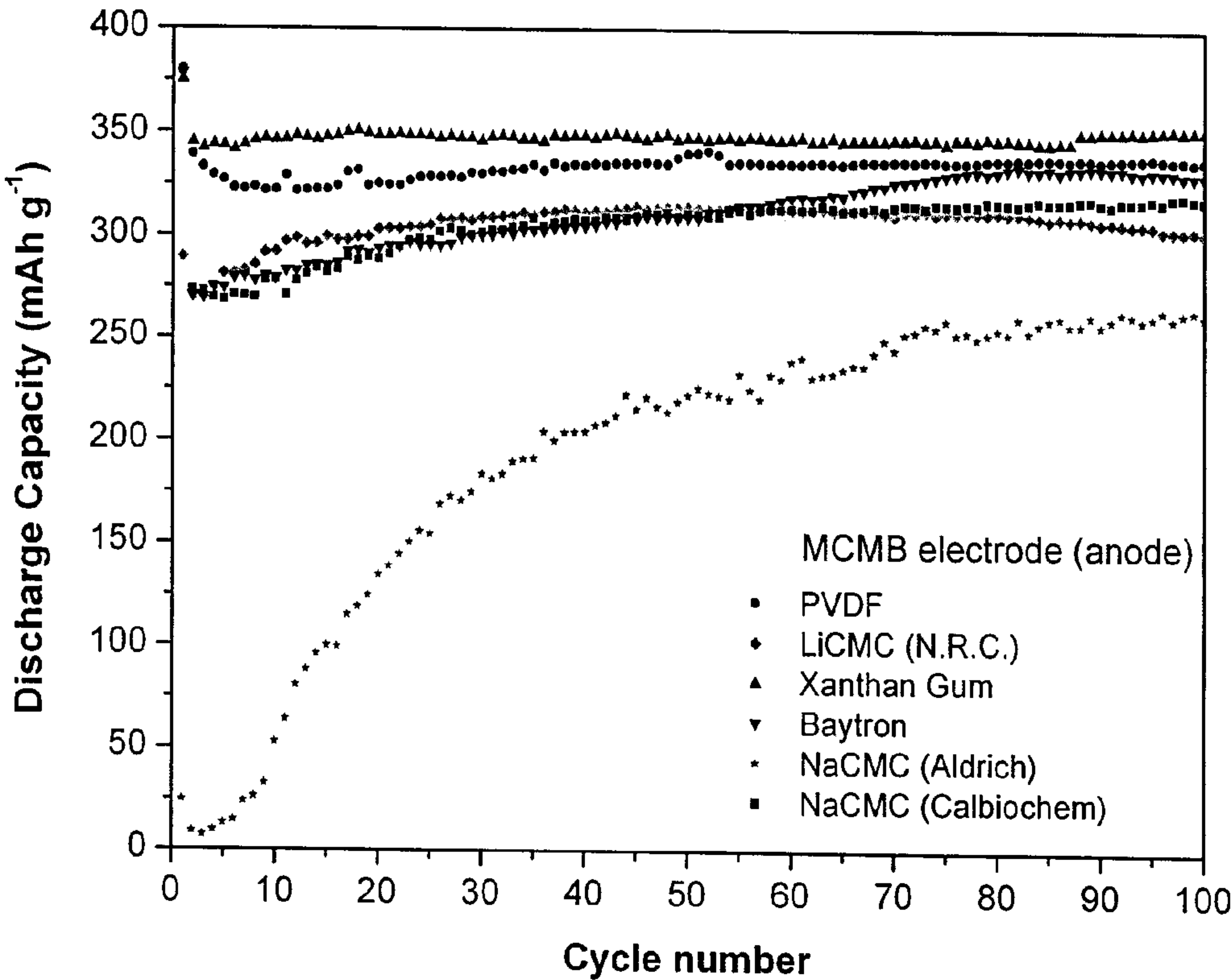




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(54) **Titre : UTILISATION DE GOMME XANTHANE EN TANT QUE LIANT D'ANODE**
(54) **Title: USE OF XANTHAN GUM AS AN ANODE BINDER**



(57) **Abbrégé/Abstract:**
Xanthan gum has been found to be a superior binder for binding an electrode, especially an anode, in a lithium-ion or lithium-sulfur battery, being able to accommodate large volume changes and providing stable capacities in batteries tested with different types of anode materials.

ABSTRACT

Xanthan gum has been found to be a superior binder for binding an electrode, especially an anode, in a lithium-ion or lithium-sulfur battery, being able to accommodate large volume changes and providing stable capacities in batteries tested with different
5 types of anode materials.

USE OF XANTHAN GUM AS AN ANODE BINDER

Field of the Invention

This invention relates to electrode binders, particularly anode binders in lithium-ion or lithium-sulfur batteries.

Background of the Invention

5 In lithium-ion batteries cathodes typically comprise a lithium-containing material and the anode is usually a lithium-free material such as graphite, a metal, a metalloid, or an oxide. In lithium-sulfur batteries, the anode is typically lithium metal and the cathode is made of sulfur or a carbon/sulfur composite. New lithium-sulfur battery devices use a metal/carbon composite as an anode and a polysulfide cathode (e.g. Li_2S). In lithium ion
10 batteries, the anode may be: i) a metal or a metalloid that can alloy with lithium, mainly elements from groups: 2B (Zn and Cd), 3A (Al, Ga and In), 4A (Si, Sn and Pb), 5A (Sb and Bi) and Sn-alloys (Sn-Fe, Sn-Co, Sn-Ni, Sn-Cu, Sn-Zn); ii) hard or soft carbon (e.g. graphite), iii) an oxide whose metal alloys with lithium such as SnO_2 , Sb_2O_3 , and silicon oxide); iv) a transition metal oxide ($\text{Li}_4\text{Ti}_5\text{O}_{12}$, titanium oxide, chromium oxide, manganese
15 oxide, iron oxide, cobalt oxide, nickel oxide, copper oxide and zinc oxide). Lithium also intercalates into nitrides, phosphides and sulfides. In lithium-ion batteries, of particular importance are anodes in which lithium is intercalated into an oxide or an oxide-carbon matrix.

20 Tin oxide reacts with lithium according to the following reaction:



25 Anodes that comprise metal (or a carbon/metal composite or metal oxide) that alloys with lithium suffer from large volume change upon lithiation/delithiation of the electrode during operation and re-charging of the battery. This volume expansion ranges from about 100% for Al to about 300% for Si. In order to accommodate this large volume

change it is necessary either to use nanoparticles or use a binder that can accommodate this volume change. Polyvinylidene fluoride (PVDF) is the conventionally used binder in battery technology; however, it does not accommodate a volume change larger than about 15-20%, such as for graphite or $\text{Li}_4\text{Ti}_5\text{O}_{12}$. PVDF does not get reduced at low potential (5 mV versus Li/Li^+) nor oxidized at high potential (5 V versus Li/Li^+) at room temperature. However, at elevated temperature, it has been reported that PVDF reacts with Li metal and LiC_6 to form LiF and some C=CF species via an exothermic reaction which will cause a risk for thermal runaway (Du Pasquier 1998; Maleki 1999; Maleki 2000). To avoid this risk, research has focused on the use of non-fluorinated binders (Gaberscek 2000; Oskam 1999; Ohta 2001; Zhang 2002; Verbrugge 2003). Even though they are still insoluble in water, reduced heat was obtained when phenol-formaldehyde, poly(vinylchloride) or polyacrylonitrile are used as binders (Maleki 2000; Du Pasquier 1998). Another disadvantage of using PVDF is its price which is about US \$20 per kg in North America (€15-18 per kg in Europe (Lux 2010)). In addition, PVDF requires the use of non-environmentally friendly solvents to process the electrode formation, such as N-methyl-2-pyrrolidone (NMP). Also, it is not easy to dispose of PVDF at the end of the battery life (Lux 2003). Thus more environmentally friendly binders are needed for preparing electrode materials for Li-ion batteries.

Some rubber-based binders such as styrene-butadiene rubber have been tested with some success, but these binders are not water soluble and there is a further need to improve their ability to accommodate volume expansion. Sodium carboxymethylcellulose (NaCMC) is a sugar-based molecule used as a thickener in the food industry and has shown good accommodation of volume expansion in the case of silicon-based electrodes (Li 2007; Buqa 2006; Beattie 2008; Hochgatterer 2008; Liu 2005), and more recently with tin oxide-based electrodes (Chou 2010). In addition to being able to accommodate the volume change, NaCMC is water soluble due to the carboxymethyl groups attached to the cellulose. This avoids the use of non-environmentally friendly solvents during the casting process, which makes fabrication of the electrode easier. As mentioned by Lux *et al.*, the use of NaCMC also makes the recycling of Li-ion battery anodes easier (Lux 2010). Indeed, by heating NaCMC at 700°C , Na_2CO_3 is obtained. In addition (Machado 2003; Kaloustian 1997), the price of NaCMC is much lower than PVDF, about US \$6 per kg in North America (€1-2 per kg in Europe (Lux 2010)).

One approach in the prior art (Satoh 2005; Satoh 2008; Satoh 2009a; Satoh 2009b) for producing negative electrodes for batteries has been to use graphite-based anodes and binders having olefinic unsaturated bonds (e.g. styrene-butadiene rubbers).

In such an approach, the graphite-based anode may be coated with metal oxides. This prior art also suggests that xanthan gum may be used as a co-binder along with the binder having olefinic unsaturated bonds. However, there has been no specific exemplification of the use of xanthan gum and the use of xanthan gum exclusively, i.e. not as a co-binder, has not been suggested. Further, the anode material is restricted to graphite or graphite coated on the surface with metal oxides. Such anode materials do not experience the very large volume expansions that oxide or oxide-carbon matrix materials undergo during the lithiation/delithiation process.

There remains a need in the art for water soluble binders which can accommodate large volume expansions upon lithiation/delithiation of electrodes in lithium-ion or lithium-sulfur batteries.

Summary of the Invention

It has now been surprisingly found that xanthan gum is an excellent water-soluble binder for anodes in lithium-ion and lithium-sulfur batteries, accommodating large volume changes and providing stable capacities in batteries tested with different types of anode materials.

Thus, there is provided a use of a binder consisting essentially of xanthan gum for binding an electrode in a lithium-ion or lithium-sulfur battery.

There is further provided a use of xanthan gum as a binder for an electrode consisting essentially of lithium intercalated into an oxide or lithium intercalated into a homogeneous matrix of an oxide and a conductive carbon.

There is further provided an anode for a lithium-ion or lithium-sulfur battery comprising a lithium-containing material bound by a binder consisting essentially of xanthan gum.

There is further provided an anode for a lithium-ion battery comprising a xanthan gum-bound electrode comprising a lithium-containing material consisting essentially of lithium intercalated into an oxide or lithium intercalated into a homogeneous matrix of oxide and a conductive carbon.

For general use in lithium-sulfur and lithium-ion batteries, the anode binder consists essentially of xanthan gum, where no co-binder is present. In lithium-sulfur batteries, the lithium-containing material of the anode is Li-S. In lithium-ion batteries, anodes may comprise a lithium-containing material, for example, a lithium alloy, lithium

intercalated into a conductive carbon (e.g. graphite, carbon black, mesoporous carbon microbeads, carbon nanotubes, graphene and mixtures thereof), lithium intercalated into an oxide (e.g. aluminum oxide, tin oxide, silicon oxide, cobalt oxide, iron oxide, titanium oxide, copper oxide and mixtures thereof), lithium intercalated into a nitride, lithium
5 intercalated into a phosphide, or lithium intercalated into silicon, or lithium inserted into a compound or composite by displacement. Lithium intercalated into a conductive carbon, lithium intercalated into an oxide or lithium intercalated into a homogeneous matrix of oxide and conductive carbon are preferred lithium-containing materials. Of the oxides, metal oxides are of particular note, more particularly transition metal oxides. Transition
10 metal oxides include, for example, chromium oxides, manganese oxides, iron oxides, cobalt oxides, nickel oxides, copper oxides and zinc oxides. Tin oxide or silicon oxide is preferred. Mixed metal oxides, for example ZnMn_2O_4 may be used.

Of particular importance are anodes which consist essentially of lithium intercalated into an oxide or an oxide-carbon matrix. The oxide-carbon matrix is
15 composite having a substantially homogeneous matrix of carbon in the oxide or an oxide in the carbon. Preferred oxides and conductive carbons are described previously. The oxide:carbon ratio (w/w) is preferably in a range of from 99:1 to 1:99, more preferably 90:10 to 10:90, most preferably 89:11 to 11:89. The oxide is preferably provided in the form of nanoparticles, forming a nanocomposite with the carbon. In this particular
20 application, the binder containing xanthan gum may further comprise a co-binder, for example, polyvinylidene fluoride, sodium carboxymethylcellulose, styrene-butadiene rubber or mixtures thereof. Preferably, the co-binder is present in the binder in an amount of less than about 75 wt% of the weight of the binder, more preferably less than about 50 wt%, even more preferably less than about 25 wt%. Most preferably, the binder
25 does not contain a co-binder and consists essentially of xanthan gum.

Also of importance are anodes comprising alloy composites incorporating at least one element (metal or metalloid) that can alloy with lithium (e.g. Zn, Cd, Pt, Al, Ga, In, Si, Ge, Sn, Pb, Sb, Bi), or a conductive carbon as discussed earlier or a second element (preferably a metal) that does not alloy with lithium (e.g. Co or Ti). The use of xanthan
30 gum helps accommodate large volume expansion (100-400%) associated with the alloying process that leads to bad battery performance. Sn, Si and Sn/Co/C alloy are of particular note.

Xanthan gum is a polysaccharide derived from the bacterial coat of *Xanthomonas campestris*. Xanthan gum is prepared by inoculating *Xanthomonas campestris* bacterium
35 with a sterile aqueous solution of carbohydrate (e.g. glucose, sucrose, lactose or mixtures

thereof), a source of nitrogen, dipotassium phosphate, and some trace elements. The medium is well-aerated and stirred, and the polymer is produced extracellularly into the medium. The final concentration of xanthan produced will vary greatly depending on the method of production, strain of bacteria, and random variation. After fermentation that can
5 vary in time from one to four days, the xanthan polymer is precipitated from the medium by the addition of isopropyl alcohol and dried and milled to give a powder that is readily soluble in water or brine to form a gum. The degree of carboxylation of xanthan gum can affect its performance as an electrode binder. Typically, a higher degree of carboxylation will confer better battery performance. The degree of carboxylation of the xanthan gum
10 may be readily controlled by carboxylation or de-carboxylation reactions to achieve the desired performance characteristics. Preferably, the degree of carboxylation is in a range of from about 0.5 to about 1.

Further features of the invention will be described or will become apparent in the course of the following detailed description.

15 Brief Description of the Drawings

In order that the invention may be more clearly understood, embodiments thereof will now be described in detail by way of example, with reference to the accompanying drawings, in which:

Fig. 1 depicts a graph showing cycling behavior of half batteries made of a lithium
20 reference and counter electrode and a meso carbon microbead (MCMB) anode. Anodes were prepared using five different binders: polyvinylidene fluoride (PVDF), sodium carboxymethylcellulose (NaCMC) from two different sources, lithium carboxymethylcellulose (LiCMC), Baytron™ (a compound of poly-3,4-ethylenedioxythiophene and polystyrenesulfonic acid) and xanthan gum. The electrodes
25 were cycled between 5 mV and 1.5 V versus Li/Li⁺ at C/12 (complete charge and discharge in 24 h).

Fig. 2 depicts a graph showing cycling behavior of half batteries made of a lithium reference and counter electrode and a meso carbon microbead (MCMB) anode. Anodes were prepared using five different binders: polyvinylidene fluoride (PVDF), sodium
30 carboxymethylcellulose (NaCMC), lithium carboxymethylcellulose (LiCMC), Baytron™ (a compound of poly-3,4-ethylenedioxythiophene and polystyrenesulfonic acid) and xanthan gum. The electrodes were cycled between 5 mV and 1.5 V versus Li/Li⁺ at different C-rates (C/12, C/9, C/6, C/3, C/2, C and 2C).

Fig. 3 depicts a graph showing a long term cycling behavior of half batteries made of a lithium reference and counter electrode and a meso carbon microbead (MCMB) anode. Anodes were prepared using xanthan gum. The electrodes were cycled between 5 mV and 1.5 V versus Li/Li⁺ at different C-rates.

5 Fig. 4 depicts a graph showing cycling behavior of half batteries made of a lithium reference and counter electrode and an anode of a composite material (nano-SnO₂/C 78%-22% in weight %). Anodes were prepared using three different binders: polyvinylidene fluoride (PVDF), sodium carboxymethylcellulose (NaCMC), and xanthan gum. The electrodes were cycled between 0.1 V and 1 V versus Li/Li⁺ at C/12 (complete charge and discharge in 24 h).

Fig. 5 depicts a graph showing cycling behavior of half batteries made of a lithium reference and counter electrode and a Sn₃Co₃Cu anode. Anodes were prepared using three different binders: polyvinylidene fluoride (PVDF), sodium carboxymethylcellulose (NaCMC), and xanthan gum. The electrodes were cycled
15 between 5 mV and 1.5 V versus Li/Li⁺ at C/12.

Fig. 6 depicts a graph showing cycling behavior of half batteries made of a lithium reference and counter electrode and an anode of a sintered mixed metal oxide (ZnMn₂O₄ spinel). Anodes were prepared using five different binders: lithium carboxymethylcellulose (LiCMC), sodium carboxymethylcellulose (NaCMC), Baytron™ (a compound of poly-3,4-ethylenedioxythiophene and polystyrenesulfonic acid), xanthan gum and polyvinylidene fluoride (PVDF). The electrodes were cycled between 10 mV and 3 V versus Li/Li⁺ at C/10 (complete charge and discharge in 20 h).

Fig. 7 depicts a graph showing long term cycling behavior of half batteries made of a lithium reference and counter electrode and a silicon anode. Anodes were prepared
25 using lithium carboxymethylcellulose (LiCMC), sodium carboxymethylcellulose (NaCMC), xanthan gum and polyvinylidene fluoride (PVDF). The electrodes were cycled between 5 mV and 2.0 V versus Li/Li⁺.

Description of Preferred Embodiments

Cyclic voltammetry and cell cycling were carried out on half cells using 2325-type
30 coin cells assembled in an argon-filled glove box. Cyclic voltammograms were recorded using a BioLogic™ VMP3 potentiostat/galvanostat. The potential of the working electrode was swept at 0.1 mV s⁻¹ from open-circuit potential down to 5 mV (or 10 mV) versus Li/Li⁺, then swept up to 1.5 V (or 2 V or 3 V) versus Li/Li⁺; afterwards cells were cycled between

1.5 V (or 2 V or 3 V) and 5 mV (or 10 mV) versus Li/Li⁺. Capacity measurements were performed by galvanostatic experiments carried out on a multichannel Arbin battery cyclers. The working electrode was first charged down to 5 mV (or 10 mV) versus Li/Li⁺ at different C-rates and then discharged up to 1.5 V (or 2 V or 3 V) versus Li/Li⁺. The mass of active material used in the calculation is the mass of the material used in the active electrode.

Working electrodes were prepared as follows. Active material (e.g. carbon graphite (MCMB), nano-SnO₂/C, Sn₃Co₃, ZnMn₂O₄ or Si) was mixed with 5 wt% of Super carbon (Timcal) and 5 or 10 wt% of binder. Electrode films were made by spreading the material onto a high purity copper foil current collector (cleaned using a 2.5% HCl solution in order to remove the copper oxide layer) using an automated doctor-blade and then dried overnight at 85°C in a convection oven. Individual disk electrodes (Ø = 12.5 mm) were punched out, dried at 80°C under vacuum overnight and then pressed under a pressure of 0.5 metric ton. A lithium metal disk (Ø = 16.5 mm) was used as a negative electrode (counter electrode and reference electrode). 70 µL of a solution of 1 M LiPF₆ in ethylene carbonate/dimethyl carbonate (1:1, v/v) was used as electrolyte and spread over a double layer of microporous propylene separators (Celgard™ 2500, 30 µm thick, Ø = 2.1 mm). The cells were assembled in an argon-filled dry glove box at room temperature.

Referring to Fig. 1, it is evident that for half batteries comprising meso carbon microbead (MCMB) anodes, the use of xanthan gum as the anode binder leads to significantly higher and more stable discharge capacity over 100 charging cycles than all of the use of any of the other binders, including PVDF. Referring to Fig. 2, it is evident that this result is consistent across a broad range of C-rates.

For half batteries comprising nano-SnO₂/C anodes, the use of xanthan gum as the anode binder leads to significantly higher and more stable discharge capacity over tens of charging cycles than with the use of PVDF, and the use of xanthan gum is comparable to the use of NaCMC (see Fig. 4).

For half batteries comprising Sn₃Co₃Cu anodes, the use of xanthan gum as the anode binder leads to significantly higher and more stable discharge capacity over tens of charging cycles than with the use of PVDF, albeit a somewhat lower discharge capacity than with the use of NaCMC (see Fig. 5).

For half batteries comprising ZnMn₂O₄ spinel anodes, the use of xanthan gum as the anode binder leads to higher and more stable discharge capacity over tens of

charging cycles than with the use of PVDF, although the use of xanthan gum led to somewhat lower discharge capacity than the use of LiCMC or NaCMC (see Fig. 6).

For half batteries comprising silicon anodes, the use of xanthan gum as the anode binder leads to higher discharge capacity retention at cycle #2 than PVDF (45% vs. 21%)
5 but lower discharge capacity retention than either LiCMC (87%) or NaCMC (95%) (see Fig. 7).

Results demonstrate that the use of xanthan gum as an anode binder provides higher and more stable discharge capacity than polyvinylidene fluoride over a broad range of anode materials, while in some cases being comparable to
10 carboxymethylcellulose.

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Other advantages that are inherent to the structure are obvious to one skilled in the art. The embodiments are described herein illustratively and are not meant to limit the scope of the invention as claimed. Variations of the foregoing embodiments will be evident to a person of ordinary skill and are intended by the inventor to be encompassed
5 by the following claims.

Claims:

1. Use of a binder consisting essentially of xanthan gum for binding an electrode active material in a lithium-ion or lithium-sulfur battery.
2. The use according to claim 1, wherein the electrode active material consists essentially of Li-S, a lithium alloy, lithium intercalated into a conductive carbon, lithium intercalated into an oxide, lithium intercalated into a nitride, lithium intercalated into a phosphide, or lithium intercalated into silicon.
3. The use according to claim 2, wherein the electrode active material consists essentially of lithium intercalated into an oxide.
4. The use according to claim 3, wherein the oxide is aluminum oxide, tin oxide, silicon oxide, cobalt oxide, iron oxide, titanium oxide, copper oxide or a mixture of two or more thereof.
5. The use according to claim 4, wherein the oxide is silicon oxide.
6. The use according to claim 3, wherein the oxide is a metal oxide.
7. The use according to claim 6, wherein the metal oxide is tin oxide.
8. The use according to claim 1, wherein the electrode active material consists essentially of lithium intercalated into a mixed metal oxide.
9. The use according to claim 1, wherein the electrode active material consists essentially of lithium intercalated into a homogeneous matrix of an oxide and a conductive carbon.
10. The use according to claim 9, wherein the conductive carbon comprises graphite.
11. The use according to any one of claims 1 to 10, wherein the electrode active material is an anode.
12. An electrode for a lithium-ion or lithium-sulfur battery comprising a lithium-containing material bound by a binder consisting essentially of xanthan gum.

13. The electrode according to claim 12, wherein the lithium-containing material consists essentially of Li-S, a lithium alloy, lithium intercalated into a conductive carbon, lithium intercalated into an oxide, lithium intercalated into a nitride, lithium intercalated into a phosphide, or lithium intercalated into silicon.
14. The electrode according to claim 12, wherein the lithium-containing material consists essentially of lithium intercalated into an oxide.
15. The electrode according to claim 12, wherein the lithium-containing material consists essentially of lithium intercalated into a metal oxide.
16. The electrode according to claim 12, wherein the lithium-containing material consists essentially of lithium intercalated into an aluminum oxide, tin oxide, silicon oxide, cobalt oxide, iron oxide, titanium oxide, copper oxide or mixtures thereof.
17. The electrode according to claim 12, wherein the lithium-containing material consists essentially of lithium intercalated into tin oxide or silicon oxide.
18. The electrode according to any one of claims 12 to 17, wherein the electrode is an anode.

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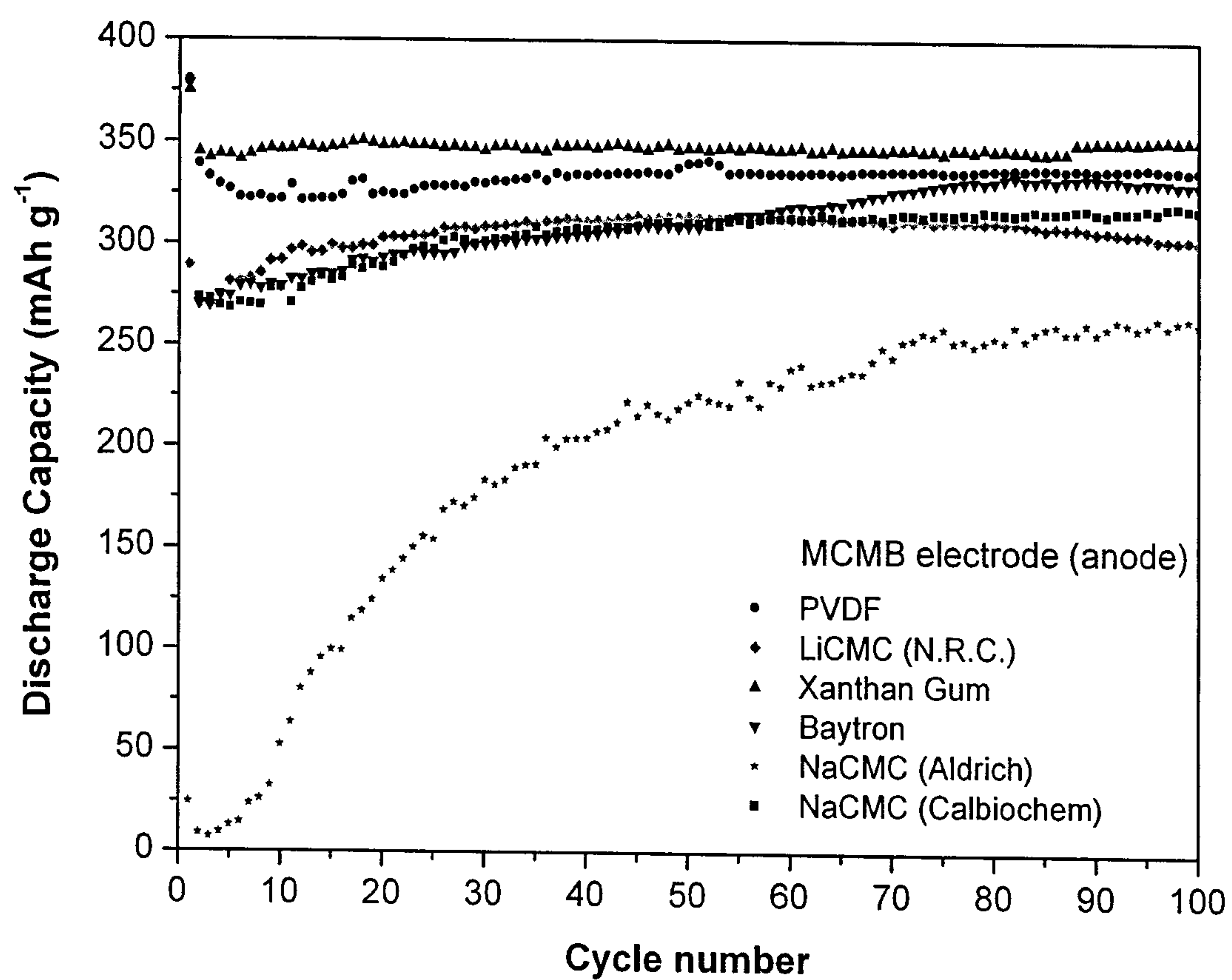


Fig. 1

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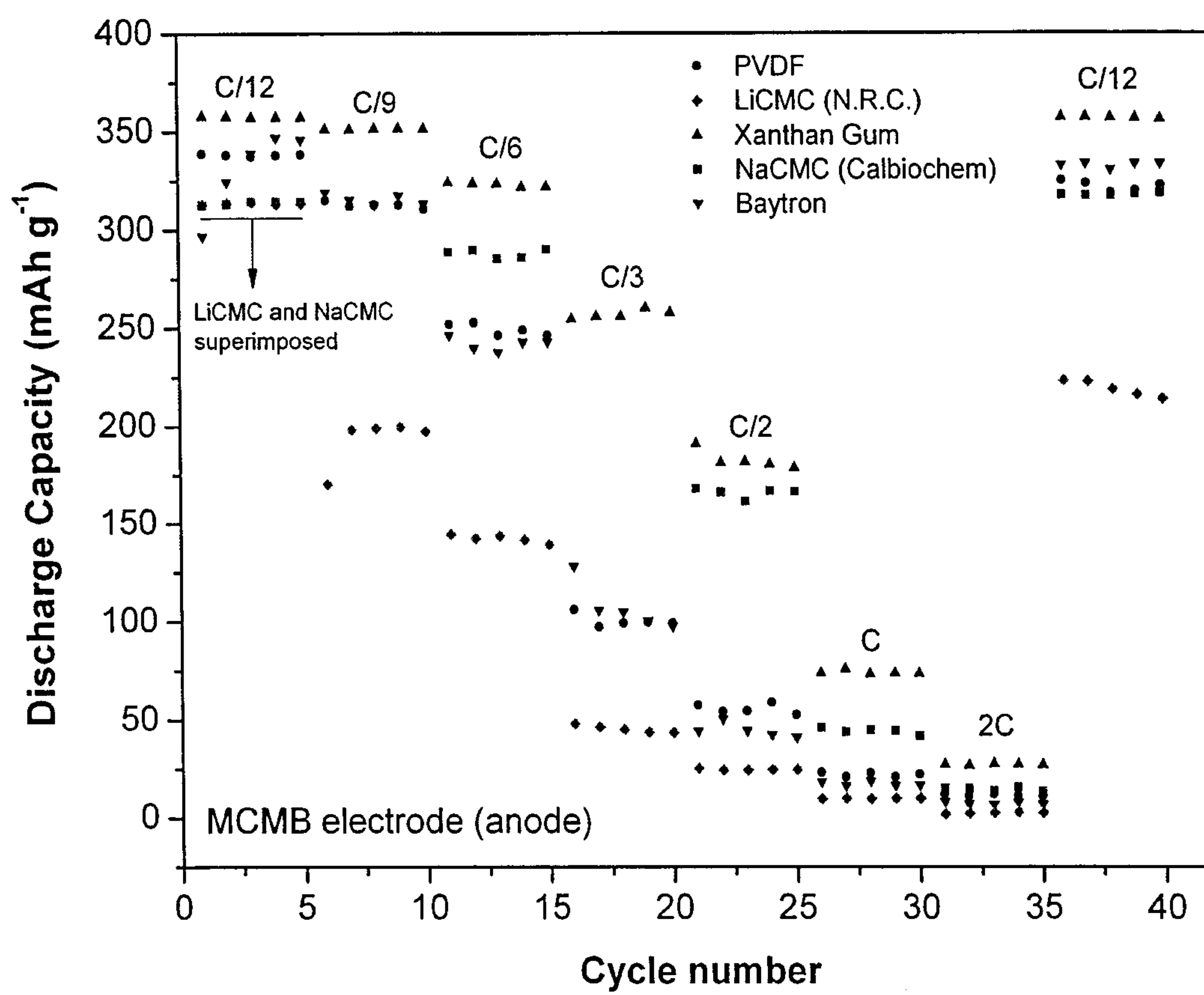


Fig. 2

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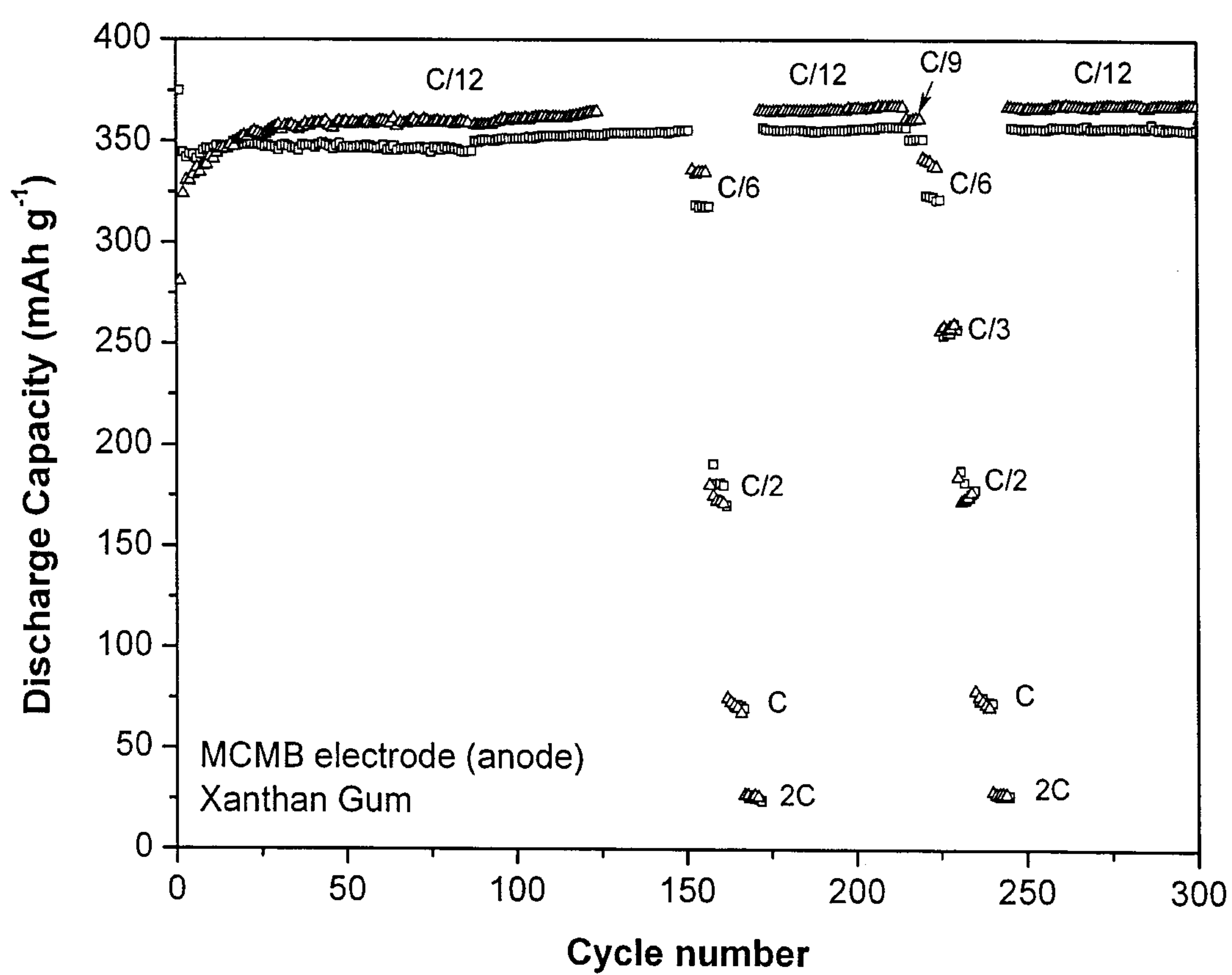


Fig. 3

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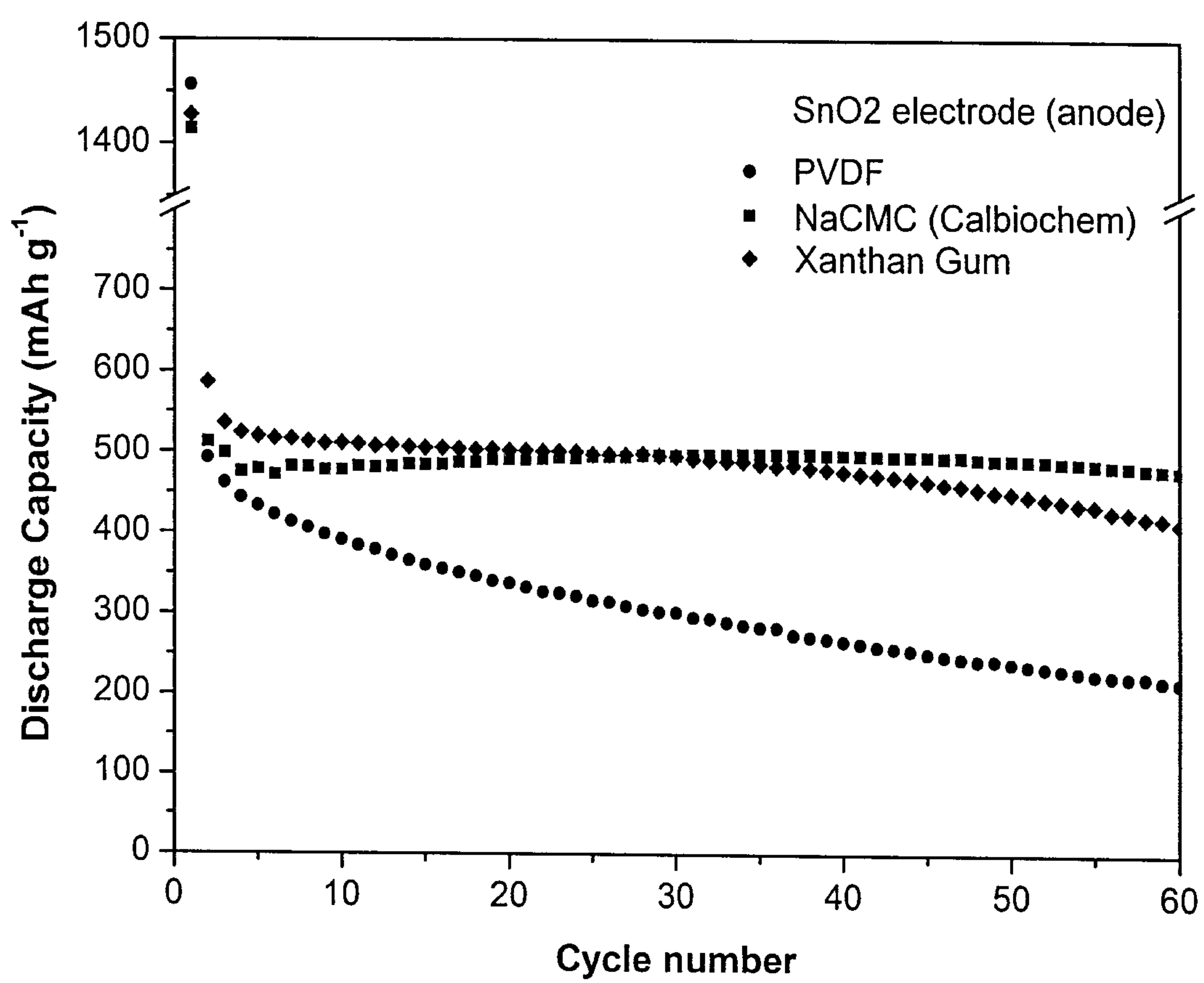


Fig. 4

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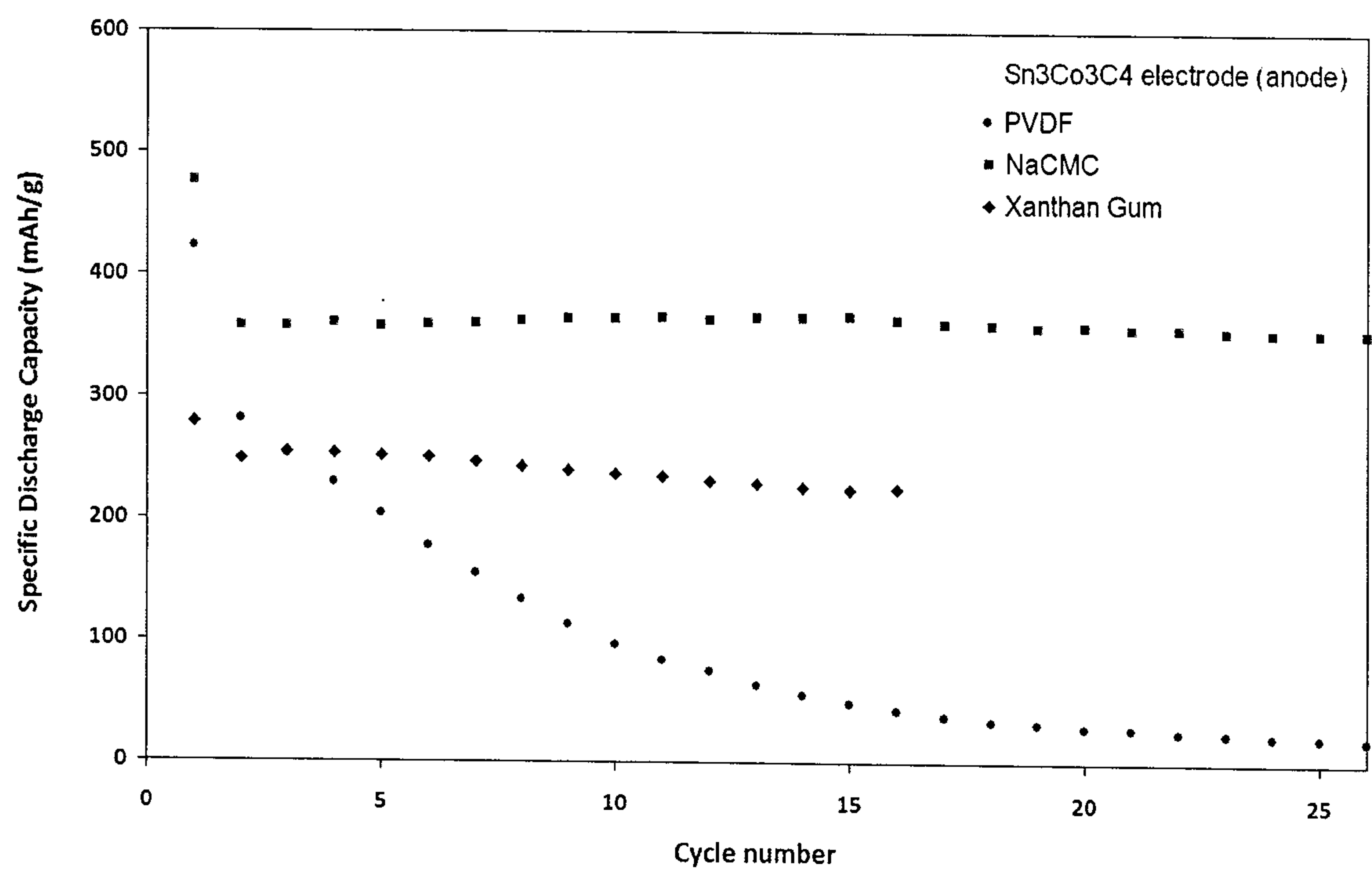


Fig. 5

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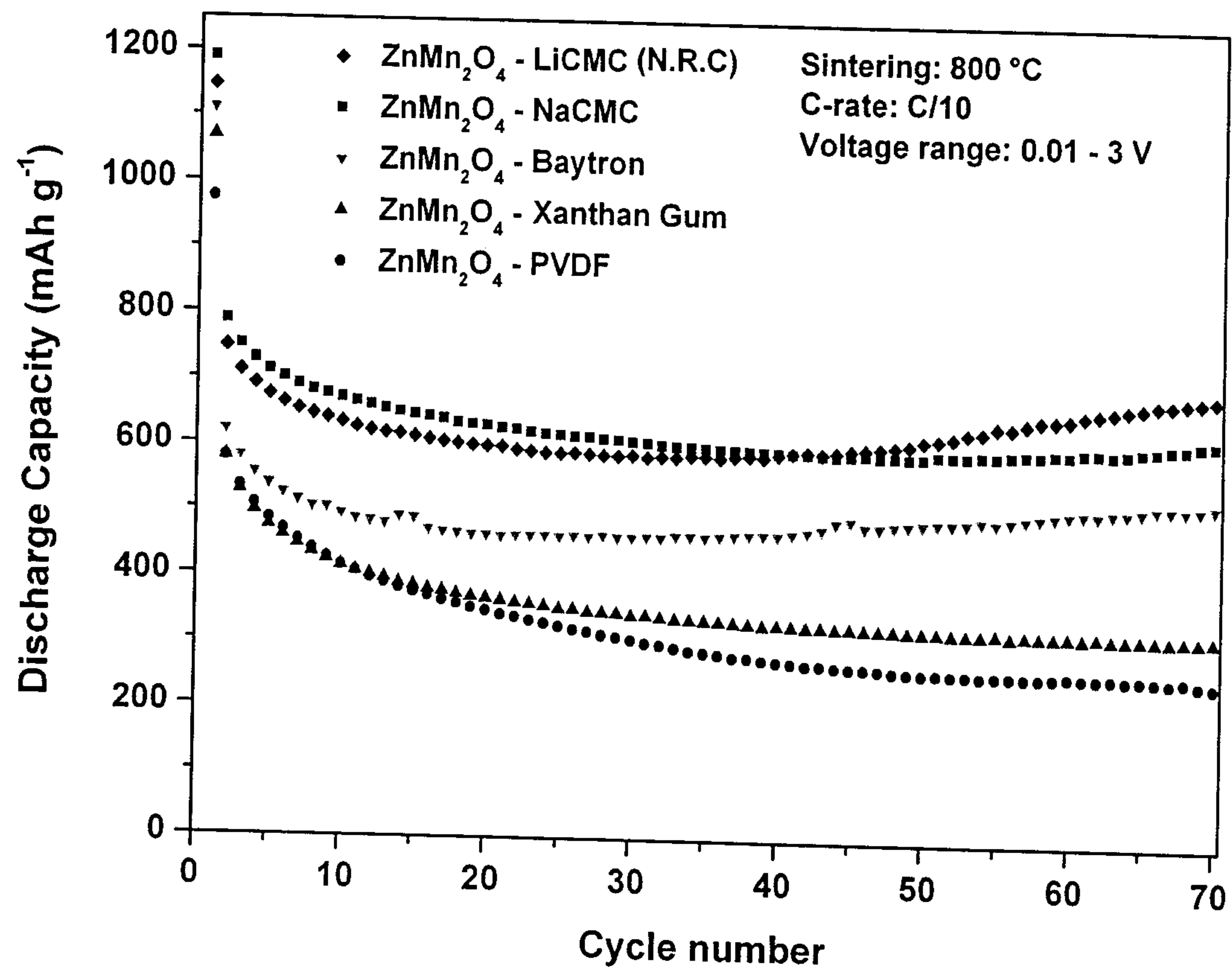


Fig. 6

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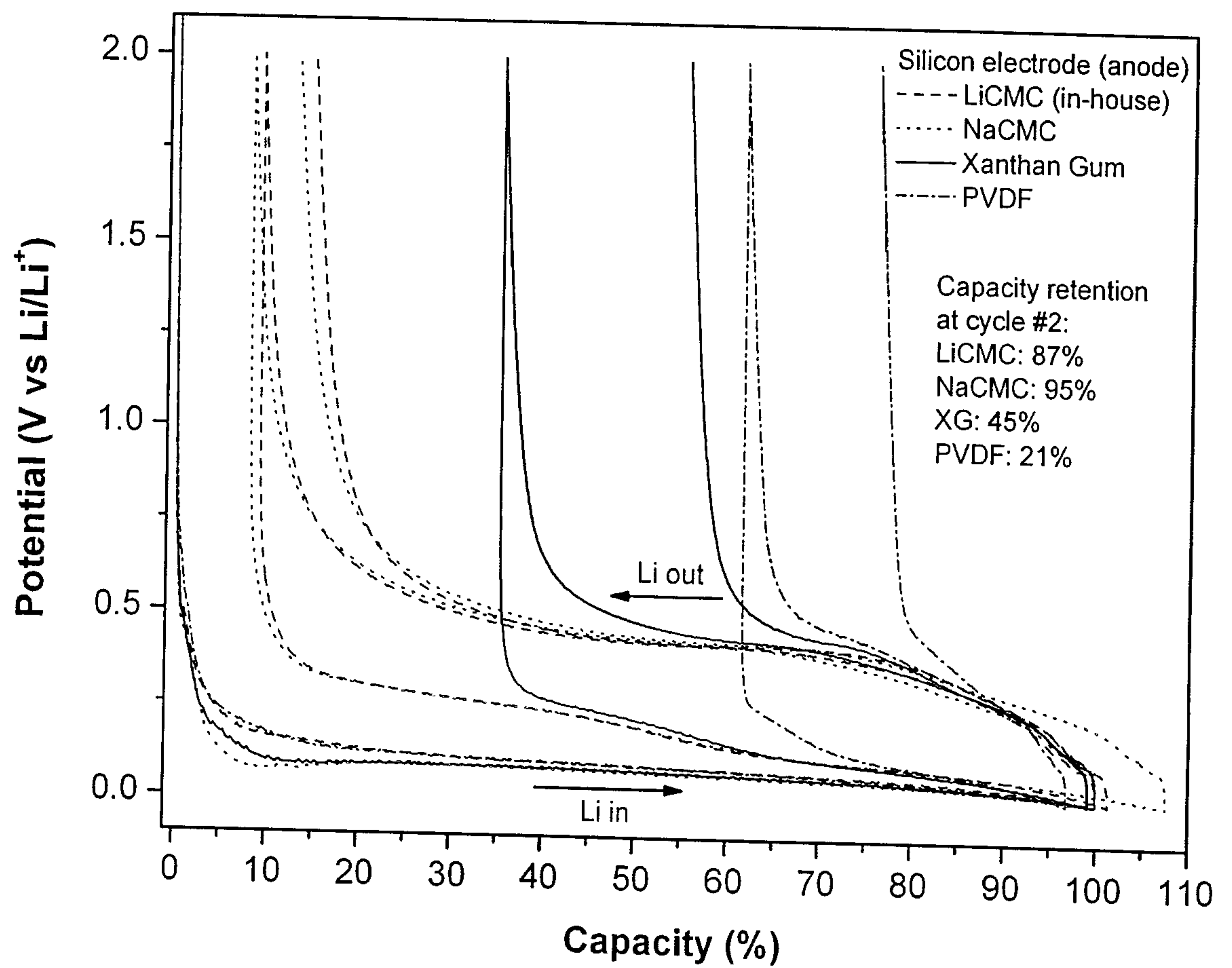


Fig. 7

