To the Mathematical Theory of the Temkin Adsorption Model

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Abstract: This study expands the mathematical theory of the Temkin adsorption model and demonstrates the way experimental data are to be additionally treated. A functional complement property of the Temkin isotherm has been studied as a particular case of the common theory of asymptotic complement. This property is shown to give rise to various analytical strategies to be employed for processing or interpretation of the experimental results, e.g. to perform mapping of data obtained in experiments onto a conjugated part of the true isotherm, to judge if presupposed energetic heterogeneity type of the surface really takes place, etc. Experimental data for adsorption of carbon monoxide on gold and of sodium oleate on steel with and without a magnetite coating have been extensively analysed.

Keywords: Adsorption heat, isotherm, Temkin, energetic heterogeneity.

INTRODUCTION

The usefulness of the Temkin isotherm in both historical and modern research can scarcely be overestimated. In its simplified form

$$A = A_0 - \frac{Q}{F} - \mathrm{const} + k\,\mathrm{lg}_{10}\, p_{\mathrm{H}_2} \tag{1}$$

(A stands for the amount of adsorbed hydrogen, p pressure of hydrogen in the gas phase; the notation exactly reproduces the origin [1]) it was discovered experimentally by A. Šlygin and A. Frumkin while determining the capacity of a platinum electrode in various electrolytes and became known first as the Šlygin-Frumkin isotherm. Just an incomplete list of its applications includes phenomena on iron and steels [2-5], zinc [6], gold, incl. nanoparticles [7], alumina and activated carbon [8], oxygenated multi-walled carbon nanotubes [9], hydrogen evolution reaction [10], sorption on clay-like minerals [11] and witnesses the highest actuality of the Temkin adsorption model in diverse fields of knowledge as, e.g., physics and chemistry of surfaces and interfaces, catalysis, corrosion, dissolution of metals, hydrogen evolution, properties of nanomaterials, environmental chemistry, pharmacology *et cetera*.

The theoretical basics of this isotherm originated in [12] as M.I. Temkin performed first *integrating over coverage* in

$$\vartheta = \int_{\mathrm{S}} \frac{a(\varepsilon)\cdot p}{1+a(\varepsilon)\cdot p}\, d\varepsilon \tag{2}$$

(p designates pressure in a gas phase or ionic activity or, provided a solution had a constant ionic power, concentration in a liquid phase, ϑ is overall surface coverage) given the law of heterogeneity $a(\varepsilon) = a_0 \exp\,(\varepsilon/kT)$, along the line

$$\varepsilon = \varepsilon_0 + (1-2t)\sigma, \tag{3}$$

where σ is a half-width of the adsorption heat range, $t \in [0,1]$, according to that the adsorption centres appeared to be ordered by their adsorption heat. This resulted in the well-known Temkin isotherm

$$\vartheta = \frac{1}{f} \log \frac{1+a_+p}{1+a_-p} \tag{4}$$

where coefficients $a_+ = a_0 \exp(\sigma) \exp\left(\frac{\varepsilon_0}{kT}\right)$ and $a_- = a_0 \exp(-\sigma) \exp\left(\frac{\varepsilon_0}{kT}\right)$ correspond to the strongly and weakly adsorbing surface areas respectively, $f = \ln\left(\frac{a_+}{a_-}\right) = \frac{2\sigma}{kT}$ is a constant, a_0p, where a_0 is a dimensioned coefficient in the definitions of a_+ and a_-, represents in both numerator and denominator of Eq. (4) dimensionless pressure (activity or concentration). The case $a_+ \gg a_-$ was defined by M. Temkin as that of "medium coverages" where the inequalities

$$a_-p \ll 1 \;\text{ and }\; a_+p \gg 1 \tag{5}$$

are valid. The former means that the weakly adsorbing areas are mainly vacant, and according to the latter, the strongly adsorbing areas are mainly occupied. Assumptions (5) result in a simplified form of Eq. (4)

$$\vartheta = \frac{1}{f} \log a_+p \tag{6}$$

(cf. Eq. (1)) that is known to be especially well-liked by experimentalists for a straight line in semilogarithmic coordinates.

The basics of a rigorous mathematical theory of the Temkin isotherm were worked out in [13] in the framework of a universal mathematical formalism for energetically heterogeneous surfaces. Employing the technique of Fourier-transformations, Temkin and Levich derived the function of distribution of adsorption centres on adsorption heat that is a constant within a given range of adsorption heats $[\varepsilon_0 - \sigma, \varepsilon_0 + \sigma]$:

$$g(\varepsilon) = \frac{1}{2\sigma} \tag{7}$$

Integrated with the local Langmuir isotherm over the range of all possible adsorption heats

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$$\vartheta = \int_{\varepsilon_0-\sigma}^{\varepsilon_0+\sigma} \frac{a(\varepsilon)\cdot p}{1+a(\varepsilon)\cdot p} g(\varepsilon) d\varepsilon, \qquad (8)$$

this is well-known [14] to reproduce Eq. (4). The summary of both works [12, 13] is well documented in [15].

The next contribution and generalization of the Temkin isotherm's mathematical theory was undertaken in [16, 17] and called in [18] "the most complete and elegant mathematical investigation of Langmuir adsorption on surfaces characterized by an exponential energy distribution"

$$g(\varepsilon) = C\exp\left(-\frac{\varepsilon}{\varepsilon_m}\right) \qquad (9)$$

In one of its parametrical limits (adsorption constant $\varepsilon_m \to +\infty$), while expanding the general solution represented by a hypergeometric function into an infinite series, one arrives in Eq. (4) again.

Although there have appeared a number of works that revisit such issues of the Temkin adsorption model as its general interpretation (s., *e.g.*, [7]), applicability fashion [19] and validation [20], those works that address its fundamental functional properties originated notably earlier. The aim of this paper is to establish one more functional property of the Temkin isotherm and adsorption model in general, that enables one to explore and to interpret experimental data in the framework of this adsorption model more extensively as well as to validate its applicability in every concrete case.

ASYMPTOTIC COMPLEMENT PROPERTY OF THE TEMKIN ISOTHERM

The reverse side of the methods [13, 16, 17] that advantageously suggest a generalized approach to consider a variety of heterogeneity types of surfaces, appears to represent its disadvantage. These don't distinguish between individual functional properties of different distribution functions. Indeed, the distribution function (7) in the Temkin case (even energetic heterogeneity of a surface) is axially symmetrical whereas the distribution function (9) in the Freundlich case (exponential energetic heterogeneity) is not (Figure **1**).

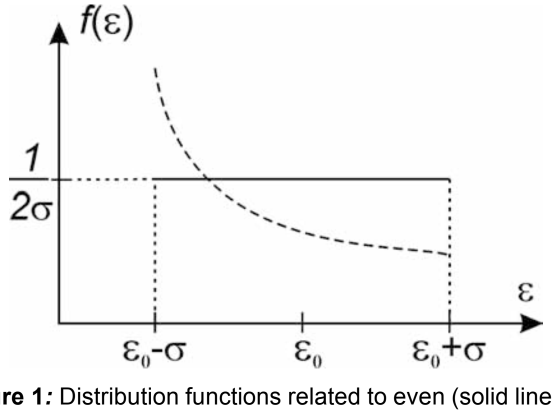

Figure 1*:* Distribution functions related to even (solid line, Eq. (7)) and exponential (dashed curve, Eq. (9)) energetic heterogeneity of a surface.

The general mathematical theory of what additional opportunities arise because of a distribution function's symmetry was developed in [21] under the name *asymptotic complement theorem* for the class of symmetrical (symmetrisable) distribution functions. When applied to the Temkin isotherm, this appears to be the following. Substitution of Eq. (7) into Eq. (8) gives the expression

$$\vartheta(q) = \frac{kT}{2\sigma}\int_{-\frac{\sigma}{kT}}^{+\frac{\sigma}{kT}} \frac{1}{1+\frac{1}{q}\exp(-x)} dx \ , \qquad (10)$$

where $q = a_0 p \exp\left(\frac{\varepsilon_0}{kT}\right)$ is hereinafter the modified dimensionless pressure. The integrand $\frac{1}{1+\frac{1}{q}\exp(-x)}$ is a monotonous function reaching its upper value at $q = 0$ and tending to zero as $q \to \infty$. The integral is invariant relative to replacement $x \to -x$ (easy to check by means of transformation of the integration variable). Considering the following difference

$$\vartheta(q=\infty) - \vartheta(q) = \frac{kT}{2\sigma}\int_{-\frac{\sigma}{kT}}^{+\frac{\sigma}{kT}} dx - \frac{kT}{2\sigma}\int_{-\frac{\sigma}{kT}}^{+\frac{\sigma}{kT}} \frac{1}{1+\frac{1}{q}\exp(-x)} dx = \cdots$$

and reducing both terms under the integral sign to a common denominator, one obtains

$$\cdots = \frac{kT}{2\sigma}\int_{-\frac{\sigma}{kT}}^{+\frac{\sigma}{kT}} \frac{1}{1+q\exp(x)} dx = \frac{kT}{2\sigma}\int_{-\frac{\sigma}{kT}}^{+\frac{\sigma}{kT}} \frac{1}{1+q\exp(-x)} dx = \vartheta(1/q)$$

on account of the above-mentioned invariance. Since

$$\vartheta(q=\infty) = \frac{kT}{2\sigma}\int_{-\frac{\sigma}{kT}}^{+\frac{\sigma}{kT}} dx = 1$$

one obtains the following equality

$$\vartheta(q) + \vartheta(1/q) = \vartheta(\infty) = 1 \qquad (11)$$

that represents an asymptotic complement property for the Temkin isotherm. It is valid for every q provided the Temkin adsorption model took place (can also be checked by a direct substitution of Eq. (4) into Eq. (11)).

The asymptotic complement property (11) of the Temkin adsorption model defines the centrosymmetric point $q^* = 1$ where coverage reads $\vartheta(q^*) = ½$ and that every point (q_1, ϑ_1) located on the left or on the right of the centrosymmetric point is to be mapped into its symmetric point (q_2, ϑ_2) located on the right or on

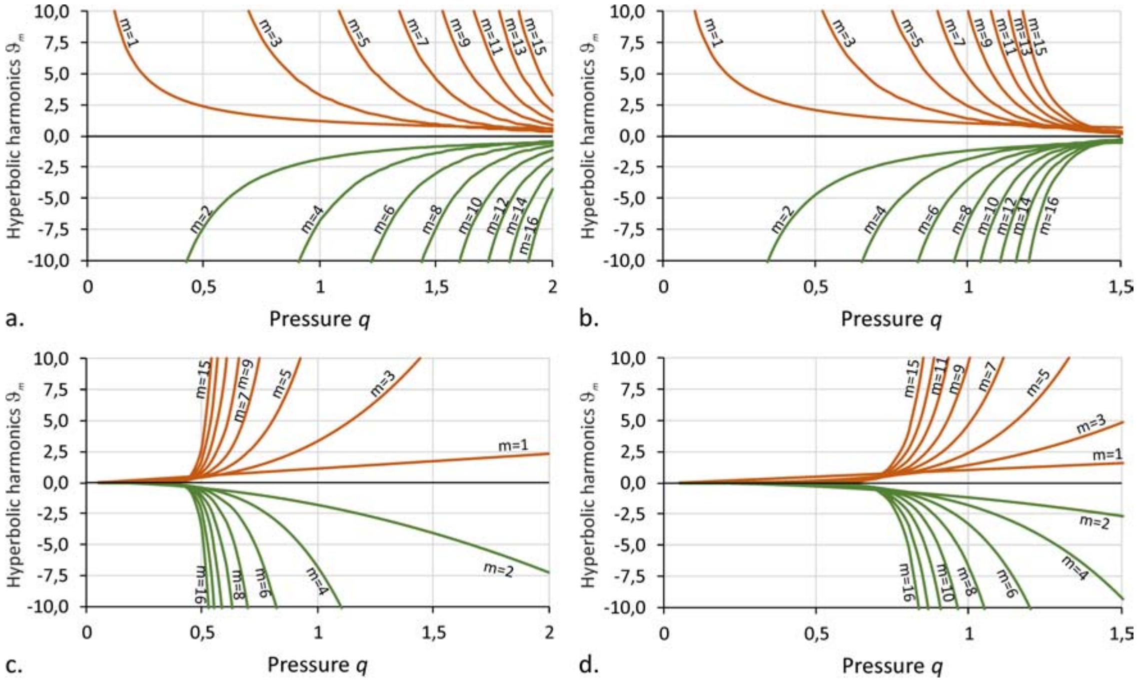

Figure 2*:* Hyperbolic harmonic components: a, c: $\frac{\sigma}{kT}=1$ and b, d: $\frac{\sigma}{kT}=0{,}5$. **a**, **b**: subtracted in Eq. (13); **c**, **d**. in Eq. (14).

the left of the centrosymmetric point respectively. q_1 and q_2 are interrelated as

$$q_2 = 1/q_1 \tag{12}$$

according to their definition due to Eq. (11).

Which additional opportunities arise because of the asymptotic complement property shows the following[1] analytical example. We find the asymptotical behaviour of the Temkin isotherm at big q:

$$\vartheta(q\to+\infty)\sim\frac{kT}{2\sigma}\left\{\log\left[qe^{\sigma/(kT)}\left(1+\frac{1}{q}e^{-\sigma/(kT)}\right)\right]-\log\left[qe^{-\sigma/(kT)}\left(1+\frac{1}{q}e^{\sigma/(kT)}\right)\right]\right\}\dots$$

Expanding logarithms into terms and then into infinite series, we obtain

$$\dots=\frac{kT}{2\sigma}\left\{\frac{2\sigma}{kT}+\frac{1}{q}e^{-\frac{\sigma}{kT}}-\frac{1}{2q^2}e^{-\frac{2\sigma}{kT}}+\dots-\frac{1}{q}e^{\frac{\sigma}{kT}}+\frac{1}{2q^2}e^{\frac{2\sigma}{kT}}-\dots\right\} \tag{13}$$

$$=1-\frac{kT}{\sigma}\sum_{m=1}^{\infty}\frac{(-1)^{m-1}}{mq^m}\sinh\left(\frac{m\sigma}{kT}\right)$$

In order to find $\vartheta(q\to0)$, one needs to apply the asymptotic complement property (11) according to that

$$\vartheta(q\to0)\sim1-\vartheta(q\to+\infty)=1-\vartheta\left(\frac{1}{q}\right)\Big|_{q\to0}=\frac{kT}{\sigma}\sum_{m=1}^{\infty}\frac{(-1)^{m-1}q^m}{m}\sinh\left(\frac{m\sigma}{kT}\right) \tag{14}$$

The convergence radius of both expansions is $R=\exp\left(-\sigma/(kT)\right)$, i.e. the higher temperature and the narrower the surface heterogeneity range are, the greater the convergence radius is. Its least upper bound is reached in the centrosymmetric point: $\sup R=q^*=1$. The hyperbolic harmonic components in Eq. (13) and (14) are presented in Figure **2**. The asymptotic complement property provides mapping of each m-th harmonics in Figure **2a** and **2b** onto the harmonics with the same index in Figure **2c** and **2d** respectively.

This means that upon determining in experiment the isotherm's points at one side of the centrosymmetric point one may not need to continue experimental determination of the isotherm's points at the opposite side of the centrosymmetric point since the asymptotic complement property (11) enables one to calculate them on the basis of the data determined first.

For the reference purposes we note here, that the Freundlich isotherm is sure not to possess the asymptotic complement property.

The Temkin isotherm obtains the following form in dimensionless units upon integrating in Eq. (10):

$$\vartheta=\frac{kT}{2\sigma}\log\frac{1+q\cdot\exp\left(\frac{\sigma}{kT}\right)}{1+q\cdot\exp\left(-\frac{\sigma}{kT}\right)} \tag{15}$$

Simple mathematical analysis shows that $\vartheta(q)$ doesn't have any inflection point in normal coordinates. It's easy to check that its 2nd derivative appears to be zero only when $\sigma=0$, otherwise $\vartheta''_{qq}<0$. Another pattern takes place in semilogarithmic coordinates whose popularity in relation to the isotherm's simplified form was already mentioned above (Eq. (6) and thereafter). The isotherm equation (15) reads then

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[1] Symbol "~" means hereinafter that the ratio of what is present on the left and on the right of "~" is equal to 1 in the limit considered, i.e. this symbol denotes an asymptotic approximation for the function on the left.

$$\vartheta = \frac{kT}{2\sigma}\log\frac{1+\exp\left(\frac{\sigma}{kT}+y\right)}{1+\exp\left(-\frac{\sigma}{kT}+y\right)}$$

where $y \equiv \log(q)$. The condition $\vartheta''_{yy} = 0$ gives the unique solution $y^* = 0$ i.e. the inflection point reads $q = 1$ and coincides with the centrosymmetric point q^* (s. also [21] for more details). The isotherm curve in semilogarithmic coordinates on the left of q^* is concave and on the right convex. The pressure in dimensional units that corresponds to the inflection point reads $p^* = a_0^{-1}\exp\left(-\frac{\varepsilon_0}{kT}\right)$. At this point $\vartheta = 0.5$.

Thus, the necessary condition that the Temkin adsorption function is valid in any concrete case arises as follows: the isotherm curves must have one inflection point in semilogarithmic coordinates and no inflection point in normal coordinates.

PRACTICAL CALCULATION OF CONJUGATED POINTS AND ENERGY CHARACTERISTICS OF A SURFACE

Let two points (p_{11},ϑ_{11}) and (p_{21},ϑ_{21}) be obtained in experiments. In order to map them in the sense of asymptotic complement property onto points (p_{12},ϑ_{12}) and (p_{22},ϑ_{22}) on dependence $\vartheta(q)$, one considers the following set of equations based on Eq. (15):

$$\vartheta_{11} = \frac{1}{2\alpha}\log\frac{1+\lambda p_{11}\exp(\alpha)}{1+\lambda p_{11}\exp(-\alpha)} \tag{16a}$$

$$\vartheta_{21} = \frac{1}{2\alpha}\log\frac{1+\lambda p_{12}\exp(\alpha)}{1+\lambda p_{21}\exp(-\alpha)} \tag{16b}$$

where $\alpha = \sigma/(kT)$ is dimensionless half-width of the whole adsorption heat range and $\lambda = \frac{q}{p} = a_0\exp\left(\frac{\varepsilon_0}{kT}\right)$ is the adsorption heat peak factor. One obtains easily from Eq. (16a) an explicit dependence of λ on α

$$\lambda = \frac{1-\exp(2\alpha\vartheta_{11})}{\exp[(2\vartheta_{11}-1)\alpha]-\exp(\alpha)}\frac{1}{p_{11}} \tag{17}$$

and comes by means of a successive substitution into Eq. (16b) to the equation

$$\vartheta_{21} = \frac{1}{2\alpha}\log\frac{1+\frac{p_{21}}{p_{11}}.\frac{1-e^{2\alpha\vartheta_{11}}}{e^{-2\alpha}e^{2\alpha\vartheta_{11}-1}}}{1+\frac{p_{21}}{p_{11}}e^{-2\alpha}.\frac{1-e^{2\alpha\vartheta_{11}}}{e^{-2\alpha}e^{2\alpha\vartheta_{11}-1}}} \tag{18}$$

whose solution represents the true value of α giving rise to the energetic heterogeneity factor σ and to adsorption heat peak factor λ. Coordinates of points p_{12} and p_{22} conjugated with p_{11} and p_{21} respectively can be calculated on the basis of equation[2]

$$p_{i1}p_{i2} = \lambda^{-2} = const \tag{19a}$$

that follows directly from Eq. (12). Consequently, for the centrosymmetric point in dimensional coordinates we have

$$p^* = 1/\lambda \tag{19b}$$

The symmetrical points are then to be calculated according to Eq. (11) as

$$\vartheta_{12} = 1 - \vartheta_{11} \tag{20a}$$

$$\vartheta_{22} = 1 - \vartheta_{21} \tag{20b}$$

To further perform calculations of this kind on the basis of real experimental data, we employ the research results having been published before.

DISCUSSION WITH APPLICATION TO EXPERIMENTAL DATA

As it follows from the above-mentioned applications of the Temkin adsorption model, there are a huge amount of experimental data having been obtained by independent research teams and interpreted in its framework. The following two examples selected to cogently demonstrate applicability of the asymptotic complement property as a tool to more extensively explore experimental data than it was typically done in the Temkin case are related to the essentially different heterogeneous systems. One of them represents a classic model reaction for general understanding the gold catalysis [23] whereas another one is subject to a corrosion process with a huge number of influencing factors.

Adsorption of Carbon Monoxide on Gold

This is a highly relevant example related to intensive interpretation of the experimental results on the basis of the Temkin adsorption model (Figure **3**) [7].

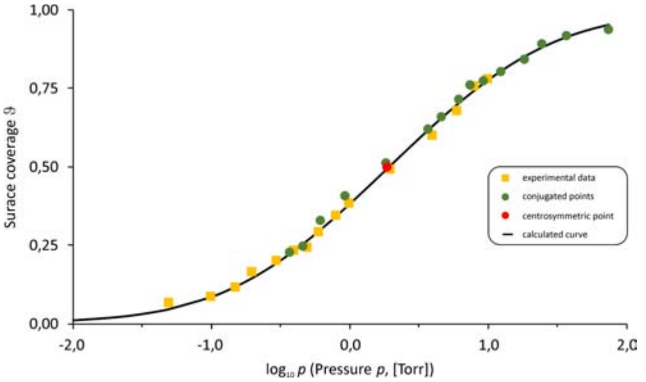

Figure 3: Dependence of coverage of the 1% Au/TiO_2 real-world catalyst surface on pressure of CO (coefficient of determination $R^2 = 0{,}9944$).

The following calculations have been performed. First, combinations of pairs of experimental points were to be chosen in order to calculate α on the basis of Eqs. (17) and (18). Upon statistical processing, averaged

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[2] A relation of this kind as auxiliary one was first formulated [22] for the special case of lateral interactions between adsorbed particles (a Fowler-Guggenheim local isotherm) with a random surface topography and a constant energy distribution.

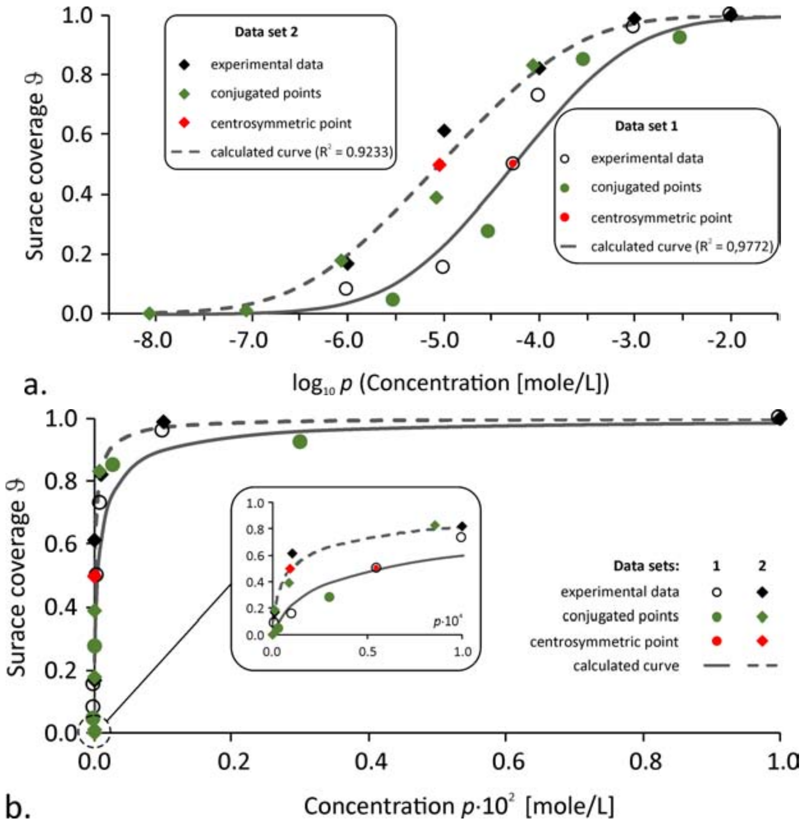

Figure 4: Dependence of surface coverage of steel electrode with and without magnetite coating on concentration of sodium oleate. a) semilogarithmic coordinates; b) normal coordinates. Data sets: 1. non-oxidized steel (6 experimental points); 2. steel with a magnetite coating (5 experimental points).

out final value of α was used for calculation of the final value of λ according to Eq. (17). The calculated curve was then generated[3] using the equation of type (16), the centrosymmetric and conjugated points were calculated according to Eqs. (19) and (20) respectively.

Coverage in the centrosymmetric point calculated appeared to be equal to 0.5 as it follows from the theoretical representation above. Really, one can see that the experimental (yellow) and conjugated with them (green) points are being mapped onto each other referred to the centrosymmetric point (red). Since more points were obtained in the experiment on the left of the centrosymmetric point than on the right, their mapping onto the right-side part of the isotherm reveals the "missing" (i.e. capable of being recovered on the basis of the existing data) points (green) far in the high-pressure area. Provided the Temkin adsorption model remained valid in that area, they would belong to the true isotherm without the need to perform an extended experiment to determine them.

That such a perfect fit of the experimental data with the Temkin adsorption model takes place is likely to be a sequence of that the blurring of a distribution function that is constant in theory of the Temkin adsorption model, is negligible. Needless is to check that there is no inflection of calculated isotherm in normal coordinates (compare with the next case where such a check has been performed). Summing up, we are to state that the experimental data [7] represent a canonical example of the Temkin adsorption model and provide a very clear presentation of the asymptotic complement property.

Adsorption of Sodium Oleate on Steel with and without a Magnetite Coating

This second example concerns a corrosion-related process. There was studied the adsorption of sodium oleate on both oxidized steel and steel with magnetite coating in a borax buffer with pH=7.4 and under natural aeration [24] (Figure **4**). The magnetite coating was produced on the steel electrode in the ammonium nitrate solution with addition of ammonium persulfate (IFHANOKS-3) that, while compared to pure NH_4NO_3, enforces thickening and porosity of the coating.

Questionable was which adsorption model should be used for interpretation of the curves. The Temkin adsorption model found itself among others to be considered.

From the stand-point of the mathematical theory developed above, the fact that the curves experience inflection in normal coordinates (Figure **4a**) and don't do in semilogarithmic coordinates (Figure **4b**) says for

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[3] In terms of work [7], in the framework of the *Heterogeneous Surface Case*.

the Temkin isotherm and therefore witnesses [18] that tailing of the distribution of adsorption centres on adsorption heat that is constant in theory, is here minimal. In other words, its smoothing (compare to Eq. (7) as idealized case) in each of the areas $\varepsilon_0 \pm \sigma$ is also minimal (effectively absent). The adsorption parameters α and λ have been calculated according to Eqs. (17) and (18). The sets of these parameters resulting from trial calculations for different combinations of points were statistically analysed. Those demonstrating the best fit with the experimental data have been chosen.

We have calculated the centrosymmetric point (Eq. (19b)) and the conjugated points (Eqs. (19a) and (20)) as well as the continuous curves itself using Eq. (15). Thus, the dependencies of coverage on concentration have appeared to be prolonged beyond the concentration range in the experiment to the area of small concentrations, by means of a direct mapping of experimental points onto conjugated ones (green in Figure **4**).

CONCLUSIONS AND SUMMARY

Both experiments related to adsorption (1) of carbon monoxide on gold and (2) of sodium oleate on steel (possibly with a magnetite coating) appear to provide a near-perfect illustration of the approach based on the asymptotic complement property to analysis of experimental data. That a higher error level is characteristic for the second experiment is likely to originate from a much more multifactor nature of the system having been studied.

The theoretical results as well as the analysis of experimental data prove the necessary requirements that the Temkin adsorption model really takes place, to be $\vartheta = 0.5$ in the centrosymmetric point and a one-to-one mapping of the isotherm's parts located on the left and on the right of the centrosymmetric point onto one another. This may not be called a *necessary and sufficient condition* at the moment because in order to state this one has to prove that there is no other adsorption model the asymptotic complement in Eq. (11) would be compatible with (of course, provided the Langmuir kernel were valid on the "homotattic surfaces" [25]). However, since the number of examples having been studied so far and based on different distribution functions is limited, we cannot propose a counterexample that would refute such a hypothesis. The Gaussian case studied earlier [21] is certainly not such a counterexample since its asymptotic complement property represents a functional dependence on dispersion of the normal distribution as well as on temperature whereas the asymptotic complement property in the Temkin adsorption model is the absolute invariant. A symmetrically curtailed Gaussian is apparently not such a counterexample either because it inevitably inherits the temperature and dispersion dependence.

While summing up, we can ascertain a variety of analytical strategies arise on the basis of the asymptotic complement property that are useful for interpretation of experimental data. Just some of them may include (*i*) verification of the behaviours of adsorption functions at $q \to 0$ and $q \to \infty$ that appear to be interrelated with one another in the framework of the Temkin adsorption model, (*ii*) given the overall asymptotic complement value that is invariant and equal to 1, one can map experimental data, (*iii*) upon determining a centrosymmetric point one can calculate the surface heterogeneity factor, (*iv*) having uncovered the Temkin adsorption model while studying the heterogeneous system, one can optimize/reduce amount of experimental data to be obtained. What is still missing can be easily restored either exactly by means of calculations or estimatively in the framework of a graphical analysis, (*v*) to check in general if the Temkin adsorption model (or an assumed type of the surface energetic non-uniformity) really takes place in a case under consideration.

Finally, we have to state on the basis of the results delivered above that a stochastic blurring of functions of distribution of adsorption centres on adsorption heat seems to have a more complex origin than simply nobleness of a metal.

ACKNOWLEDGEMENTS

One of the authors (M.V.) is grateful to Prof. C. Pursell (Triniti University) for granting the experimental data in a digital form and to Prof. Yu. Kuznetsov (The A.N. Frumkin Institute of Physical Chemistry and Electrochemistry, Moscow) for valuable discussions of results.

This work was partly (L.T.) supported by Russian Science Foundation, Project No. 18-16-00006.

THE CONFLICT OF INTEREST STATEMENT

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Received on 31-01-2020 Accepted on 18-02-2020 Published on 03-04-2020

DOI: https://doi.org/10.6000/1929-5030.2020.09.02