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IMPACT OF BRUNAUER EMMETT TELLER ISOTHERM ON RESEARCH IN SCIENCE CITATION INDEX EXPANDED

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Abstract

A bibliometric analysis along with a brief historical and modifications overview and scientific applications was carried out to reveal the impact of Brunauer-Emmett-Teller (BET) isotherm on scientific research. The data was based on the Science Citation Index Expanded (SCI-EXPANDED) database of the Thomson Reuters Web of Science. BET isotherm received a total of 10,418 citations from its publication to 2012. Among them, 9,117 (88%) were research articles by 20,108 authors with 95% manuscripts in English. Geographical distribution revealed that North America was the most productive continent, while Africa contributed the least citations. In terms of the institutions and research areas, Spanish National Research Council of Spain and chemistry took the lead. Citations on nanoscience and nanotechnology and environmental science categories increased significantly in the last five years. *Journal of Colloids and Interface Science* published the most BET isotherm cited articles, while "adsorption", "surface", and "properties" were the most frequently used words in title.

Key words: adsorption, bibliometric, BET isotherm, physical chemistry

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1. Introduction

The academic strength of a publication could be quantitatively measured by the number of citations as it reveals how a research is acknowledged among scientific community. Citation testifies scholarly contribution or research visibility (Lefaivre et al., 2011). Without publication, science is dead (Piel, 1986) and a study without citation is like a body without soul. There are many databases providing citation records, but the most acclaimed among the scientific community is the Thomson Reuters Web of Science database. The database latest journal citation system is termed as Science Citation Index Expanded (SCI-EXPANDED) which is one of the databases available under the banner of Web of Science (Oxley, 1998).

In recent years, highly cited works have been investigated in water resources (Chuang et al., 2011), chemical engineering (Ho, 2012), environmental sciences (Khan and Ho, 2012), and adsorption (Fu et al., 2012) research fields. The classic works are of particular importance as their citation count is an indication of high impact and visibility to a broad range of research community (Wohlin, 2005). Most of the highly cited works lead to subsequent advances in molecular biology (Picknett and Davis, 1999), providing beneficial effects to human race. Highly-cited articles nevertheless provide an interesting and useful insight into which authors, articles and topics are influencing the research profession over time (Smith, 2008). An isotherm study was initiated when Boedeker in eighteenth century postulated an isotherm for acidic and basic radicals adsorption onto soils (Boedeker, 1859). It was reported that the most frequently cited article in adsorption field was "adsorption of gases in multimolecular layers" by Brunauer et al. (1938) in

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the *Journal of the American Chemical Society* in 1938 (Fu et al., 2012). This is well-known and the most frequently applied multi-layer adsorption isotherm, so-called BET (Brunauer, Emmett, and Teller) isotherm. The article had high annual citation growth rate, and thus great vitality. The BET isotherm has had a strong and lengthy influence on the adsorption field. The article had not only the highest total citations but also the highest citations in 2011 in adsorption field (Fu et al., 2012).

In this work, we characterized the impact of BET isotherm, using SCI-EXPANDED database.

2. Methodology

The documents information used in this study was based on the SCI-EXPANDED database of the Thomson Reuters Web of Science. According to Journal Citation Reports (JCR) of 2012, it indexes 8,471 journals with citation references across 176 scientific disciplines in science edition. The Web of Science was updated on 31 August 2012. A total of 10,418 citations were found from Web of Science for the original paper for BET isotherm published by Brunauer, Emmett, and Teller (Brunauer et al., 1938). All citations were calculated by Microsoft Excel 2007.

Articles originating from England, Scotland, Northern Ireland, and Wales were reclassified as being from the United Kingdom (UK) (Chiu and Ho, 2005). Federal Republic of Germany (Fed Rep Ger), German Democratic Republic (Ger Dem Rep), West Germany, East Germany, and Germany were reclassified as being from Germany (Ho, 2012). USSR, Bessr, and Russia were reclassified as being from Russia. Italia and Italy were reclassified as being from Italy. Czechoslovakia and Czech Republic were reclassified as being from Czech Republic (Ho, 2013). Belgique and Belgium were reclassified as being from Belgium. Hong Kong and China were reclassified as being from China (Chuang et al., 2011).

3. Results and discussion

3.1. BET isotherm, historical overview and modification

3.1.1. BET isotherm

In 1938, Stephen Brunauer, Paul Hugh Emmett, and Edward Teller affiliated to Bureau of Chemistry and Soils and George Washington University made a remarkable contribution to surface science by formulating an isotherm for the "Adsorption of gases in multimolecular layers" published in *Journal of the American Chemical Society* (Brunauer et al., 1938) was the most frequently cited article (Fu et al., 2012) and was the only one article with more than 10,000 citations in adsorption research field. The BET isotherm has been considered as a fundamental milestone in the interpretation of multilayer sorption isotherms, especially for the types II and III of the Brunauer classification (Brunauer et al., 1940). It has been applied to (ad-)sorption data of gases and vapours on the surfaces and/or porous solids as well as to (ab-)sorption data of vapours, especially water, by polymers and homogeneous materials (Timmermann, 1989). The study was divided into three parts. First, they evaluated the effect of polarization of the second layer of adsorbed gas by the first layer by taking non-polar argon as a model gas. They assumed that the separation of the argon atoms in the adsorbed state is about the same as in the solid state. They observed that the polarization of the second layer of adsorbed gas by the first layer is already too small to constitute the major portion of the binding energy between the two adsorbed layers, at least in those instances in which the gas molecules do not possess considerable permanent dipole moments. To check whether the same force that produce condensation responsible for the binding energy of multimolecular adsorption, they in second part of their work carried out derivation of the isotherm equation for multimolecular adsorption by a method that is a generalization of Langmuir's treatment of a unimolecular layer.

They obtained an isotherm equation as (Brunauer et al., 1938) (Eq. 1):

$$v = \frac{v_m c p}{(p_o - p)\{1 + (c-1)(p/p_0)\}} \tag{1}$$

where: v is the total volume, v_m is the volume of gas adsorbed when the entire adsorbent surface is covered with a complete unimolecular layer, c is a constant, p is the pressure, p_0 is the saturation pressure of gas. An S-shaped isotherm was obtained with these considerations. If constant $c < 1$, then isotherm consists of two regions. A low pressure region which is concave to pressure axis and a high pressure region which is convex to pressure axis.

For $p << p_0$, Eq. (1) could be written as Eq. (2). Further simplification of Eq. (1) will give Eq. (3).

$$v = \left(\frac{v_m c}{p_0} p\right) \Big/ \left(1 + \frac{c}{p_0} p\right) \tag{2}$$

$$\frac{p}{v(p_0 - p)} = \frac{1}{v_m c} + \frac{c-1}{v_m c}\frac{p}{p_0} \tag{3}$$

The BET isotherm in linearized form for solid-solution system is given as Eq. (4):

$$\frac{C_e}{(C_e - C_s)q} = \frac{1}{K_B q_m} + \left(\frac{K_B - 1}{K_B q_m}\right)\left(\frac{C_e}{C_s}\right) \tag{4}$$

where: K_B and q_m are the BET isotherm constants. K_B is the expressive of energy of interaction with the surface and q_m is the amount of solute adsorbed per unit weight of adsorbent in forming a complete monolayer on the surface (mg/g).

The third part of the original paper of BET isotherm covers applicability of the isotherm on a number of catalysts and adsorbents.

3.1.2. Historical overview and modifications

BET (multilayer) isotherm is considered as another milestone towards the development of adsorption science, was proposed to overcome the shortcomings of Langmuir (monolayer). Brunauer and Emmett presented multilayer isotherms in 1930s (Emmett and Brunauer, 1934; 1937; Brunauer and Emmett, 1935; 1937). Initially, BET isotherm was derived from the kinetic considerations (Emmett and Brunauer, 1934). In 1938, BET isotherm was reported by Brunauer, Emmett, and Teller (1938). After that, Cassie (1945) for the first time introduced statistical thermodynamic derivation to BET isotherm model. Further modifications in BET isotherm were made by Pickett (1945) due to limitation of using the isotherm at higher pressures. Modified BET isotherm with heat of adsorption in second and next several layers less than the heat of liquefaction was also investigated by Anderson (1946). Brunauer and co-workers included Deming, Deming, and Teller, proposed an extended BET theory known as BDDT isotherm by introducing additional contribution to the adsorption energy that results from the forces of capillary condensation (Brunauer et al., 1940). The modified isotherm was successfully applied over a wide range of relative pressure unlike BET isotherm. The proposed model deals with the identification of five principal types of adsorption isotherms for gases and vapors. International Union for Pure and Applied Chemistry had accepted it as a basis for a more complete classification of adsorption isotherms.

The notion of multilayer adsorption of gases on substrates to water adsorption on sites offered by electrolytes was studied (Stokes, 1948). It was postulated that in strong electrolyte solutions, the attachment of water to electrolytes could be viewed as an adsorption process, and if this was true, then the water activity should be described by an adsorption isotherm similar in form to the BET adsorption isotherm for gases. The modified BET adsorption isotherm was highly successful in explaining the relationship between water activity and molality of strong electrolyte solutions. This development opened up a completely new avenue for the BET theory. Several decades later, the modified BET adsorption isotherm to explain the properties of bridging electrolytes was developed (Abraham et al., 1993; Abraham, 2004). Later, in 1993, the activities of salts and water, excess properties, and solid phase equilibria in aqueous media by applying the general properties of solutions to extensions of the Stokes-Robinson application of the Brunauer-Emmett-Teller (BET) adsorption isotherm was studied (Ally and Braunstein, 1993). Minimal number of parameters to represent a wide variety of thermodynamic properties with reasonable accuracy over a relatively wide range of temperature and concentration were the major merits of proposed method.

3.1.3. Scientific applications

BET isotherm is a most acceptable standard method to determine specific surface areas and pore size distribution (Sing et al., 1985). The BET adsorption isotherm (for gas adsorption) is obtained by measuring the amount of gas adsorbed across a wide range of relative pressures at a constant temperature. Conversely, desorption isotherm is achieved by measuring removal of gas with reduction in pressure. There are many gases such as argon (Gaines and Rutkowski, 1959; Mayer, 1999), carbon dioxide (Farha et al., 2010; Mahajan, 1991), carbon monoxide (d'Alnoncourt et al., 2003), oxygen (Yu et al., 2000), and hydrogen (Drenan and Hill, 1949; Farha et al., 2010) used as adsorbate for BET isotherm in different circumstances but the most commonly used gas is nitrogen at 77K (Farha et al., 2010; Sing, 2001). To determine specific surface area of a given material only monolayer part of the BET adsorption isotherm is required, but to determine pore size distribution multilayer part of BET adsorption isotherm is essential. In addition, Rains and coworkers have applied BET isotherm to determined mean ionic activity coefficient of water-nitric acid system (Rains et al., 1999). They have selected Davis and DeBruin's activity data as basis of study (Davis and DeBruin, 1964) and in addition the data of Hamer and Wu (1972) was used to confirm the identity of the activity coefficient. They conclude that a model can very accurately predict mean ionic activity coefficients and osmotic coefficients in water-nitric acid system in between 3 and 30 mol range.

3.2. Bibliometric analysis

3.2.1. Document types and language of citations

According to SCI-EXPANDED database, among 10,418 citing BET isotherm documents, 9,117 (88%) were research articles by 20,108 authors. The proceeding paper articles spotted next with 555 (5.3%), followed by reviews with 321 (3.1%). The top three document types cover 96% of the total documents. The document types showing less significance were proceedings papers (n = 163), notes (154), letters (76), meeting abstracts (20), editorial materials (7), discussions (2), book chapter reviews (2), and correction (1). As the document type of article presents new findings along with their interpretation by researchers. Only articles were extracted for the further analysis. Document language plays a major role in transferring research ideas and their implementation. Among citing BET isotherm articles, 8,643 (95%) of the manuscripts were published in English, presenting monopoly of a language among the others. The dominance of English as a major language might be because the most of the journals in SCI-EXPANDED database was in English (Chiu and Ho, 2007). The remaining 5.2% articles were in German (109 articles), French (103), Russian (76), Spanish (63), Japanese (45),

Polish (27), Portuguese (18), Chinese (10), Korean (9), Italian (4), Czech (3), Hungarian (2), Romanian (2), and one article for each of Finnish, Norwegian, and Serbo-Croatian, respectively.

3.2.2. Impact of BET isotherm on countries and institutions

The citing countries and institutions were estimated by the affiliation of at least one author to the cite articles. The global distribution of articles citing BET isotherm was analysed. Altogether, 7,777 articles cited the BET isotherm with address information in the Web of Science originated from 3,984 institutions in 102 countries. According to the geographical distribution in Fig. 1, the maximum number of cited articles was from North American countries and the minimum was from African countries.

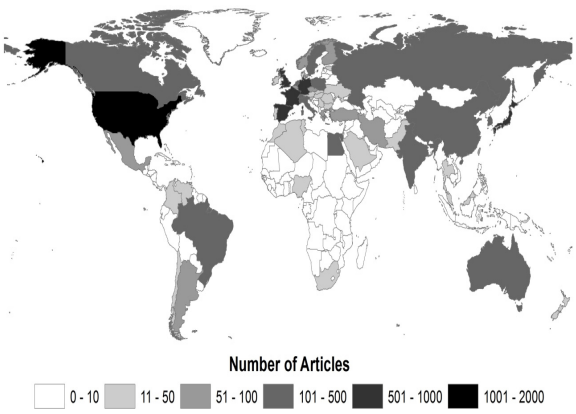

Fig. 1. Distribution of impact of BET isotherm in the world

Ever since the postulation of BET isotherm way back in 1938, a continuous increase in citations was observed, but an obvious increase in citations was observed during last ten years (Fig. 2).

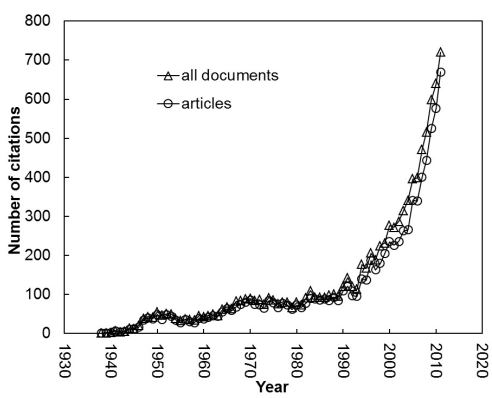

Fig. 2. Impact of BET isotherm since it publication

It has been noticed that citing the original paper not only respects the work of the authors who presented a novel research idea but also discussed this idea in detail in the body of their paper (Ho, 2011). Top 20 countries with BET isotherm cited articles were the USA and Canada from North America; Brazil from South America; France, Germany, Spain, the UK, Italy, Poland, Sweden, Switzerland, Netherlands, and Czech Republic from Europe; Japan, China, India, South Korea, and Russia from Asia; Egypt form Africa; and Australia. Among them, the USA (1,859 articles) has the highest number of citing articles, followed by France (852), Germany (628), Spain (553), Japan (523), and the UK (511). The top ten institutions citing BET isotherm article were the CSIC of Spain (136 articles), the CNRS of France (102), University of Montpellier 2 of France (74), Kent State University of the USA (58), University of Cordoba of Spain (58), Tokyo Institute of Technology of Japan (56), Chinese Academy of Sciences of China (55), University of Estadual Campinas of Brazil (55), Oak Ridge National Laboratory of the USA (53), and University of California, Berkeley of the USA (49).

3.2.3. Journals, Web of Science subject categories and research areas

The BET isotherm covers a broad range of article's subject category on Web of Science. This also reflects the versatile applicability of BET isotherm. The original BET isotherm article was cited by 1,450 journals among 127 science categories and six social science categories in 86 research areas. The top five research areas were chemistry (4,671; 51%), materials science (2,110; 23%), engineering (1,582; 17%), physics (762; 8.4%), and environmental sciences and ecology (581; 6.4%). Out of the 127 Web of Science subject categories in science, physical chemistry articles were at the top of the list covering 2,867 (31%) articles, followed by multidisciplinary material science (1,635; 18%), multidisciplinary chemistry (1,283; 14%), chemical engineering (1,144; 13%), applied chemistry (636; 7.0%), environmental sciences (571; 6.3%), polymer science (466; 5.1%), nanoscience and nanotechnology 430 (4.7%), analytical chemistry (402; 4.4%), and food science and technology (374; 4.1%). Fig. 3 showed annual impact of BET isotherm of the aforementioned subject categories. The postulation of BET isotherm till 2011, the impact on physical chemistry manuscripts was revealed. The impact of BET on newly introduced subject categories is demonstrated in Fig. 4.

Since last five years, the impact of BET isotherm on nanoscience and nanotechnology and environmental science showed appreciable increase.

Among the 1,450 journals, *Journal of Colloids and Interface Science*, a physical chemistry journal led the list with 331 (3.7%) articles, *Langmuir* spotted next with 188 (2.1%), followed by *Microporous and Mesoporous Materials* (175; 1.9%), *Carbon* (147; 1.6%), *Journal of Physical Chemistry* (130; 1.4%), *Chemistry of Materials* (128; 1.4%), *Journal of the American Chemical Society* (126; 1.4%), *Journal of Materials Chemistry* (123; 1.3%), *Geochimica et Cosmochimica Acta* (118; 1.3%), and *Journal of Catalysis* (105; 1.2%).

Twelve of the top 20 journals belong to physical chemistry subject category.

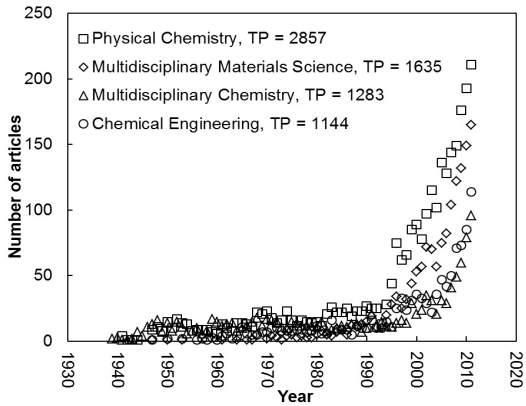

Fig. 3. Impact of BET isotherm on top four subject categories (TP > 1,000)

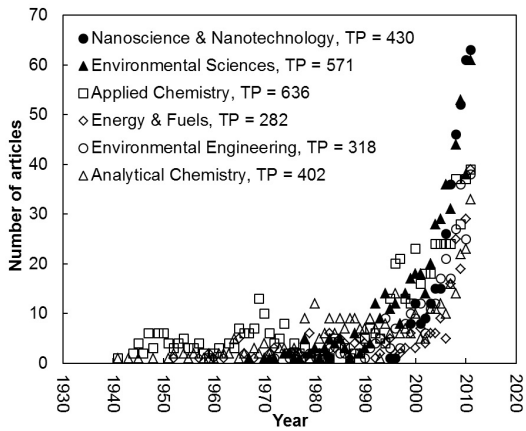

Fig. 4. Impact of BET isotherm in six new categories with more than 30 articles in 2011

3.2.4. Analysis of article titles

The article titles describe research theme in a very precise manner. It could help to evaluate research focuses (Li et al., 2009) as it includes words expressed as an essence of research work by the authors (Tanaka and Ho, 2011). All the single words in the title, have been statistically analysed, discarding prepositions such as "of" and "in" as they are meaningless for research analysis (Zhang et al., 2010). After eliminating these words, twenty most frequently used single substantive words in article titles were traced out.

"Adsorption" 1,501 (16%) citing articles was the most frequently used word, followed by "surface" (959; 11%), "properties" (772; 8.5%), "carbon" (734; 8.1%), "silica" (614; 6.7%), and "water" (613; 6.7%). Insight on aforementioned top five article title words showed that surface science specifically interfacial chemistry was a dominating research area. The title words along with the number of articles and their respective ranks are listed in Table 1.

Table 1. Top 20 most frequently used words

Words in title	*TP*	*Rank (%)*
adsorption	1,501	1 (16)
surface	959	2 (11)
properties	772	3 (8.5)
carbon	734	4 (8.1)
silica	614	5 (6.7)
water	613	6 (6.7)
sorption	578	7 (6.3)
characterization	505	8 (5.5)
synthesis	491	9 (5.4)
mesoporous	462	10 (5.1)
porous	436	11 (4.8)
catalysts	354	12 (3.9)
structure	346	13 (3.8)
preparation	327	14 (3.6)
area	325	15 (3.6)
materials	324	16 (3.6)
influence	295	17 (3.2)
isotherms	273	18 (3.0)
pore	263	19 (2.9)
oxide	249	20 (2.7)

TP: total number of articles

4. Conclusions

The original article for BET isotherm has been cited by 20,108 authors from 3,984 institutions in 102 countries. It has also been cited by 9,117 articles published in 1,450 journals from 127 Web of Science categories and six social science categories in 86 research areas.

Brunauer Emmett Teller Isotherm has been mostly applied in the category of physical chemistry and the research area of chemistry. A significant increase of citations suggested that authors not only still respected the work of the authors who presented a novel research idea but also discussed this idea in detail in their papers.

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