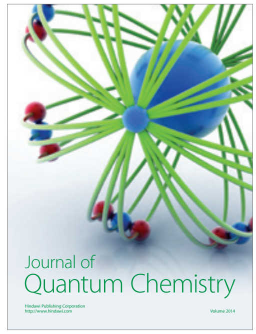
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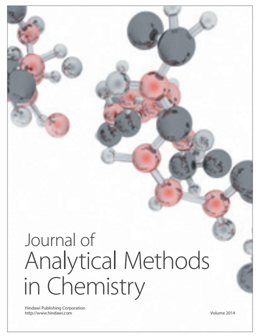
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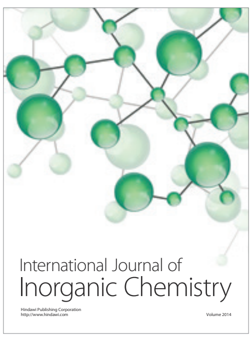
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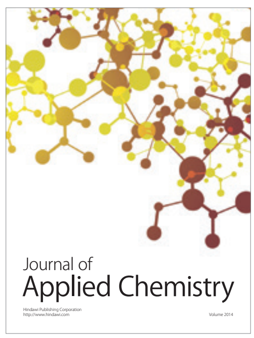
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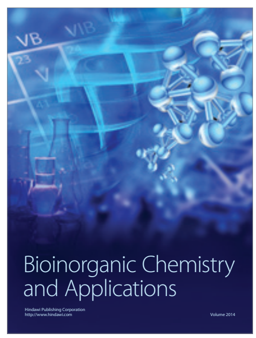
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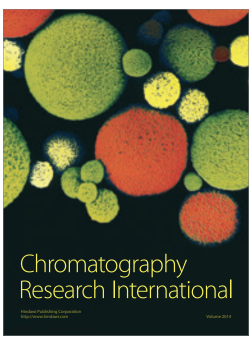
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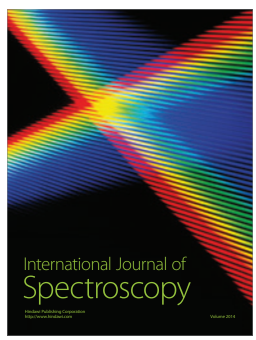
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