**Supporting information**

**S1. CAS extraction and purification**

Waste milk was pasteurized at 85 °C for 15 min. After treatment, it was cooled at 4 °C for 12 h, and the surface cream formed was removed. For casein precipitation, 100 mL of waste milk was heated at 40 °C, mixed with 300 mL of distilled water, and stirred at 150 rpm. Later, 4 mL of acetic acid 1 M was dropped and stirred for 10 min to promote the CAS precipitation. The final product was washed with 40 mL ethanol, centrifuged at 5000 rpm for 7 minutes, and dried at room temperature for 5 days.

**S2. FTIR characterization of CAS obtained from waste milk**

It can be seen a characteristic peak located at 3277 cm-1 associated with the OH/NH vibration and signals of methyl/methylene C-H stretching located at 2924 cm-1 and 2858 cm-1, respectively. Also, a signal at 1745 cm-1 related to a C=O stretching of carboxylic acid and the peaks of primary and secondary amide bonds of characteristic α-casein located at 1635, 1516, and 1395 cm-1, respectively (Picchio et al., 2018). According to the reported studies (Eigel et al., 1984; Xie and Hsieh, 2003), CAS is conformed with polar groups such as -COOH, -NH2, and -OH; in a relation, *i.e.*, 26 %, 15 %, and 15 %, respectively. In this order, the chemical composition of CAS obtained from cow milk is attributed to the presence of *ca*. 21 different amino acids, which mainly include leucine, lysine, valine, and phenylalanine, involved with the protein metabolism and proteins phosphorylation (Rafiq et al., 2016), with a molecular weight *ca.* 19,000 to 25,000 kDa.



**Figure S2.** FTIR spectra of CAS obtained from waste milk. Casein hydrolysate (Sigma Aldrich CAS No. 91079-40-2) was used as a reference, and obtained CAS from waste milk was purified with ethanol and ethanol/ether for comparison.

**S3. Mechanical performance values (Tensile strength and Breaking elongation) of electrospun CAS/PEO fibers.**

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|  | CAS/PEO 90/10 | CAS/PEO 80/20-2% TA | CAS/PEO 90/10-2% TA | CAS/PEO 90/10-4% TA |
| Tensile strength, *TS*  (MPa) | 0.91±0.19 | 1.67±0.22 | 0.50±0.18 | 1.88±0.31 |
| Breaking elongation, *BE* (%) | 93.74±20 | 182.91±10 | 125.16±22 | 274.56±15 |

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