Some Challenges in the Naming and Measuring of Nanocellulose

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Due to its renewable nature, its inherent strength, and many other favorable attributes, nanocellulose (NC) has drawn increasing attention for many potential applications. A diverse and complex assortment of NC products have been reported, and these are most commonly classified based on some contrasting procedures of preparation. The research community is facing a continuing challenge to adequately measure and quantify morphological features of various NC products. In principle, it ought to be possible to quantify and name NC based on such attributes as "degree of branching," "breadth of particle size," and "aspect ratio distribution," *etc.* However, the ability to measure and compute such quantities still lies beyond what can be achieved in practical amounts of time in typical laboratories. Meanwhile, there has been tension between researchers proposing additional descriptive names, while at the same time there have been efforts at achieving uniformity and simplicity in nomenclature. It is proposed in this essay that this state of affairs is largely a reflection of complexity itself, such that NC products that have the same nominal description can be very different from each other when examined closely. The diversity itself may turn out to be a good thing, as researchers work to come up with varieties of NC that can survive an expected relentless competition from existing plastic-based or cellulose-based materials.

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Emergence of a Hot Field of Study

In recent years, research around nanocellulose (NC) has become a hot topic. It has been claimed that NC provides a new platform for sustainable production of a wide range of high-performance products towards applications in engineering plastics, textiles, food, biomedical, paint, adhesives, electronics, and so on. Although it exhibits great opportunities, there are still some challenges related to the development of NC, requiring continuous efforts from a lot of talented scientists.

Challenges in the Logical Nomenclature of NC Products

As is well known, the term "nanocellulose" refers to cellulosic materials having at least one of their dimensions in a nanometer scale. To be regarded as NC, the cellulosic material must have separated fibrils having diameters within the range of about 2 to 100 nm. However, depending on how the material has been prepared, the morphology and properties of a specific specimen of NC specimen can be very different from another. Therefore, in addition to basing terminology on dimensions, many of the most important classifications of NC varieties are based on the processes employed to obtain the material.

As a first means of differentiation, NC can be divided into about five groups: nanocrystalline cellulose (NCC or CNC), nano-fibrillated cellulose (NFC or CNF), micro-fibrillated cellulose (MFC or CMF), bacterial nanocellulose (BNC), and electrospun cellulose (Klemm *et al*. 2011). The cellulose nanocrystals are prepared by digestion of the cellulosic material in a harsh chemical solution, such as concentrated sulfuric acid, which breaks down and dissolves the amorphous regions of the cellulose. By contrast, the fibrillated types of NC are prepared by processes that rely heavily on mechanical shearing. Pretreatments, such as with cellulase enzymes or (2,2,6,6-tetramethylpiperidine-1-oxyl) (TEMPO)-mediated oxidation are popular laboratory methods to decrease the amount of shearing energy that is required to achieve a nano-sized, fibrillar structure. Although the classification according to the process of preparation has a strong relationship to the morphology of the resulting NC, the labels are too rough to describe the real state of NC fully. Some nano-structured cellulose with special morphology, such as tree-like and nano-network, cannot be differentiated by the terms listed above. In addition, there are many modified NCs, with functional groups (such as carboxyl, carboxymethyl, sulphate group, acetyl group, *etc*.) or polymers (such as hemicellulose, residual lignin and others). How to classify and name these NCs is also worthy of further discussion. A more detailed definition about NC varieties can be found in "Nanotechnologies-Standard terms and their definition for cellulose nanomaterial (PD ISO-TS 20477-2017)".

The features of NC are diverse, including the dimension, morphology, aggregation structure, and surface characteristics. Most NC specimens are actually a mixture, with a wide size distribution in most cases (Fig. 1). Although these characteristics of NC are fundamental and should be considered for classification, they are difficult to be defined and described comprehensively. On the other hand, these features of NC are strongly dependent on a variety of factors, such as the resources, pre-treatment, and nano-defibrillation process.

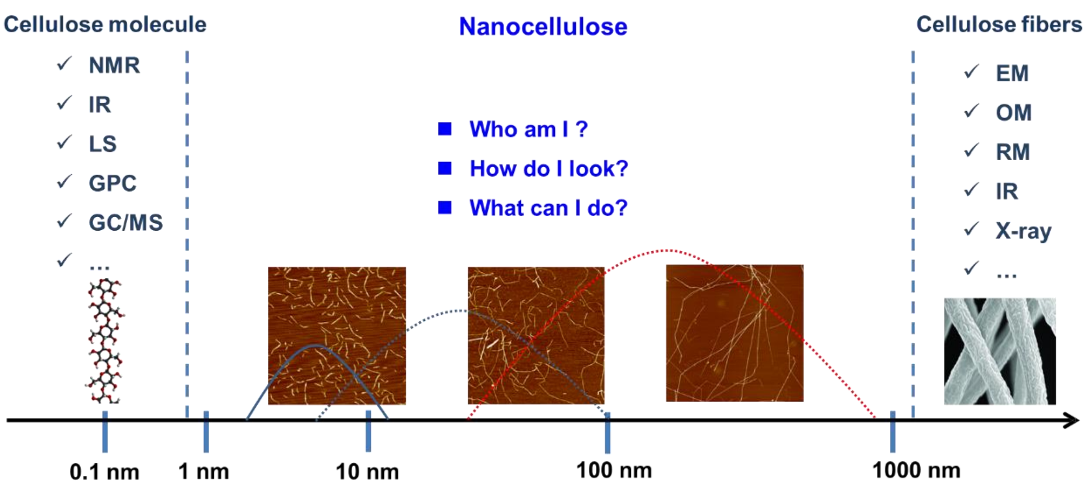

Fig. 1. Some common but important issues about nanocellulose; NMR = nuclear magnetic resonance; IR = infrared spectrometry; LS = light scattering; GPC = gel permeation chromatography; GC/MS = gas chromatography in combination with mass spectrometry; EM = electron microscopy; OM = optical microscopy; RM = rheological measurement; X-ray = X-ray diffraction

Even for the same type of NC, therefore, it is hard to make an accurate and reasonable comparison with the results from the published papers of different groups. In fact, there are only limited features such as average diameter of nanocellulose provided in most works. This reduces the reference value of the research results shared. Thus, the type, structure, and characteristics of NC should be clarified as much as possible for a better understanding and efficient communication.

Challenges in the Characterization of NC

As reported, nanocellulose-based materials could be characterized by numerous techniques. For example, the size and morphology of NC can be examined by microscopy. Evaluations of the chemical and physical properties can also be carried out using X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), thermogravimetric analysis (TGA), and solid-state NMR. However, these characterizations can be regarded as traditional technologies for cellulose fibers or molecules, rather than the direct, precise, and powerful methods for NC. Some of these methods are rendered less useful because NC often is dispersed in an aqueous solution. The drying of NC can drastically change the morphology and aggregate the structure of NC. There has been a need for more effective, rapid, and reproducible NC characterization that can be carried out in the wet state. Dynamic light scattering (DLS) and neutron scattering can provide some information about dispersed NCs, but the underlying theories are mainly based on an assumption of spherical particle shape. Therefore, the results can be misleading when applied to NC. To gain knowledge of the real morphology of NC in the wet state, more efforts should be attempted using *in-situ* measurements. Moreover, it is still a challenge to characterize the surface features of NC, including the content and distribution of functional groups. The more complex the structure of NC is, the more difficult the characterization is. Considering its nomenclature and potential applications, however, it should be necessary to establish more scientific and effective standards for the evaluation and characterization of NC.

Challenges in the Application of NC

Fascinating physicochemical properties of NC, such as transparency, excellent mechanical strength and stiffness, light weight, large surface area, and biodegradability, have allowed the development of NC-based functional materials. In addition, the unique structure of NCs provides them with a strong interaction with surrounding polar species, such as water, nanoparticles, polymeric compounds, living cells, and certain organic compounds. Recently, NC has demonstrated good performance in numerous applications, including specialty paper (Hu *et al.* 2013), building materials, cosmetics and personal care products, hydrogels/aerogels (Lavoine and Bergström 2017), flexible materials, transparent films, coatings, medicines, and high-efficiency filter materials. However, NC seems not to be the sole option for most such potential applications. For example, the functional hydrogels/aerogels can be alternatively prepared by cellulose solvent systems. Acting as a stabilizer and hydrophilic agent, carboxymethyl cellulose is used in most of the compositions of cement and building materials. Therefore, most NC-based materials are faced with relentless competition from existing plastic-based or cellulosic chemical-based materials, and there is a strong need to utilize the irreplaceable value of NC itself by exploiting the inherent nanostructures of NC that cannot be artificially reconstructed. Applications of NC as a kind of "industrial MSG" in various fields are possible but not inevitable. Moreover, the performance of NC-based functional materials is relatively uncontrollable and unstable because of the generally wide distributions of nanoparticle-size and structure. After research and development for decades, the applications of nanomaterials including NC are still limited to some small fields. This is not only due to their relatively high price, but also because of the lack of a clear direction. That is, we should think about the real value of NCs before using them.

Summary and Perspectives

Some issues around NC have been discussed briefly in this editorial, including the nomenclature, characterization, and applications. Obviously, these challenges are not all for NC. These topics are very common, and we are used to receiving the views of most researchers. In fact, NC is a class of unique materials, which are very different from other generally nanomaterials. The conducting of research in NC with its salient properties to meet the world's needs is still an uphill task. Anyhow, taking the time to deeply understand the fundamental knowledge of NC may be beneficial for its development and usage in the future.

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