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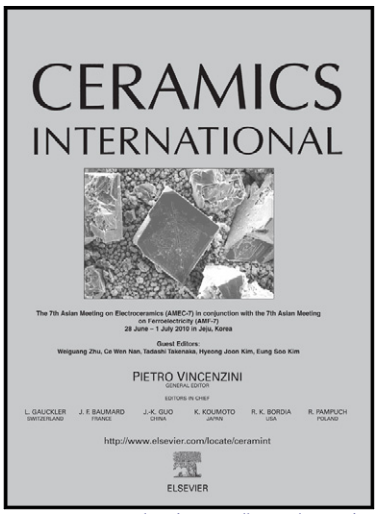

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Gel Casting of Sialon Ceramics based on Water Soluble Epoxy Resin

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Abstract

A recently developed gel casting system based on water soluble epoxy resin was successfully applied to sialon ceramics. The solids loading could reach 48.9% without any pH adjustment, and all slurries exhibited shear-thinning behaviour. Flexural strengths of sialon ceramic obtained from 44.5 vol.% slurry were 295.2 MPa at room temperature and 281.4 MPa at 700 °C, respectively; the microhardness was 16.5 GPa. The net-shape manufacturing of sialon ceramics could benefit from this investigation.

Key words: Gel-casting; Epoxy resin; Sialon powders; Ceramics

1. Introduction

Sialons were discovered in the early 1970s [1-4]，and have been developed for structural engineering applications due to their distinct advantages[5,6], such as very high strength, good thermal shock resistance and exceptional resistance to wetting or corrosion, et al. (α+β) sialon composites are very promising from the stand-point of tailoring for specific applications because of the flexibility of their phase compositions and microstructure [6].

The accurate microstructural control of ceramics is essential when specific requirements have to be achieved [7,8]. The fabrication techniques play an important role in the control of microstructure. Among the techniques, colloidal processes [9,10] are the frequently-used techniques, which allow better quality control, leading to homogeneous architectures. Gel casting is such a kind of colloidal process invented by Janney and Omatete in 1990s [11,12], and has several merits compared with

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traditional colloidal processes, such as shorter production cycle and higher green strength, especially the much more homogeneous microstructures of the products [13, 14]. Generally, the gel casting process includes dispersion of a ceramic powder in a monomer solution to form a slurry which is subsequently gelled in a mould (to form a homogeneous wet green body) [13]. After drying, binder removal and sintering are carried out as in other ceramic processes. However, the neurotoxicity of the monomer, such as acrylamide in gel casting, limits its application due to the environment and health concern. A lot of efforts hence have been paid to develop alternative gelling agents with low toxicity or nontoxicity [15, 16]. Recently, a ring-open polymerization mechanism was brought into the gel casting process [16, 17]. The polymerization mechanism is based on the poly-addition reaction between a water-soluble epoxy resin and an amine hardener, and is not affected by oxygen. This new technique were successfully applied for different materials and applications, such as PZT for ultrasound devices [18-20], zirconia for mirospanner [21], alumina for microgear[22, 23], BST for pillar arrays[24], Y-TZP for helical springs [25], SiO_2[26] et al, making it very versatile in the colloidal process of ceramics.

However, few investigations have been done about sialon using the above technique. Therefore, the present study reports for the first time on gelcasting of sialon based on water soluble epoxy resin, and the goal is to develop an epoxy resin gel casting system which is suitable for near net-shape manufacture of sialon ceramics.

2. Experimental

Sialon 050 powders (International Syalons Newcastle, England, UK) were used as the

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raw material. Ethylene glycol diglycidyl ether (EGDE, Nasage, Japan) and bis(3-aminopropyl) amine (Sigma-Aldrich, Germany) were used as epoxy resin and hardener, respectively. The resin content was fixed at 25%, based on the total weight of water and resin for all the slurries. The ratio of resin and the hardener content was fixed at 1:0.23. Ammonium polyacrylate (NH_4PAA) solution (Allied Colloids, Bradford, UK) was used as the dispersant. Distilled water (17 MΩ·cm) was used as the vehicle. The aqueous solution containing EGDE was prepared by dissolving resin in distilled water with dispersant prior to the preparation of slurries. Slurries with various solids loadings were prepared by adding sialon powders into the premixed solution, and were ball milled for 24 hours with alumina milling media. Then, hardener was added into the slurries. The resulting slurries were cast into suitable silicone moulds of the desired shapes and degassed in a vacuum system for 1 minute to aid moulding. After drying slowly in a sealable box at room temperatures for 4-5 days, demoulding was carefully done and the green bodies were transferred into a 40 ºC oven for further drying for 24 hours. Sintering was carried out in nitrogen. A slow ramp of 1 ºC/min to 600 ºC was conducted to burn out the organics followed by 5 ºC/min ramp to 1730ºC, holding for 2 hours. All the green and sintered samples were ground and polished for observation and measurement. The rheological behaviour was characterised with an AR 500 rheometer (TA instruments, USA) with a cone and plane system (Truncation: 59 µm; Core: 4 cm, 2°). The flow behaviour of the slurries after adding hardener was measured at 20°C under the continuous shear mode within the shear rate range of 0.1~600 s^{-1}. The drying shrinkage rates were measured based on

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the mould size and sample size after drying. The sintering shrinkage rates of the samples were obtained based on the measurements of the sample dimensions before and after sintering. An optical digital camera (Olympus SP350, Olympus Co (UK) Ltd) was used to examine the shape of the green samples. Scanning Electron Microscopy (SEM) (JSM 7000, Jeol, Tokyo, Japan) was employed to observe the microstructure of the samples. All the densities in this study were measured by the Archimedes method. The flexural strength of sintered samples (at least 6) were conducted in four-point flexure with an universal mechanical testing machine (Instron 4505, England, UK) equipped with a 10 kN load cell at 20 and 700 °C. The Vickers microhardness tests were performed on the sintered samples using a Mitutoyo MVK-H1 microhardness tester (1 kg load for ten seconds) (Mitutoyo Ltd, UK).

3 Results and Discussion

The information of the sialon powders used in this study is exhibited in Figure 1. Figure 1(a) shows the image of the powders at low magnification and large agglomerates can be observed. This is because the powders were obtained by the spray drying method. However, the agglomerates belong to soft agglomerates, which can be broken easily through ball milling process. As shown in Figure 1 (b) at higher magnification, the primary particle size could be analyzed, and it is around 300nm. Moreover, the surface area of the powder (BET) is 11.96 cm^2/g.

The rheological properties of slurries are shown in Figure 2. Figure 2(a) shows the flow behaviour of slurry with 44.5 vol.% solids loading. When the shear rate was increased, the viscosity decreased and the shear stress increased, which indicates a

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typical shear-thinning behaviour. This is in agreement with Mao's work [16].

The SEM micrographs of the fracture surface of the green body (obtained from 44.5vol.% slurry) is shown in Figure 3(a). Pores generated during the fabrication process could be observed, but most of them can be eliminated during sintering. A typical sintered long bar sample for mechanical test is shown in Figure 3(b). The microstructure of the sintered long bar is shown in Figure 3(c), and the sialon grains showed relatively homogeneous needle morphology.

Some properties of the green or sintered samples obtained from the above slurry (44.5 vol.%, 25wt.% resin) are displayed in Table 1. The using of 25 wt.% resin content was based on our previous study[21], and the purpose is to obtain strong green bodies suitable for the following mechanically machining. The density of the green body was 2.04 g/cm^3, while that of the sintered sample reached 3.14 g/cm^3, nearly 97% of theoretical density. The microhardness of the sintered ceramic was 16.5 GPa. The flexural strengths for the ceramics were 295.2 MPa at 20 °C and 281.4 MPa at 700 °C, respectively.

Moreover, slurries with higher solids loading had been successfully prepared using the same procedure as the 44.5 vol.% slurry. Figure 2(b) shows the effect of the solids loading on the rheological properties of these slurries. It can be observed that all the slurries showed shear-thinning behaviour. Viscosities increased with the increase of the solids loading. Especially when solids loading increased from 46.8 vol.% to 48.9 vol.%, the viscosity increased markedly, for example, the viscosity increased from 2.55 to 7.64 Pas at the shear rate of 100 s^{-1}. More details can be found in Figure 2(c).

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This tendency that higher solids loading leads to higher viscosity is in agreement with many published work [8,27]. In order to characterize the effect of solids loading on the shape control, drying shrinkage rates for green sample and sinter shrinkage rates for sintered sample in two directions, as indicated in Figure 3(b), were measured and presented in Table 2. The shrinkage rates either in direction D1 or D2 decreased with increase of the solids loading, irrespective of green or sintered samples. Increasing the solids loading is helpful to realize near-net-shape manufacture. Among these samples, 48.9 vol.% solids loading led to the smallest shrinkage rates (4.27%, 3.65%, 22.30% and 21.01%, respectively) which is the best for the net-shape manufacture at current stage.

4. Conclusions

In summary, sialon ceramics could be successfully prepared by using this water-soluble epoxy resin gelcasting system. For all the slurries, when the shear rate was increased, the viscosity decreased and the shear stress increased, indicating a typical shear-thinning behaviour. The density of sialon ceramic obtained from 44.5 vol.% slurry reached 3.14 g/cm^3, nearly 97% of theoretical density, and the microhardness was 16.5 GPa; the flexural strengths reached 295.2 MPa at room temperature and 281.4 MPa at 700 °C, respectively. Moreover, higher solids loading led to the smaller shrinkage rate, which is better for the net-shape manufacture.

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References

[1] V.A. Izhevskiy, L.A. Genova, J.C. Bressiani, F. Aldinger, Progress in SiAION ceramics, J. Eur. Ceram. Soc., 20(13) (2000) 2275-2295.

[2] K.H. Jack, W.I. Wilson, Ceramics based on Si-Al-O-N and related systems, Nature-Physical Science, 238(80) (1972) 28-&.

[3] Y. Oyama, O. Kamigait, Solid solubility of some oxides in Si_3N_4, Jpn. J. Appl. Phys., 10(11) (1971) 1637-&.

[4]I. Wilson, K.H. Jack, New ceramics materials based on Al-Si-N-O related systems, Am. Ceram. Soc. Bull., 51(4) (1972) 430-&.

[5] G.Z. Cao, R. Metselaar, Alpha-Sialon ceramics- a review, Chem. Mater., 3(2) (1991) 242-252.

[6] H. Mandal, New developments in alpha-sialon ceramics, J. Eur. Ceram. Soc., 19(13-14) (1999) 2349-2357.

[7] C. Gomez-Yanez, H. Balmori-Ramirez, F. Martinez, Colloidal processing of $BaTiO_3$ using ammonium polyacrylate as dispersant, Ceram. Int., 26 (6) (2000) 609-616.

[8] G. Liu, D. Zhang, T.W. Button, Preparation of concentrated barium titanate suspensions incorporating nano-sized powders, J. Eur. Ceram. Soc. 30(2) (2010) 171-176.

[9] J.A. Lewis, Colloidal processing of ceramics, J. Am. Ceram. Soc., 83 (10) (2000) 2341-2359.

[10] T.S. Suzuki, Y. Sakka, K. Nakano, K. Hiraga, Effect of ultrasonication on the microstructure and tensile elongation of zirconia-dispersed alumina ceramics prepared by colloidal processing, J. Am. Ceram. Soc., 84(9) (2001) 2132-2134.

[11] O.O. Omatete, M.A. Janney, R.A. Strehlow,Gelcasting-a new ceramic forming process, Am. Ceram. Soc. Bull., 70 (1991) 1641-&.

[12] A.C. Young, O.O. Omatete, M.A. Janney, P.A. Menchhofer, J. Am. Ceram. Soc., 74(10) (1991) 612-618.

[13] R. Gilissen, J.P. Erauw, A. Smolders, E. Vanswijgenhoven, J. Luyten, Gelcasting, a near net shape technique, Mater. Design, 21(4) (2000) 251-257.

[14] S. Ananthakumar, K. Prabhakaran, U.S. Hareesh, P. Manohar, K.G.K. Warrier, Gel casting process for Al_2O_3-SiC nanocomposites and its creep characteristics, Mater. Chem. and Phys., 85(1) (2004) 151-157.

[15] K. Cai, Y. Huang, J.L. Yang, Alumina gelcasting by using HEMA system, J. Eur. Ceram. Soc., 25(7) (2005) 1089-1093.

[16] X. Mao, S. Shimai, M. Dong, S. Wang, Effects of coarse particles on the gelcasting of ceramic foams, J. Am. Ceram. Soc., 91(10) (2008) 1354-1356.

[17] M. Dong, X. Mao, Z. Zhang, Q. Liu, Gelcasting of SiC using epoxy resin as gel former, Ceram. Int., 35(4) (2009) 1363-1366.

[18] S.M. Olhero, L. Garcia-Gancedo, T.W. Button, F.J. Alves, J.M.F. Ferreira, Innovative fabrication of PZT pillar arrays by a colloidal approach. J. Eur. Ceram. Soc., 32(5) (2012) 1067-1075.

[19] R. Xie, C. Liu, Y. Zhao, P Jin, K.C. Zhou, D. Zhang. Gelation behavior and mechanical

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properties of gelcast lead zirconatetitanate ceramics. J. Eur. Ceram. Soc. 35(7) (2015) 2051–2056.
[20] R. Xie, Y. Zhao, K.C. Zhou, D. Zhang, Y Wang, H.L.W. Chan, Fabrication of fine-scale 1-3 piezoelectric arrays by aqueous gelcasting, J. Am. Ceram. Soc., 97(8) (2014) 2590-2595.
[21] G. Liu, M.M. Attallah, Y. Jiang, T.W. Button, Rheological characterization and shape control in gel-casting of nano-sized zirconia powders. Ceram. Int., 40 (2014) 14405-14412.
[22] S.Vijayan, R. Narasimman, K. Prabhakaran, Gelcasting of alumina using the hexamethylenediamine-paraformaldehyde monomer system. Ceram. Int., 40 (2014) 3185-3191.
[23] R. Xie, D. Zhang, X.Y. Zhang, K.C. Zhou, T.W. Button. Gelcasting of alumina ceramics with improved green strength. Ceram. Int., 38(2012) 6923–6923.
[24] S.M. Olhero, A. Kaushal, P. Antunes, J.M.F. Ferreira, Microfabrication of high aspect ratio BST pillar arrays by epoxy gel casting from aqueous suspensions with added water soluble epoxy resin. Mater. Res. Bull., 60 (2014) 830-837.
[25] M.M. Zhuang, M.C. Wang, Y.N. Zhao, B. Zheng, A. Guo, J.C. Liu. Fabrication and high-temperature properties of Y-TZP ceramic helical springs by a gel-casting process. Ceram. Int., 41(2015) 5421-5428.
[26] W. Wan, Y.B.Feng, J. Yang, Shuang Xu, T. Qiu. Preparation of mesoporous silica ceramics with relatively high strength from industrial wastes by low-toxic aqueous gel-casting. J. Eur. Ceram. Soc., 35(7) (2015) 2163-2170.
[27] E. Adolfsson, Gelcasting of zirconia using agarose, J. Am. Ceram. Soc., 89(6) (2006) 1897-1902.

Figure Captions

Figure 1 Powder information: (a) secondary electron SEM images of sialon powder at low magnification, (b) at high magnification.

Figure 2 (a) Typical flow behaviour of the 44.5 vol.% slurry with 1wt.% dispersant and 25 wt.% resin content; (b) flow behaviour of slurries with different solids loadings with 25 wt.% resin content;(c) details of the viscosities shown in Figure 2(b).

Figure 3 (a) Secondary electron SEM micrographs of the fracture surface of the green bodies obtained from 44.5 vol.%; (b) optical image of typical sintered long bar; (c) microstructure of the sintered sialon long bar.

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Table 1 Some properties of the green or sintered sample obtained from 44.5 vol.% slurry

Solids loading (vol.%)	44.5
Green density (g/cm^3)	2.04
Sintered density (g/cm^3)	3.14
Microhardness (GPa)	16.5±0.8
Flexural strength (at 20 °C) (MPa)	295.2±38.1
Flexural strength (at 700 °C) (MPa)	281.4±33.1

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Table 2 Drying shrinkage rate and sinter shrinkage rate in two directions

	Direction	44.5 vol.% (%)	46.8 vol.% (%)	48.9 vol.% (%)
Drying	D1	5.83	4.41	4.27
	D2	5.61	4.77	3.65
Sinter	D1	24.07	22.42	22.30
	D2	22.88	22.76	21.01

The direction of D1 and D2 are shown in Figure 3(b).

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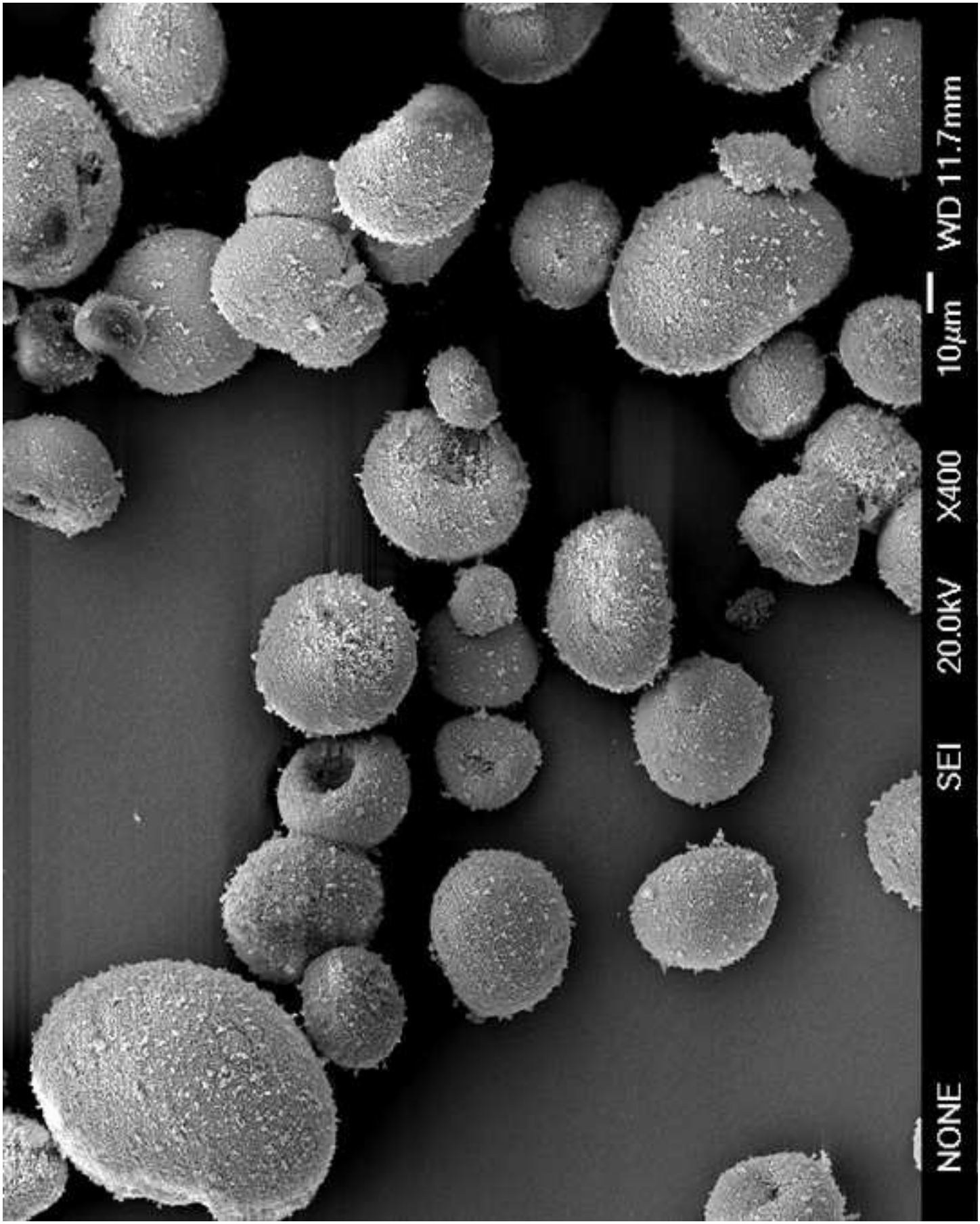

Figure 1(a)

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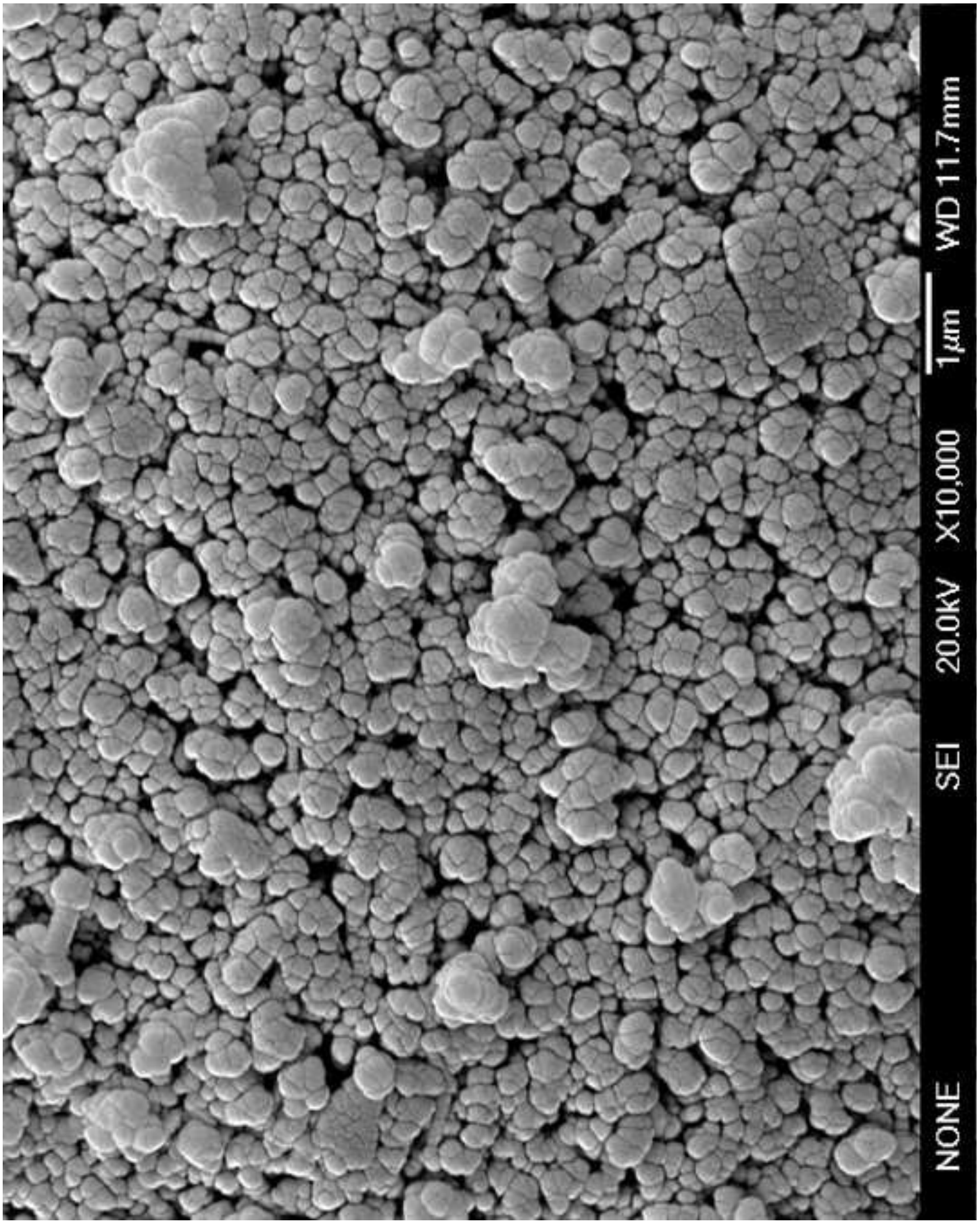

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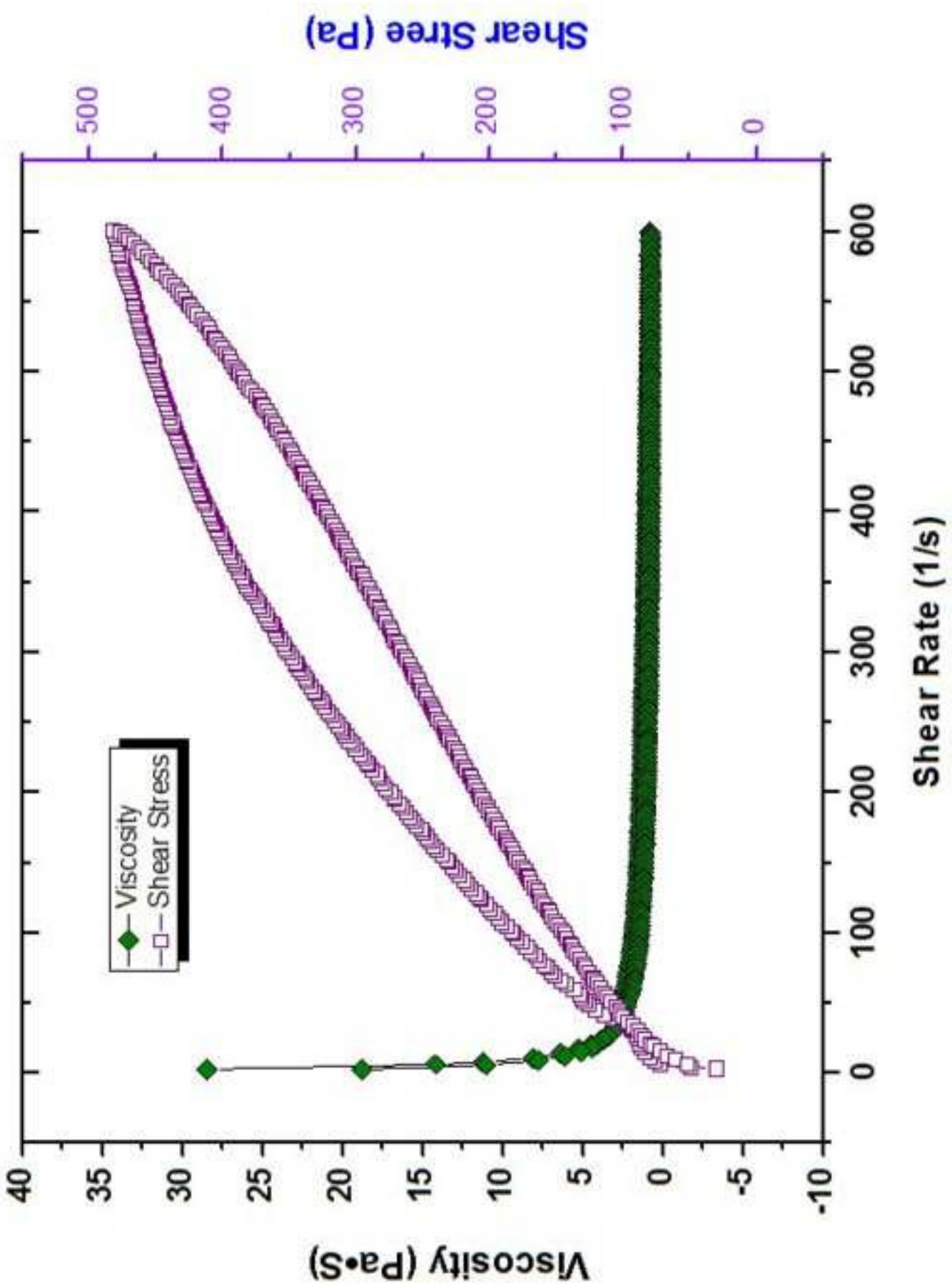
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Shear Stree (Pa)
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300
200
100
0
Shear Rate (1/s)
0
100
200
300
400
500
600
Viscosity (Pa•S)
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30
25
20
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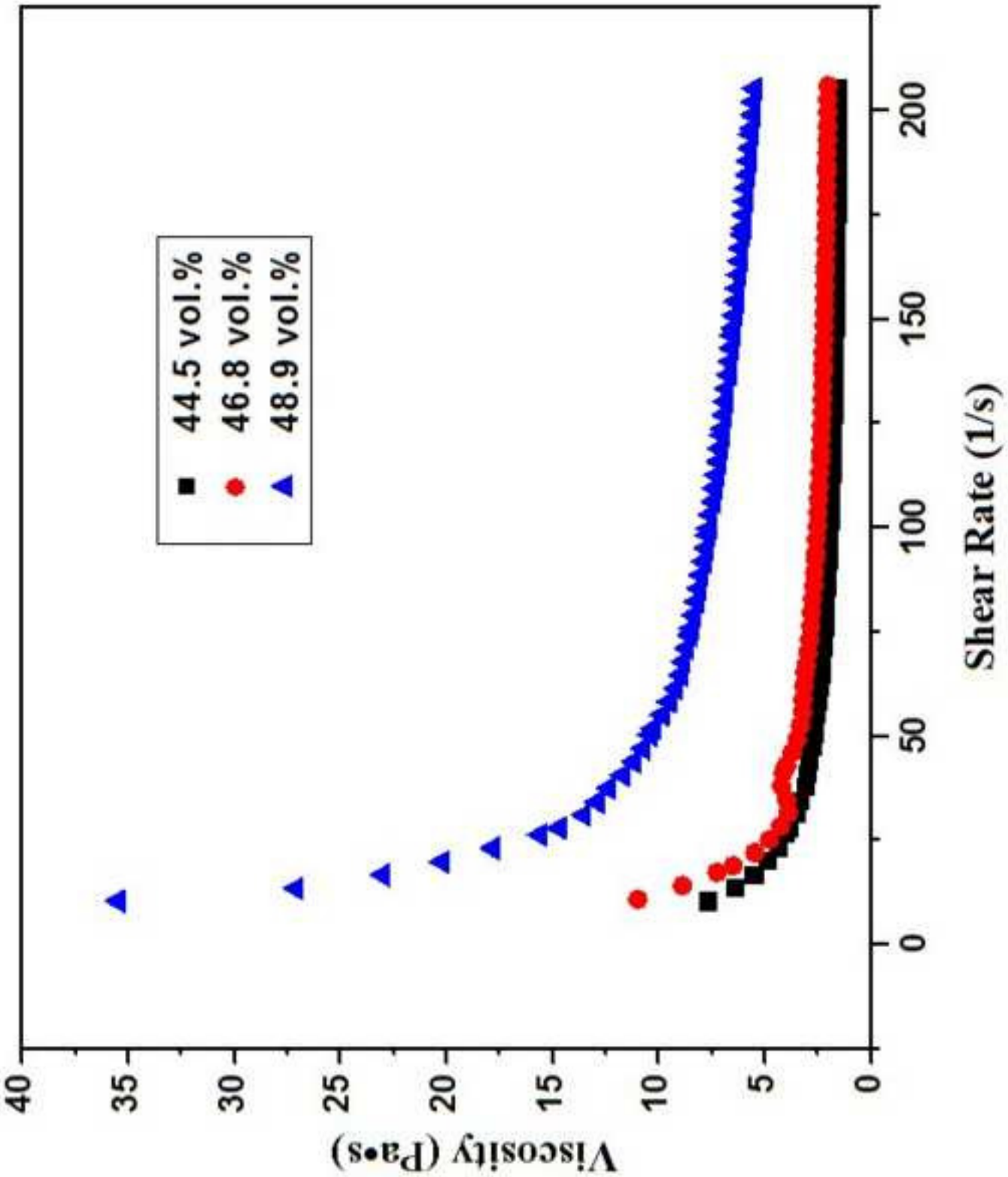
10
5
0
-5
-10

Figure 2(a)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46.8 vol.%
48.9 vol.%
Viscosity (Pa•s)
Shear Rate (1/s)
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35
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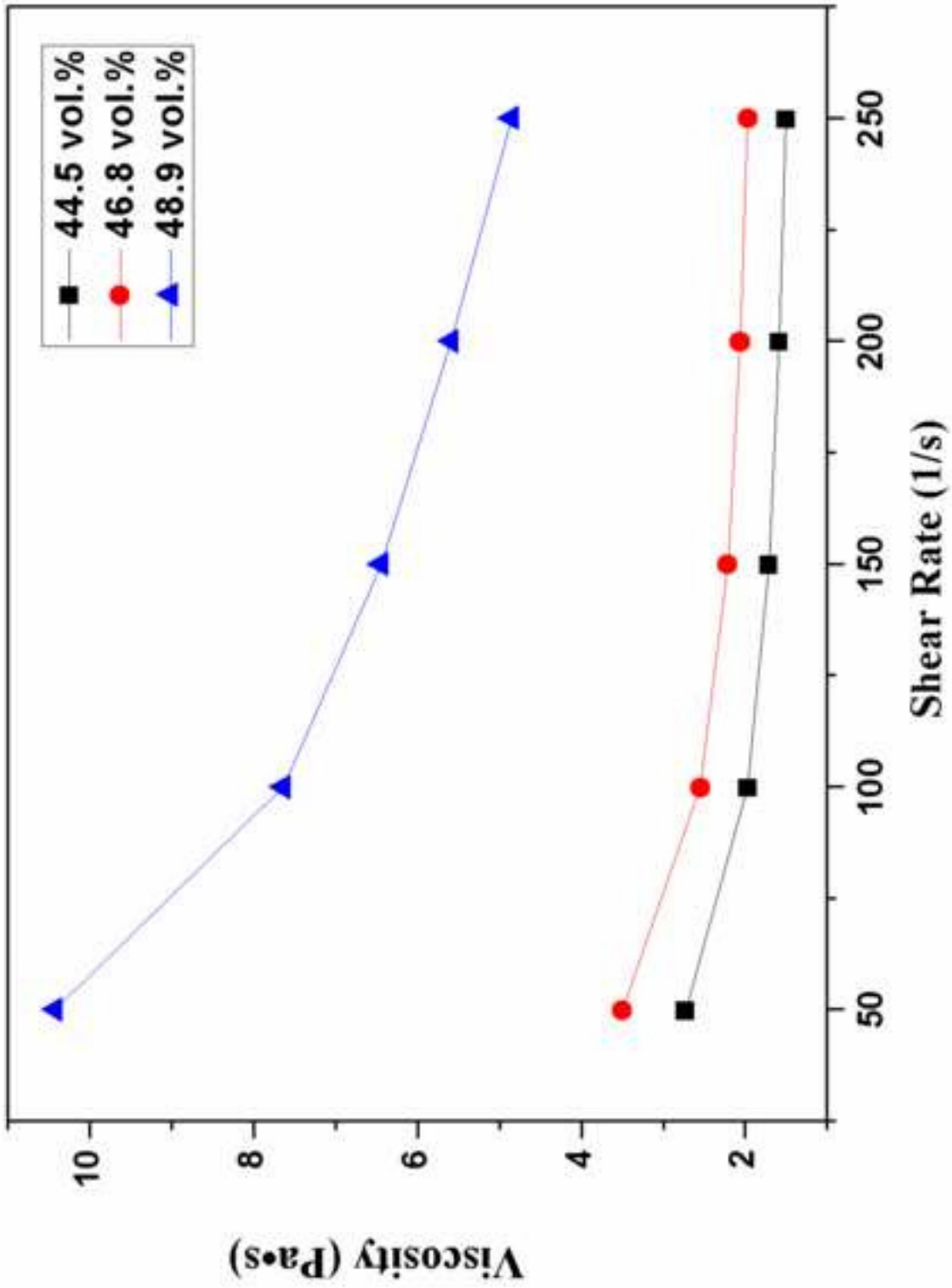
100
150
200

Figure 2(b)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46.8 vol.%
48.9 vol.%
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Shear Rate (1/s)

Figure 2(c)

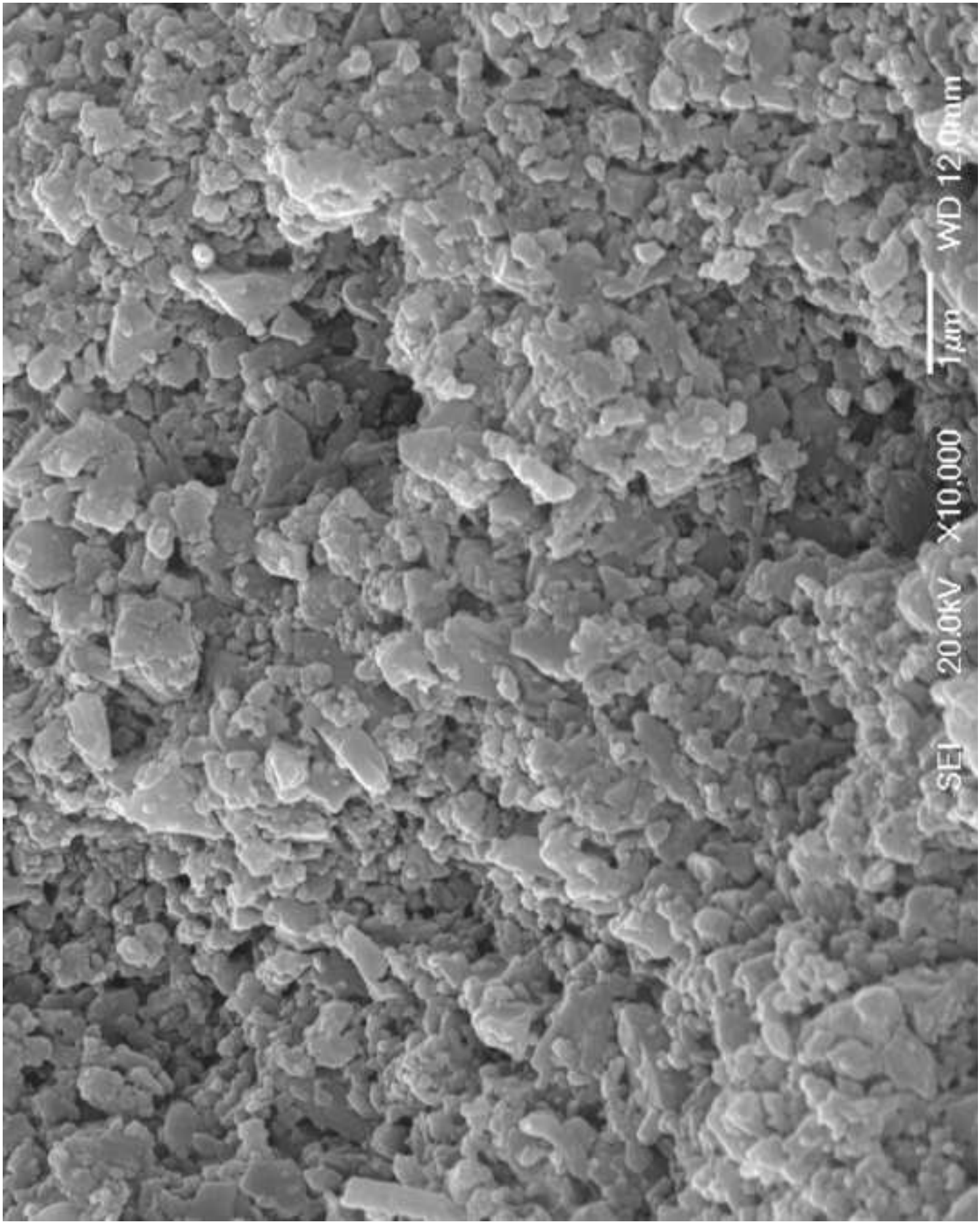

Figure 3(a)

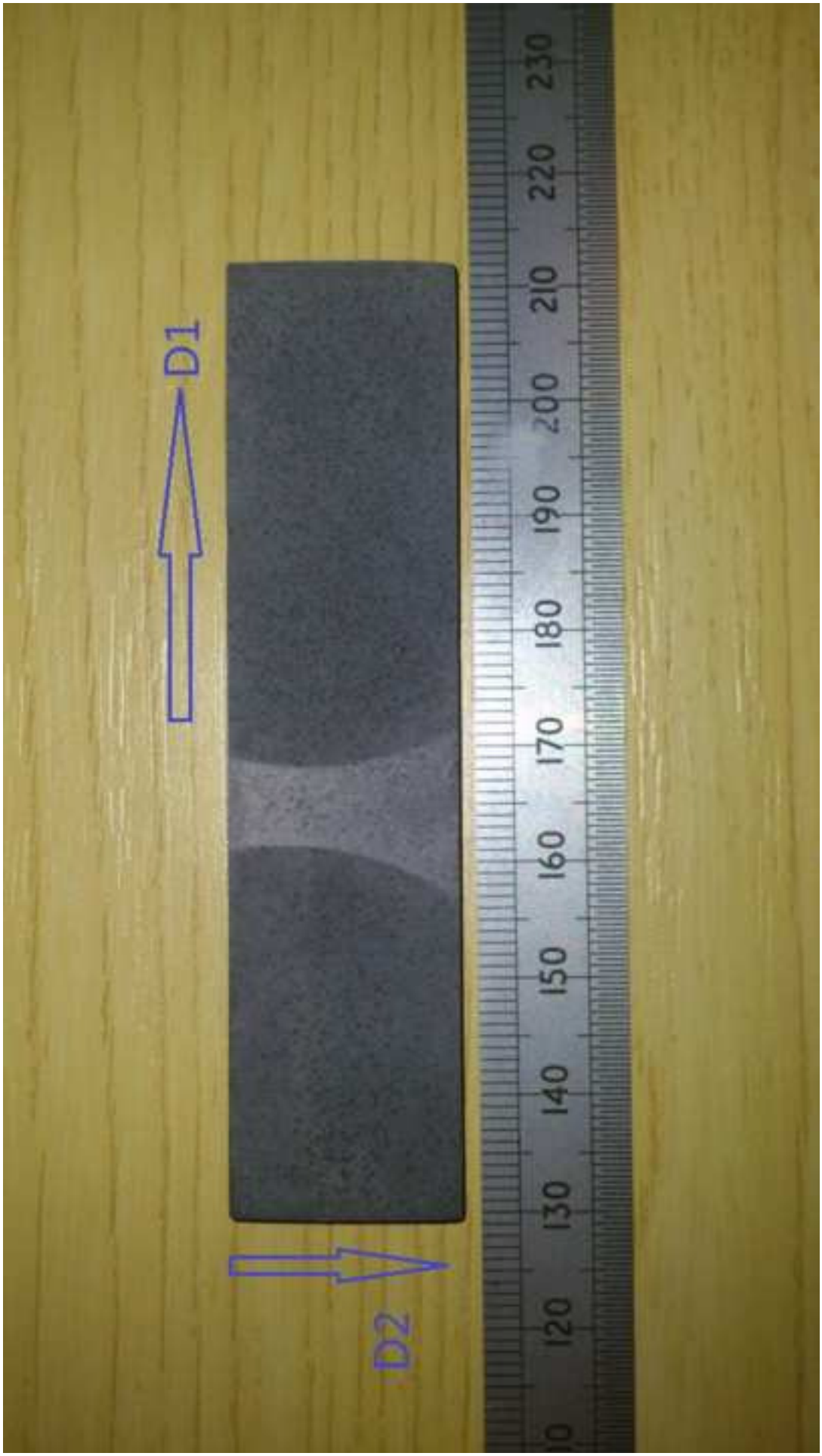

Figure 3(b)

Figure 3(c)