Pore classification in the characterization of porous materials: A perspective

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Abstract: Classification of pores is one of the basic requisites of comprehensive characterization of porous solids. There are various categorizations of pores described in the literature, but it is difficult to give a consistent global classification of porous substances including catalysts, adsorbents, oxides, carbons, zeolites, organic polymers, soils etc. The purpose of each of these classifications is to organize pores in classes by grouping them on the basis of their common characteristics like structure, size, accessibility, shape etc. In this study, a summary of the most used classifications of porous materials is done. Some common properties or behavior for individual classifications could be found, but many differences mainly in pore size are still subject of intensive discussions. Therefore, it is the purpose of this review to provide a general description of the concept and classification of pores in porous solids, to deal with complexity of the matter and to organise our knowledge in decision-making proccesses of pore characteristics determination.

1 Introduction

The importance of porous materials has been recognized since antiquity when porous charcoal was used for its medicinal properties [1]. The worldwide interest [2] in environmental protection and energy conservation has revived the research on porous materials, which have numerous applications such as in catalysis, separation, insulation, sensors, chromatography, etc. A basic motivation for interest and application of pore structures is their ability to promote states of matter, which cannot exist in the external world

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of homogeneous bulk matter [1]. The relative performance of different porous solids in above-mentioned applications are highly dependent on the internal pore structure of each material. Therefore, in order to better understand a particular physical process, taking place within a porous medium, it is necessary to fully characterize the various properties of porous materials like internal geometry, size, connectivity, etc [3].

This paper describes the commonly used classifications of porous bulk materials and existing differences mainly in pore size categorization. For this purpose a summary and discussion have been made in order to organize pores into appropriate classes, corresponding to the basic physical processes that take place in pores, sensitivity of the commercially available analytical techniques and sophistication of the model used to interpret the achieved data. A proposal and a presentation of the unified pore classification close this study.

2   General pore classifications in solids

Numerous studies attempt to categorize pores in solid materials based on defined physical frames [4–8], but many more examples could be found in the literature [9–12]. It is difficult to give a consistent classification of porous structure in solid materials. A useful attempt, at classification based on pore origin, structure, size and accessibility to surroundings Fig. 1 has been made by Kaneko [8]. With respect to their origin and structure, pores are well categorized into two main categories. The intraparticle pores are allocated to individual particles and predominantly are defined as structurally intrinsic pores. In some processes due to the following reactions such as specific evolution, leaching, specific surface reaction, etc. the new porous material could be created. The newly formed pores are classified as injected intrinsic pores. Extrinsic type of intrapores are formed by the reaction in which a foreign substance is impregnated in the parent material in advance and subsequently removed by the above modification procedures. When the material has not been "contaminated" by the foreign substance, which means that the compound is fully eliminated, the newly formed extrinsic pores are called pure type. Another type of extrinsic pores is pillared, produced by the use of pillaring materials such as metal hydroxides. In some cases, extrinsic intrapores could also be interparticle pores [8].

Some porous materials are consolidated, existing as relatively rigid, macroscopic bodies whose dimensions exceed those of the pores by many orders of magnitude. They may be called agglomerates. Others are unconsolidated, being non-rigid, more or less loosely packed assemblages of individual particles, and they may be referred to as aggregates. The particles themselves may be nonporous such as in sand, and therefore surrounded by a network of interparticle voids. These voids have properties dependent only on the size, shape and manner of packing of the constituent particles. In other cases as in spray-dried catalysts, the particles themselves may be significantly porous. This means that a distinction between internal (or intraparticle) voids and interparticle voids should be made. In general, internal pores will be smaller, both in size and in total volume, than the voids between particles. Nevertheless, they will often provide the dominant contribution to the

surface area of the solid [10].

Other bigger arrangements of particles can contribute to introduction of different pore types within the soil structure. These arrangements can be categorized as interaggreation (intracluster) and intercluster pores as mentioned by J. Kodikara [9].
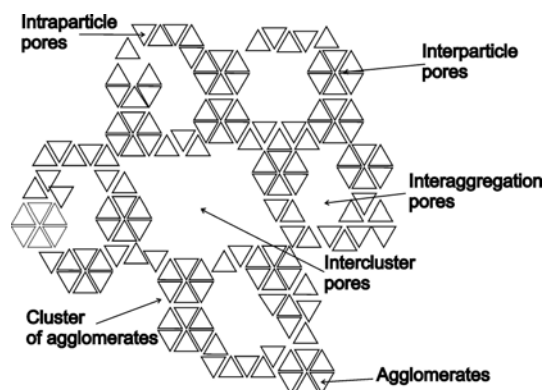

Fig. 1 Structural elements and pore types (modified from Kaneko [8]).

Pores can be also classified according to their accessibility to surroundings Fig. 2. The pores communicating with the external surface are named open pores, like (b), (c), (d), (e) and (f). They are accessible for molecules or ions in the surroundings. Some may be open only at one end (b and f). They are then described as blind (i.e. dead-end, or saccafe) pores. Others may be open at two ends (through pores, (e)). If the porous solids are insufficiently heated, parts near the pores' outer shell collapse, thus inducing closed pores that have no communication to the surroundings. Closed pores (a) are also product of insufficient evolution of gaseous substance. Although the closed pore is not associated with adsorption and permeability of molecules, but it influences the mechanical properties of solid materials.
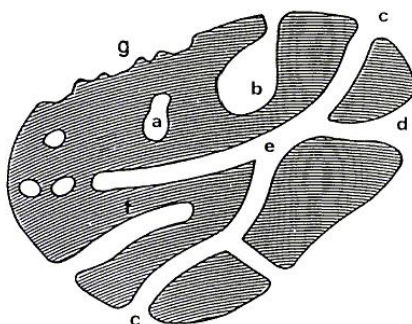

Fig. 2 Schematic pores classification, according to their availability to surroundings (modified from IUPAC [10]) a - closed pores, b, f - pores open only at one end, c, d, g - open pores, e - open at two ends (through) pores.

According to Ruike et al. [11], the term "closed pore" is used for pores that cannot be penetrated by He at 303 K. The other interpretation of the term "closed pore" is an open

pore whose width is smaller than the probe's molecular size (ultrapores). Such effectively closed pores and chemically closed pores should be designated as latent pores [8]. Other possible classifications of pores mentioned in Kaneko's review but used also by IUPAC [10] and detected in the study done by Bindra [13] are based on the pore geometry. According to Kaneko classification, pores are divided based upon the following geometrical shapes Fig. 3: cylinder, slit-shape, cone-shape and ink-bottle. It is almost the same as the shape classification proposed by IUPAC the difference being the funnel shaped instead of the cone-shape geometry as proposed in Kaneko's categorization.
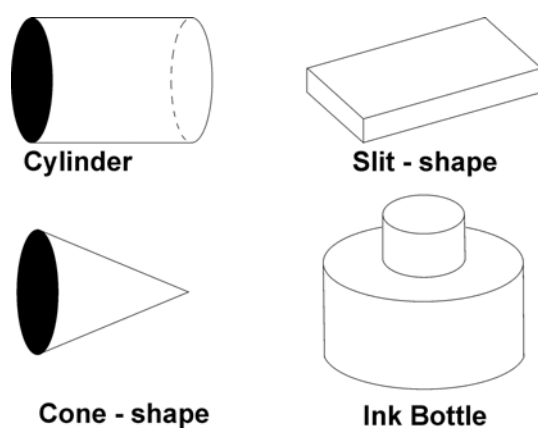

Fig. 3 Pores geometry classification (modified from Kaneko [8]).

Rhomboid, elliptical and square are the other types of pore shapes, which are reported in literature. For simplicity and mainly due to the irregularity in geometry, the pores' shape is often based on model systems. Pore modeled systems are preferably described in terms of different geometric bodies like cylinders (this may be the case for activated oxides like alumina or magnesia), prisms (some fibrous zeolites), cavities and windows (other zeolites), slits (possible in clays and activated carbons), or spheres (although, most often, the pores are, on the contrary, the voids left between solid spheres in contact with each other, as it happens with gels: silica gel, zirconia gel etc [10]. Combination of these forms in pore categorization and their modeling is also used in pore description depending on the arrangement of structural elements. Model development and its application is based on several criteria for full description of porous system such as in terms of geometry, pore size, orientation, location and type of connectivity.

3   Pore size categories

The classification of pores is an important topic. Porous materials with the same apparent porosity, but with pores of different size and geometry react in a different way under the same conditions. Taking these into consideration, questions regarding classification of pores based on sizes have risen.

The "pore size" is a property of major importance in practical applications of porous materials, but it is even less susceptible to precise definition. Pore size has a precise

meaning when the geometrical shape of the pores is well defined and known [10] (e.g. cylindrical, slit-shaped, etc).

There are various categories of pore sizes described in the literature. A summary of the most frequently used pore size classifications is presented in Table 1. It is difficult to give a consistent classification due to differences mainly in ranges of macro-, meso- and micropores, which are still a subject of intensive discussions.

Table 1 Summary of the most frequently used pore size classifications.

Classification	Specified types of pores, d [nm]					
	Macro-	Meso-	Micro-	Supermicro-	Ultramicro-	Submicro-
IUPAC	> 50[4, 16]	$2 \div 50$[4, 16]	< 2[4]; $(0.4 \div 2$[16]$)$	$0.7 \div 2$[4]	< 0.7[4]	< 0.4[16]
Dubinin [5]	$> 200 \div 400$	$200 \div 400 > d > 3 \div 3.2$	$< 1.2 \div 1.4$	$3 \div 3.2 > d > 1.2 \div 1.4$	-	-
Cheremskoj [6]	> 2000	-	$2000 > d > 200$	-	$< 2 \div 4$	< 200
Kodikara [9]	$10^4 \div 10^6$	-	$10^3 \div 3 \times 10^4$	$25 \div 10^3$	$< 3 \div 4$	-

A convenient classification based on Dubinin's [14] early investigations is adopted in Everett [4] and Sing et.al [15]. The classification is for catalysts and divides their pores according to their width i.e. diameter of a cylindrical pore, distance between two sides of a slide-shaped pore or the smallest dimension in fissure pore. They are roughly classified into the following groups: macropores > 50 nm, mesopores in the range of $2.0 \div 50$ nm while micropores < 2.0 nm. It may be desirable to subdivide micropores into those smaller than about 0.7 nm as narrow micropores or ultramicropores and those in the range of 0.7 to 2.0 nm be called supermicropores.

In active carbon, the pores are divided into two general categories: transport pores that constitute macropores with radius > 25 nm and adsorbing pores that are further classified as mesopores ($1 \div 25$ nm), micropores ($0.2 \div 1$ nm) and submicropores (< 0.2 nm) [16]. Coconut shell derived activated carbon (YKAC) and coal based activated carbon (SXAC) used as adsorbents in different pore size measurements show pore size distribution in the mesoporous range of $8 \div 20$ nm and $7 \div 15$ nm, respectively [24]. Other industrially applied sorption materials like zeolites and silica gels show slightly different pore size distribution. The most common commercial zeolites, types A and X used as molecular sieves, have precisely defined ultramicro pore size, which is 0.3, 0.4 and 0.5 nm for KA, NaA and CaA types respectively. The pore opening of the sodium form of zeolite X (13X) is approximately 0.8 nm. As opposed to zeolites, silica gels have larger pores ($1 \div 4$ nm for narrow pore and $6 \div 12$ nm for wide pore silica gel). Silica gels with macro pores up to 300 nm are also known. For investigation of the theory of capillary phenomena mesoporous molecular sieves of M41S type have been synthesized with pore sizes covering the whole mesoporous domain of $2 \div 20$ nm [25].

In discussing the mechanism of adsorption and capillary effects in adsorbents, especially activated charcoals, Dubinin [5] has distinguished the following type of pores:

macropores ($r > 100 \div 200$ nm), mesopores ($1.5 \div 1.6 < r < 100 \div 200$ nm), supermicropores ($0.6 \div 0.7 < r < 1.5 \div 1.6$ nm) and micropores ($r < 0.6 \div 0.7$ nm) where,

r is effective pore radius measured normal to the direction of movement of the molecule during the filling of the pore. The radius itself can serve as a characteristic in the case of the cylindrical pore, and as the half-width in the case of the fissure pore.

Cheremskoj [6] also classified individual pore types by using criteria dedicated to the relative pore size in basic structural elements of the porous material. In this classification several pore types are defined as follows:

Macropores ($r > 1000$ nm) - are specified as pores with size considerably higher than individual element size;

Micropores ($100 < r < 1000$ nm) - are pores with approximately the same size as the structural elements;

Submicropores ($r < 100$ nm) - have pores that are considerably smaller then structural particles;

Ultramicropores ($r < 1 \div 2$ nm) - could be found inside structured elements.

On the basis of evidence from literature, J. Kodikara [9] proposed pore width in clay structures to be divided into the following categories: intercluster (macropores with $10^4 \div 10^6$ nm); interaggregation (micropores with $1 \div 30 \times 10^3$ nm); interpatricle range (submicropores with $25 \div 1000$ nm); intraparticle (ultramicropores with $< 3 \div 4$ nm). Another area, which defines pores, is membrane science technology. Different types of membranes and the separation processes done are also based on pore size distribution or correctly speaking on molecular sieve sizes. Microfiltration ($100 \div 5000$ nm), ultrafiltration ($5 \div 50$ nm), dialysis ($2 \div 5$ nm) and reverse-osmotic membranes ($0.2 \div 0.5$ nm) are well distinguished [17] in this field.

4   Discussion and proposals

The IUPAC classification, in which pores are classified into macro-, meso-, and micropores is mostly based on the different mechanisms occurring in these pores during N_2 isothermal adsorption at 77 K and 1atm. pressure. Multilayer adsorption, capillary condensation, and micropore filling are the processes that relate to macropores, mesopores, and micropores, respectively. The pore width classes correspond to the application of the capillary condensation theory, according to the different relative pressures P/P_O ratio. The pore width of 50 nm is related to the relative pressure of 0.96. Above this value, adsorption isotherms experiments are considerably difficult to be interpreted and applicability of the capillary condensation theory has not been sufficiently examined.

Unstable behavior of the layer of N_2 adsorbed due to tensile strength effect is observed at P/P_O=0.39 and the Kelvin equation cannot be applied to pores having width smaller than 2 nm, which corresponds to this relative pressure. This means that in pore range of $2 \div 50$ nm (mesopores) capillary condensation phenomena is taking place. In the case of so-called micropores (pore width, $w < 2$ nm) the pore filling occurs in a continuous way. The maximum size ($w < 0.7$ nm) for the term ultramicropores corresponds to the bilayer thickness (monolayer on solid surfaces is 0.354 nm) of nitrogen molecules adsorbed, near the entrance of the pores. The gaps between ultramicropores and micropores are termed

as supermicropores (0.7 nm $< w <$ 2 nm).

The range between minimum and maximal micropore radius could be calculated based on the critical diameter of N_2 molecule (d_{cr}=0.380 nm), penetrating into porous space [18]. The minimum micropore width (≈ 0.4 nm) corresponds to the N_2 critical diameter and the maximal (≈ 2 nm) is 2.5 x d_{cr}. So according to IUPAC [16] instead of the maximal micrpore size the micropore range ($w = 0.4 \div 2nm$) has been given. The pores smaller than micropores are named submicropores having width lesser then 0.4 nm.

A new concept of an ultrapore was introduced in materials as proposed by K. Kaneko [8] and M. Ruike [11], where He wass used as gas. Since He molecules are smaller then N_2 an ultrapore designates the smallest open pore through which He can diffuse at 303 K but N_2 cannot at 77 K. It should be remembered that the ultrapore is not the same as the ultramicropore that have a pore width less than 0.7 nm as defined by IUPAC. Their size is less then 0.35 nm, which is a monolayer thickness of N_2 and that is considered in Brown analysis [19] to be a minimum pore size (the thickness of one layer of absorbed molecules) which can be modeled.

The micropores in activated carbon fibers have been classified by M. Ruike [11] into larger and smaller open micropores, ultrapores, and closed pores. Ultra- ($w < 0.35$ nm) and closed pores are those accessible and inaccessible by He at 303 K respectively. In this review closed pores are the ones whose size is impossible to be measured by available analytical techniques. Based on investigations done by K. Kaneko [20, 21] and application of Dubinin-Radushkevich equation, the critical pore width between smaller and larger micropores have been assumed to be in the range of 1.2÷1.5 nm.

Another classical approach has been proposed by Dubinin [5] in adsorbents, especially for activated charcoals. They distinguish between macropores ($r > 100 \div 200$ nm), mesopores ($1.5 \div 1.6 < r < 100 \div 200$ nm), supermicropores ($0.6 \div 0.7 < r < 1.5 \div 1.6$ nm) and micropores ($r < 0.6 \div 0.7$ nm) based on differences in mechanism of adsorption and capillary effects.

Micropores are considered to have dimensions of the molecule being adsorbed. Physical adsorption in these pores is characterized by bulk filling of the space that is available for adsorption and pore volume is so low that the adsorbent - adsorbate interaction could be regarded as a single-phase system.

In order to distinguish the transitional area between micro- and macropores, Dubinin has proposed an introduction of supermicropores with $0.6 \div 0.7 < r < 1.5 \div 1.6$ nm. Movement along such pores involves a gradual alteration from micropore to mesopore properties.

Mesoporous range ($1.5 \div 1.6 < r < 100 \div 200$ nm) corresponds to the upper and lower limits of application of Thomson-Kelvin capillary condensation equation. These pores posses highly developed specific surface on which monomolecular, polymolecular and finally pore filling occur through capillary condensation governed by various factors largely of kinetic origin.

The coarser pores ($r > 100 \div 200$ nm) are therefore classified as macropores, where the adsorption process is negligible and transport processes predominantly occur.

In comparison to IUPAC pore size classification, the Dubinin proposal seems to be precise with respect to adsorption mechanisms and corresponding linear pore size ranges. The term supermicropore in both classifications does not correspond to the same pore size. In IUPAC supermicropores have a width less than micropore. In order to distinguish the slight changes in micropore structure and expanding the role of mesopores, supermicropores have been introduced by Dubinin. Since the prefix "super" can be interpreted as something bigger, the IUPAC recommendation for supermicropores is not stylistically correct. In order to avoid a possible misunderstanding our proposal is to keep the term "supermicropore" and their values as proposed by Dubinin.

Another "hot spot" in the classifications is the position of submicropores. In some papers like [22], the submicropores are called pores with diameter less than about 1.5 nm, i.e. below Dubinin micropores. In case no other subdivision of micropores is presented submicropores are considered to be the smallest pores that exist. According to IUPAC classification these pores are considered to be the smallest, being even smaller then ultramicropores.

Shields and Lowell [23] have additionally referred submicropores to ultramicropores. This is in contradiction with the IUPAC classification and could be misleading in the interpretation of the results achieved in porous material investigations.

The Cheremskoj [6] classification clearly placed ultramicropores as pores having size smaller then submicropores. This classification is well applicable in case of investigations aiming to reveal the physical aspects of porous phenomena and interactions of pores with other defects in investigated structures in terms of vacations, dislocations, etc [18]. In our unified pore size classification presented in Table 2 Cheremskoj approach has been taken into consideration.

Table 2 Unified pore size classification.

Pore types	Macro-	Meso-	Supermicro -	Micro-	Submicro-	Ultramicro-	Subultramicro-
Pore size, nm	$d > 50$	$50 > d$ $d > 3 \div 3.2$	$3 \div 3.2 > d$ $d > 1.4$	$d = 1.2 \div 1.4$	$1.2 > d$ $d > 0.7$	$0.7 > d$ $d > 0.35$	$d < 0.35$

Comparing the pore size of submicro- and ultrapores < 0.4 nm and < 0.35 nm respectively given by IUPAC [16], Kaneko [8] and Brown [19] no huge difference is observed and our proposal is to keep the Kaneko and Brown pore size value but to use the term "subultramicropore". For micropores, the size of < 1.4 nm is proposed by us which also corresponds to 4 monomolecule layers N_2 on solid sorbents. Multilayer adsorption phenomena are applied for bigger values which is typical for mesopores.

Despite the high differences in comparison to Dubinin classification, in most of the revised papers the IUPAC classification regarding meso- and macropores size has been used. The preferences are mainly due to practical reasons because in relatively higher pressures than 0.96 (pore size 50 nm) the interpretation of the isotherms achieved is complicated. So in our approach of classification mesopores fall in the range of $3 \div$

$3.2 < w < 50$ nm at which hysteresis is observed and pores show capillary condensation. Macropores are considered to be higher then 50 nm.

5 Summary

In this paper original classification of porous materials is proposed. A comparison has been used in order to group the pores in different categories with respect to their origin, structure, pore size etc. At present, it appears that pore size is the main parameter characterizing porous materials. No consistent agreement was observed mainly between proposed pore size ranges in individual classifications. A general proposal for pore types and size classes is summarized and presented in Table 2. It contains several micropores types as well as meso- and macropore ranges based on physical assumption and interpretation of adsorption isotherm data.

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