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Structural elucidation of silica present in kidney stones coming from Burkina Faso

Élucidation structurale de la silice présente dans des calculs rénaux prélevés au Burkina Faso

Arnaud Dessombz [a, b, *], Gérard Coulibaly [c], Brahima Kirakoya [c], Richard W. Ouedraogo [c], Adama Lengani [c], Stéphan Rouziere [b], Raphael Weil [b], Lise Picaut [b], Christian Bonhomme [d], Florence Babonneau [d], Dominique Bazin [b, d], Michel Daudon [e]

[a] *INSERM, UMR 1138, Laboratoire de physiopathologie orale moléculaire, Centre de recherche des Cordeliers, équipe Berdal, Université Paris-5, Université Paris-6, Université Paris-7, Paris, France*
[b] *CNRS, Laboratoire de Physique des Solides, Université Paris-11, 91405 Orsay cedex, France*
[c] *Service de néphrologie et hémodialyse, CHU Yalgado Ouédraogo, Ougadougou, Burkina Faso*
[d] *Sorbonne Universités, UPMC UNiversité Paris-6, ULR CNRS 7574, Laboratoire de chimie de la matière condensée de Paris (LCMCP), Collège de France, 11, place Marcelin-Berthelot, 75005 Paris, France*
[e] *AP-HP, Hôpital Tenon, Service d'explorations fonctionnelles, 4, rue de la Chine, 75970 Paris cedex 20, France*

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ABSTRACT

Hundred kidney stones obtained from the University Hospital of Ouagadougou (Burkina Faso) were finally characterized by a panel of complementary spectroscopic and diffraction tools. The most surprising result is the high occurrence of opaline silica as a component in these kidney stones. Opaline silica is a scarce mineral phase in renal calcification; however, we found that at least 48% of the stones had a detectable proportion of silica. SEM images demonstrate the presence of micrometric objects (of spheroidal shape) in close association with monohydrated calcium oxalate crystals. X-ray fluorescence, XRD and ^{29}Si solid state MAS NMR demonstrate unambiguously the presence of amorphous silica, whose composition is comparable to that of natural opals. As NMR is a local spectroscopic probe, other nuclei can be probed. We demonstrate that traces of aluminium are present in the kidney stones by using ^{27}Al solid state MAS NMR. These experiments may offer the first clues of pathological processes that are responsible for these stones.

Mots-clés:
Calculs rénaux
Silice opaline

RÉSUMÉ

Cent calculs rénaux obtenus du CHU de Ouagadougou (Burkina Faso) ont été caractérisés par une série d'outils spectroscopiques et de diffraction complémentaires. Le résultat le plus marquant a été la très haute prévalence de la silice opaline comme composant de ces

* Corresponding author.
E-mail address: arnaud.dessombz@crc.jussieu.fr (A. Dessombz).

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calculs. La silice opaline est un composant rare des calculs rénaux. Cependant, nous l'avons trouvée dans près de 48% des calculs, dans des proportions suffisantes pour être détectables par analyse infrarouge. Les images au MEB démontrent la présence d'objets micrométriques (de forme sphérique) en association intime avec des cristaux d'oxalate de calcium monohydraté. La fluorescence des rayons X, la diffraction des rayons X et la RMN du solide du ^{29}Si démontrent sans ambiguïté la présence de silice amorphe, de manière comparable au cas des opales naturelles. Comme la RMN est une sonde spectroscopique locale, d'autres noyaux peuvent être détectés. Nous avons ainsi démontré que des traces d'aluminium sont présentes dans les calculs rénaux étudiés en utilisant la RMN du solide du noyau ^{27}Al. Ces expériences peuvent offrir les premiers éléments de réponse concernant le processus pathologique conduisant à la formation de ces calculs.

1. Introduction

Renal lithiasis is a consequence of biocrystallization [1–5] of different chemical compounds in the urinary tract corresponding to various pathologies encompassing genetic disorders [4], infection [5], acquired metabolic diseases and most often metabolic risk factors related to dietary habits [6]. Among these relations with pathologies, the prevalence of calcium oxalate (CaOx) stones is associated with significant changes in the dietary habits in industrialized countries [7,8]. In contrast, the presence of silica in kidney stones is quite rare and often related to the administration of drugs [9–13]. None of the large series of stone analyses reported in the literature has identified silica among the chemical phases present in kidney stones.

Recently, a set of 100 consecutive kidney stones coming from the CHU of Ougadougou (Burkina Faso) has been analyzed following the classical analysis procedure based on morphologic examination combined with Fourier-transform infrared (FTIR) analysis [14]. In a significant number of these kidney stones (48%) [15], the presence of amorphous silica has been observed. Silica was present in the nucleus of 42 stones (42%) and was especially frequent in the core of stones from women (72.7% of cases). Moreover, while the content of silica is usually low, silica was the major component of 18% of the stones. Such unusual prevalence of silica in kidney stones has motivated an investigation through physical techniques in order to describe precisely the status of Si atoms.

We and other authors have already demonstrated in different studies the advantages of using physical methods to precisely characterize kidney stones and more generally pathological calcifications [16–18]. Among them, we can quote scanning and transmission electron microscopy (SEM, TEM) [19,20], in situ atomic force microscopy [21], X-ray and neutron scattering [22–24], atomic absorption spectrometry [25], infra-red and Raman vibrational spectroscopies [26–30], micro-computed tomography [31,32], fluorescence induced by X-ray or neutrons [33–35], Nuclear Magnetic Resonance [36–39] as well as techniques specific to synchrotron radiation [40–45].

In this paper, we gathered structural information on silica present in a set of selected kidney stones in order to assess its biological origin. For this purpose, we initially determined stone composition through FTIR experiments, confirmed the amorphous state of silica through X-ray scattering, underlined the presence of different elements through X-ray fluorescence and collected images of different kidney stones at the mesoscopic scale by SEM. Finally, valuable structural information regarding the silica phase was gathered through solid state NMR. This spectroscopic technique, which is a local probe in nature, can bring invaluable structural information regarding the environment of Si [46].

2. Materials and methods

2.1. Patients

We investigated 100 kidney stones collected after open surgery ($n = 99$) or spontaneous expulsion ($n = 1$) from the University Hospital of Ouagadougou (Burkina Faso). The patients were 85 adults (64 men aged 47.0 ± 18.5y and 22 women aged 39.1 ± 15.3y) and 14 children (10 boys aged 9.8 ± 4.7y and four girls aged 11.4 ± 5.5y). All participants, and legal tutors for minor children, gave their verbal consent for use of the material. Ethical approval for the study was obtained from the ethics committee of the Tenon Hospital.

2.2. FTIR (FTIR) spectroscopy

All the samples have been characterized by Fourier transform infrared (FTIR) spectroscopy. To do so, an FTIR spectrometer, Vector 22 (Bruker Optics, Champs-sur-Marne, France), was used according to the analytical procedure previously described [47,48]. Data were collected in the absorption mode between 4000 and 400 cm^{-1} with a resolution of 4 cm^{-1}.

2.3. Scanning electron microscopy (SEM)

A Zeiss SUPRA55-VP SEM was used for the observation of the microstructure [49]. This field-effect “gun” microscope (FE-SEM) operates at 0.5–30 kV. High-resolution observations were obtained by two secondary electron detectors: an in-lens SE detector and an Everhart-Thornley SE detector. To maintain the integrity of the samples, for both SEMs, measurements were taken without the usual deposits of carbon at the surface of the sample.