J Mater Sci (2020) 55:12637–12641

Editorial

1000 at 1000: reflecting on "Review: Current international research into cellulose nanofibres and nanocomposites"

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Published online:
22 June 2020

The path towards high citations of a research paper is a strange and not an always predictable one. Of course, many factors play a role in determining whether a paper receives citations. It is true to say for all journals, including the multidisciplinary, highly cited, some say high impact journals such as *Nature*, *Science*, that review articles attract many more citations than traditional research papers. The cynic in me would say that references to such papers may be an easy way to make general statements about an area, in the opening sentences of a standard paper, without the need to read or understand the bulk of the content of such papers. There may be some truth in this, but good review articles can at least inform readers of what research is out there, and make them aware of the context of the research being undertaken.

Review papers can particularly help postgraduate (or graduate) students, who, let's face it are probably the vanguards of research, to find references for all the vital work in a current area. I cannot remember a time in my career so far when I wasn't more connected to the current and past research on cellulose as I was when I was a Ph.D. student. The story of how I came to publish the review on cellulose nanomaterials and nanocomposites [1] should perhaps start from that moment in my life.

My love for cellulose began with my masters in wood science. Following that, during my Ph.D., following the masters, I witnessed a gradual decline in the paper industry, while in the Paper Science department in UMIST (both these institutions are no longer with us). I could see a gradual move towards using cellulose as a material in other applications, other than paper and textiles. Cellulose

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https://doi.org/10.1007/s10853-020-04961-4

nanocomposites had started to emerge in the mid-to-late 1990s, around the time of my Ph.D. So, timing wise I personally was at the right point to face this new area of research from its outset. I subsequently moved from the Paper Science department over to the Materials Science Department in Manchester. There I initiated a research project into natural fibres and composites. I am indebted at this time to the guidance and mentorship of one of our former editors—Professor Bob Young FRS FREng—who had the idea that I might galvanise all the research I had done, and that others were doing at that time in this area into a review for *Journal of Materials Science*. So, I assembled a group of international researchers who were working on this subject. It was good fortune that the area of natural fibre composites was, and still is, a truly international effort. Homegrown talent included a retired Professor in the Materials Science department, the late Professor Ken Entwistle. Ken, on retirement, had abandoned his long career in 'conventional materials', and was now focussing on wood and the measurement of the fibril angles, the measurement of the micro-fibril angle in soft-wood [2], the derivation of the micro-fibril angle in soft-wood using wide-angle synchrotron X-ray diffraction on structurally characterised specimens [3] and mechanosorptive creep, the recovery of mechanosorptive creep strains [4] properties of the material. I remember with fondness his building of homemade devices to measure creep strains in wood using pen lasers and mirrors! In a prelude to the 2010 article, a paper entitled "Review: Current international research into cellulosic fibres and composites" was published in *Journal of Materials Science* [5]. This proved to be the template and winning formula for the later article, itself receiving a lot of attention.

Wood is perhaps a good starting point to discuss why the 2010 article both came about, and why it has received so many citations. The word 'material' derives from the late Latin word *māteria*, which has another meaning of 'wood', and takes its root from the word *māter*, which means 'mother'. Etymology is important here since our concepts of materials are rooted in the natural world, and in times when sustainability issues are high on the agendas of many countries and governments we do well to remember these connections. Materials Science, at least how it is conventionally taught, however often makes little reference to wood, or natural composites in general. Indeed, in the days when Ken and I were working on the topic, it was seen by our colleagues as somewhat of an oddity amongst conventional materials. I recall one other retired Professor at the coffee break in Materials teasing Ken jokingly for his interest in wood. Ken pointed out that there were few engineering materials that could boast such a range of mechanical properties—a near times twenty range of mechanical stiffness from within one tree! This is something to be desired for engineering materials.

Wood is of course the perfect composite material, and possessing a hierarchical structure [6], it has nanoscale features that we are at pains to replicate with our own synthetic analogues. The growth of interest in cellulose-based nanocomposites is no doubt inspired by this natural structuring seen in wood, and so it was in 2010 when I came to write and assemble "Review: current international research into cellulose nanofibres and nanocomposites".

Another factor in a review article, or any publication for that matter, gaining a high number of citations is of course timing. In 2010, we were perhaps witnessing an upward trend in the publication of articles relating to cellulose nanocomposites, nanomaterials, etc. The graph in Fig. 1 perhaps demonstrates this growth in the area, although it is noted that other areas show similar growth trajectories. The arrow pointing to 2010 just highlights the argument for timing, since this indicates when the article appeared, just at the beginning of the upward trend. Timing is, of course, often serendipitous, and who knows where to, and how a new area of research will develop. It is true that areas reach their maximum, and it may be true that this is already taking place for the 2010 article; Fig. 1a shows evidence for a tailing off of papers published in this area. I am all too used to graphs like this showing near exponential growth, in the middle of the Covid-19 epidemic. In fact, mathematical models based on Markovian processes of 'infection' can be applied in the situation of a 'birth–death' lifetime of a published article. Cumulative citations to an article over its 'lifetime' will ultimately follow a sigmoidal curve—the peak of which can be predicted using models—as can be seen from Fig. 1b. The predicted number of citations will likely reach no more than around 2200 on current data, although the odd citation could appear every now and then ad infinitum. Inevitably articles get "forgotten", especially when they set their sell-by-date by using the word 'Current' in the title!

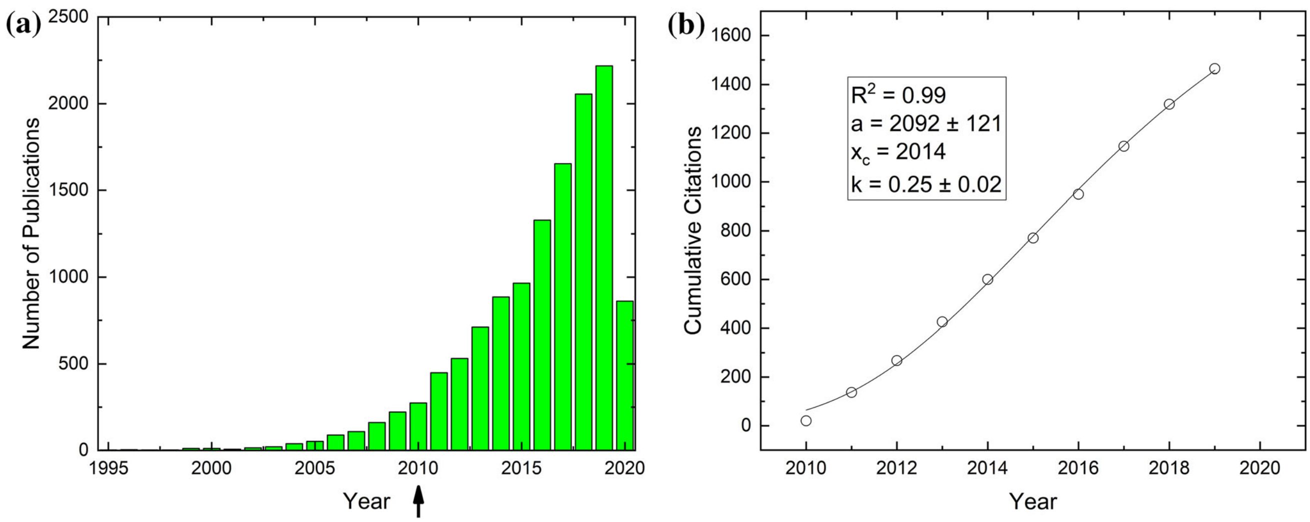

Figure 1 **a** The number of publications by year using the search criteria "cellulose and (nanocomp* or nanomat*)". *—wildcard notation. Arrow to highlight 2010. Data retrieved from Web of Knowledge (21/05/2020); **b** Cumulative citations to the 2010 review article "Review: Current international research into cellulose nanofibers and nanocomposites" as a function of year. Solid line represents a Gompertz equation fitted to the data. Parameter a predicts the maximum number of citations (asymptote), k is the growth rate, and x_c is a scaling factor for the x-axis.

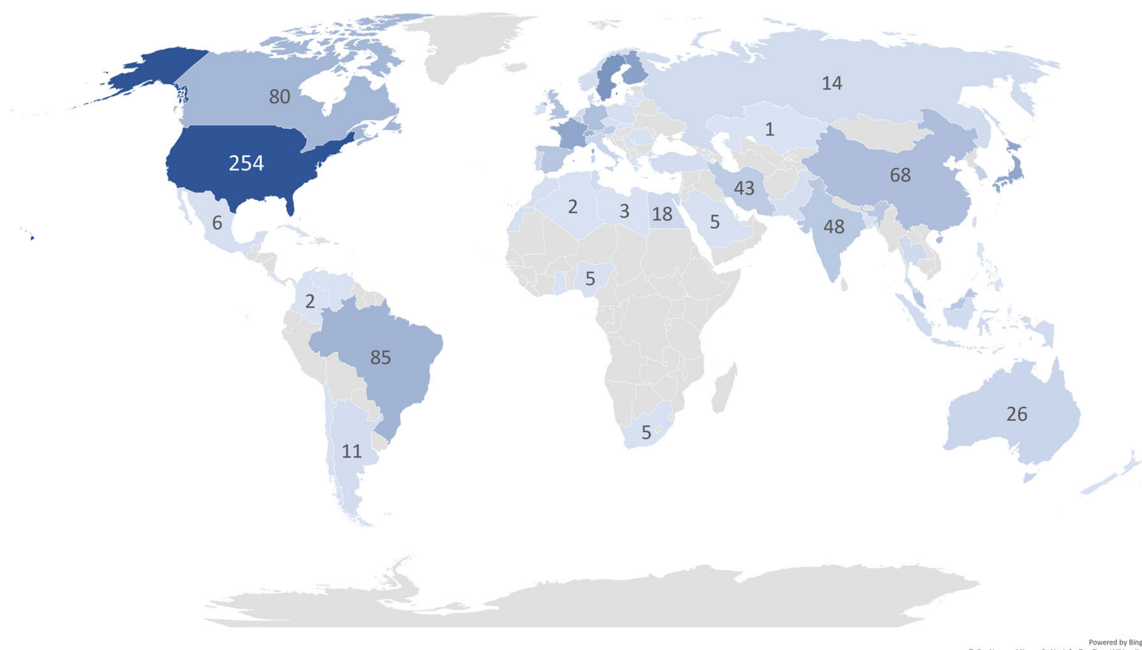

Figure 2 Geographical location of citations to the 2010 review article "Review: Current international research into cellulose nanofibers and nanocomposites". Figures given are extracted from Web of Knowledge (from 21/05/2020).

The article was authored by researchers from across the globe (France, Argentina, USA, Switzerland, UK, Austria, Japan, and Sweden). This range of international authors is perhaps another reason why the article has attracted so many citations (now > 1500). It is well known that papers published with a range of international authors tend to attract a lot of attention, because then the article will typically reach a much wider audience. This reach of the article is reflected in the geographical location of citations on the map presented in Fig. 2.

It is interesting to note that largest numbers of citations are from China (321) and the USA (254)—both countries with large populations, and therefore, high numbers of total publications in this area anyway. A better way to present these data might be *per capita*, but then you would have to define as a publishing population. Other notable countries citing the review are Sweden (142), Finland (118), England (109), France (109), and Japan (107), which compared to the USA and China, are remarkably high given *per capita* considerations. This probably reflects the fact that major activities around 'nanocellulose' are to be found in these countries, with perhaps the exception of England, where there can only be a handful of researchers working in this area—maybe the bulk of those citations are self-citations?

Perhaps more importantly, we should turn to the subject and content of the 2010 review, instead of navel gazing at citations and metrics. In any case, we should be moving away from assessing an article's worth based on metrics, and as a personal signatory of DORA (https://sfdora.org/) I should be upholding this principle. The article contains an introduction, with a reference to Chaucer and 'The Parson's Tale'. Many of my co-authors questioned its inclusion, and reading back I don't think I made a good case for it. The reason for its inclusion was that we have a very ancient attachment to 'wood' and its association with the spiritual. Not being a religious person myself, but mindful that perhaps that natural world connection has been lost somewhere down the line, I thought it was a good idea to point ourselves in that direction. Sustainability is not a new concept in some cultures (*e.g.* aboriginal, First Nation groups). In those cultures, it is most intimately connected to language and natural capital, and none more so in its description of wood. In Japan, people still retain the concept of trees having a spirit, and so the material is revered in that way. Maybe we wouldn't be so careless with our natural resources in the Global North if this was still the case in the so-called developed world? Years later, when I was honoured enough to be receiving the Hayashi Jisuke Prize from the Japanese Cellulose Society, I made reference to this cultural connection with wood, quoting the famous haiku poet Matsuo Bashō:

> "butt of the tree
> see in it the cut end
> today's moon"

So, I believe an article that draws on international cultural references to cellulose, plants, wood, is all important. We are the custodians of the material from which nanocellulose derives, but we should also seek to sustain those cultures which respect their natural capital.

In the introduction, we were also blessed by a late addition of a graph drawn by members of the Queen Mary team showing how the aspect ratio of fibres makes a big difference to their reinforcing efficacy; higher aspect ratios are favoured in this respect, and governed by the Halpin–Tsai equation. I still think this is an important point that is often missed in the literature, with some groups wondering why they don't achieve the reinforcement, and thereby mechanical properties that they would wish for with a relatively low aspect ratio cellulose nanofibre. Following the introduction, the paper was then split into sections, each one highlighting some work being carried out by groups from around the world. Leading the charge on this was a section written by Alain Dufresne (Grenoble Institute of Technology (INPG), International School of Paper, Grenoble, France) on the physical properties of cellulose whiskers and nanofibres, showing the different morphologies that one can obtain from the materials. It is perhaps unfortunate that we did not go into the classifications of cellulose nanofibres enough here, since IUPAC have since declared that "cellulose nanowhiskers" should now be known as "cellulose nanocrystals"—our article consistently refers to the former. Next, there followed a section about cellulose nanofibres and their interactions with polyurethanes, which is still not a typical matrix associated with these materials, but one that can display interesting shape memory properties. Some of our own work on monitoring the deformation of cellulose nanocrystals inside epoxy resin using Raman spectroscopy was then presented. Then, Stuart Rowan (then at Case Western Reserve University, now at University of Chicago) and Chris Weder (also then at Case Western, now at the Adolphe Merkle Institute) presented their work that they published in *Science* [7] on water activated shape memory cellulose nanocrystal-polymer composites, a material that mimics the sea cucumber. Wim Thielemans then introduced the modification of the surface of cellulose nanocrystals, using amongst other things ring-opening polymerisation of ε-caprolactone, grafting of styrene, etc. This is a self-contained area of research, and several subsequent reviews in other journals have covered this topic. A contribution from Virginia Tech (Scott Renneckar—now at UBC, Canada—and Maren Roman) then followed. This section showed how it is possible to fluorescently label cellulose nanocrystals and use them as tags in biomedicine applications. The review then turned to promising applications. I think, and still to this day, that we picked a few quite niche areas of application, including cellulose nanofibres as reinforcement in adhesive joints (for wood) in construction (Wolfgang Gindl and Stefan Veigel, BOKU, Vienna and Josef Keckes, University of Leoben, Austria), optically transparent films for display devices and semi-construction materials (Hiroyuki Yano, Kyoto University, Japan), cellulose/DNA hybrids (John Simonsen, Oregon State University, USA), hierarchical composites (Alex Bismarck, Imperial College, UK—now at University of Vienna), novel foam materials (Lars Berglund, KTH, Sweden), and all-cellulose composites (Ton Peijs, Queen Mary—now at University of Warwick, UK). It is interesting to reflect now, nearly 10 years on what applications of nanocellulose have been developed, and by whom. I am aware that Professor Yano has made a nanocellulose car in very large Japanese industry/academic project [8]. The work on foam materials at KTH led to prototype bike helmets, although not yet mass-produced ones, using nanocellulose-based foam materials [9]. The path towards applications is not a smooth one, particularly where cellulose nanomaterials are still effectively a disruptive material in an existing supply chain for many industries and products. I think if there was to be a repeat review article, then the focus should be on applications. There is still much to be understood about the properties of cellulose nanomaterials though, and we should not lose sight of fundamental research to underpin these developments.

So, on the anniversary of the journal, it is right to celebrate what became, for a while, the most cited paper in the journal ever since its inception. But looking ahead to a world where sustainable materials will play more of a role in our lives, we do well to remember cellulose's place in the natural world, and how we can better make use of its properties that mother (or *māter*, Moeder, Maman, Morsa, Mutter) nature has imbued over millions of years of evolution. Realising this vision is not without some risk that we don't fall into the old traps of exploitation, and the production of yet still persistent materials in our environment that cannot be degraded, recycled, or at best entered into a circular economy. These challenges still present themselves to us for cellulose nanomaterials, and I look forward to the next 10 years of their development.

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