Supporting Information. Materials. N,N-dimethylaminoethylacrylamide (DMAEAm; Monomer-Polymer and Dajac Labs, Feasterville, PA) was twice distilled through a short-path. N-isopropylacrylamide (NIPAAm; Aldrich, 97%) was recrystalized from hexanes and dried under vacuum prior to use. 4-Cyano-4-(dodecylsulfanylthiocarbonyl)sulfanyl pentanoic acid (DCT) was synthesized as described in the literature[63]. 2,2-Azobis(2-methylpropionitrile) (AIBN; Aldrich, 98%) was recrystalized from methanol. Methanol (EMD, 99.8%), hexane (EMD, 89.2%), dimethylformamide (Aldrich, 99.9%), tetraglyme (Aldrich, 99%), $HAuCl_4$ (Aldrich, 99.9%), $KAuCl_4$ (Aldrich, 99.9%), $Fe(CO)_5$ (Aldrich, 99.9%), and 2kDa or 20 kDa MWCO Dialysis membrane (Pierce, Rockford, IL) were used as received. Magnets were NdFeB, 5 cm x 1.27 cm x 0.63 cm, B_r max =12,100 Gauss (Force Field Magnets.com)

Polymer Synthesis Procedure. The first polymerization was performed as previously described[31], except that 4-Cyano-4-(dodecylsulfanylthiocarbonyl)sulfanyl pentanoic acid (DCT; a RAFT chain transfer agent[63]) was used in place of 2-(Dodecylsulfanylthiocarbonylsulfanyl)-2-methylpropionic acid. The target molecular weight was 5 or 15 kDa, with a target degree of polymerization (dp_n) of 44, or 132, respectively. Further purification and dialysis of pNIPAAm was subsequently undertaken as described below. The second RAFT polymerization proceeded by loading a flask with 83 μmol (1.32 g) of the 15 kDa purified pNIPAAm mCTA, 1.24 mmol (0.19 g) DMAEAm, 1.24 mmol (0.15 g) NIPAAm, 8.3 μmol (1.4 mg) AIBN (free radical initiator), and 8 mL methanol (MeOH). The flask was thoroughly purged with N_2 followed by heating (60ºC, 24 hrs.) and subsequent evaporation of MeOH. The product was dissolved in 5 mL of tetrahydrofuran and precipitated thrice into n-pentane. Further purification and dialysis of the polymer was also performed as described below.

^{1}H-NMR of Polymers. The ^{1}H-NMR spectrum in $CDCl_3$ obtained on a Bruker AV300 ^{1}H-NMR spectrometer showed peaks corresponding to the NIPAAm hydrogens at δ (ppm) = 1.15 (s, $R\text{-}CO\text{-}NH\text{-}CH\text{-}(C\underline{H}_3)_2$) and at δ = 4.00 (s, $R\text{-}CO\text{-}NH\text{-}C\underline{H}\text{-}(CH_3)_2$). Other peaks corresponding to the CTA hydrogens were observed at δ = 1.27 (s, $CH_3\text{-}(C\underline{H}_2)_9\text{-}(CH_2)_2\text{-}CS_3\text{-}R$); δ =2.51 (m, $R\text{-}C\underline{H}_2C\underline{H}_2\text{-}COOH$); δ = 3.23 (br t, $R\text{-}C\underline{H}_2\text{-}CS_3\text{-}R$). ^{1}H-NMR results from the ACR-dB preparations confirmed the presence of

DMAEAm with an additional peak observed at $\delta = 2.26$ (s, R-N(C$\underline{H}_3$)$_2$). The integrated ^{1}H-NMR peak ratio of the DMAEAm peak at 2.26 to the NIPAAm peak at 4.00 showed that the DPN of DMAEAm in the final diblock polymers was ~8-12. The content of DMAEAm in the 7.2 kDa and 16 kDa ACR-dB polymers was ~17.6% and ~8.3%, respectively. All ^{1}H-NMR spectra showed peak broadening due to the polymeric nature and polydispersity of the sample.

SEC Chromatography. Size-exclusion chromatography was performed (8 mg/mL in dimethylformamide) on a Viscotek VE2001 GPC Solvent/Sample module equipped with a VE3580 refractive index (RI) detector. Results are shown in Figure S1. Molecular weight and polydispersity data were obtained by comparison to PMMA standards.

Purification of pNIPAAm and ACR-dB. The following was used to further purify the precipitated pNIPAAm and ACR-dB polymers prior to use. Each polymer was dissolved in deionized (DI) water at a concentration of ~15 mg/mL. 5M NaCl in DI water was added drop-wise under vigorous stirring until the polymer precipitated out of solution. This precipitate was collected through centrifugation at 40ºC / 10,000 RPM for 8 minutes. The polymer was then dialyzed with a molecular weight cutoff of 2 kDa for several days against frequent replacement of 4L of DI water at 4 ºC. The dialyzed solution was frozen under liquid nitrogen, and lyophilized to obtain the pNIPAAm mCTA or the ACR-dB copolymer as a slightly yellow powder.

Magnetic Enrichment of mNPs. The precipitated dark brown material (~60 mg) was dried overnight under vacuum, and dissolved in 25 mL of pH 8 phosphate buffered saline (PBS). 5M NaCl was added dropwise under vigorous mixing until the material was observed to precipitate (indicated by a sharp increase in turbidity). A permanent magnet was then affixed to the side of the tube, and the material was allowed to collect at the surface of the magnet (NdFeB, 12,100 Gauss) for 5 minutes. The supernatant was decanted, and the magnetically enriched material was re-suspended in DI water. The solution was then dialyzed with a molecular weight cutoff of 20 kDa against frequent replacement of 4L of deionized water for 48-72 hours, and obtained as a powder through freeze drying.

Magnetic Enrichment of Au-mNPs. A permanent magnet was affixed to the side of the Au-mNP reaction vial. The vial was placed in a 45 ºC oven for 15 minutes. The supernatant was decanted, and the captured material was resuspended in 150 μL of DI water. This material was then dialyzed against 4L of DI water for 24 hours, and subsequently taken for measurement by UV-Vis-NIR absorbtion spectroscopy, or TEM.

Figure S1. Size exclusion chromatography traces of the ACR-dB and mCTA polymers. Homo-pNIPAAm mCTA chains with lengths of 5 kDa (black dots) and 15 kDa (solid line) were used in RAFT copolymerization of DMAEAm-co-NIPAAm in the second block. The resulting ACR-dB polymers had molecular weights of 7.2 kDa (white dots), and 16 kDa (dashed line).

Figure S2. Absorbance spectrum of the AuNPs shown in Figure 3A as a function of temperature. The absorbance spectrum of the 7.2 kDa ACR-dB / AuNPs dissolved in pH 9 PBS was taken at room temperature (black), 38.9 ºC (orange), and cooled back to 24.2 ºC (blue). Spectra were normalized to maximum absorbance peak height in the 540-580 nm range.

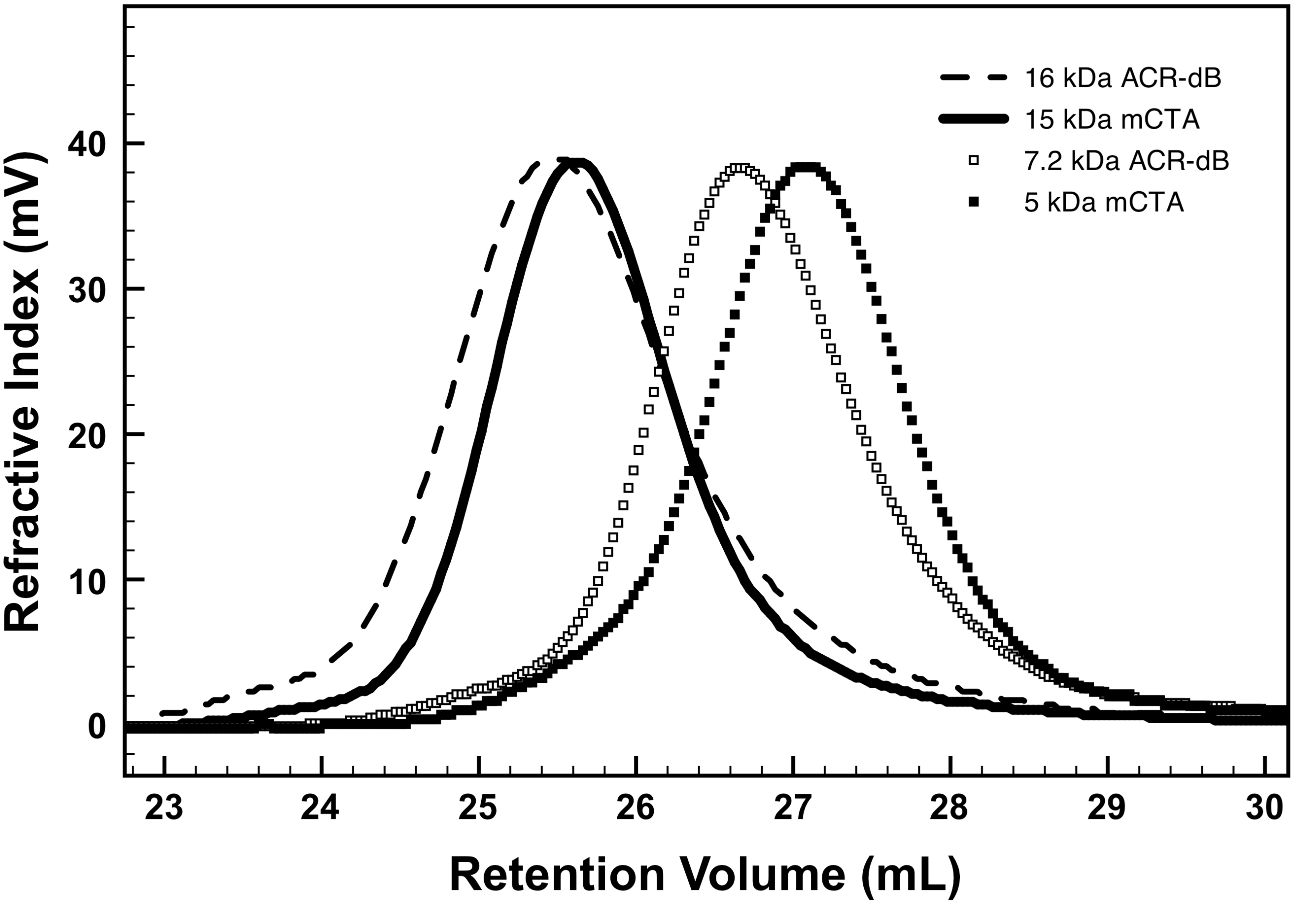
Figure S3. Field magnetization curve of mNPs. These mNPs were synthesized with the 7.2 kDa ACR-dB copolymer. Magnetization values were normalized per gram of inorganic material as determined by thermal gravimetric analysis (data not shown).

Figure S4. Field-cooled / zero-field cooled magnetization curve of 7.2 kDa ACR-dB mNPs. The blocking temperature was found to be 22 K. Magnetization values were normalized per mass of inorganic material as determined by thermal gravimetric analysis (data not shown).

Figure S5. HR-TEM image and corresponding EDXS spectrum of a single AuNP formed during Au-mNP synthesis. This particle was found to have a FeK α:CuK β peak ratio equal to background levels, indicating there was no Fe present in the sampling volume; the gold signal remained strong.

Figure S6. HR-TEM image and corresponding EDXS spectrum of a small cluster of mNPs that remained in the sample after Au-mNP synthesis. This cluster was found to have a FeK α:CuK β peak ratio of 0.56, with no gold signal present.

Figure S7. TEM image of Au-mNPs demonstrating particle fusion and elongation of fused products.



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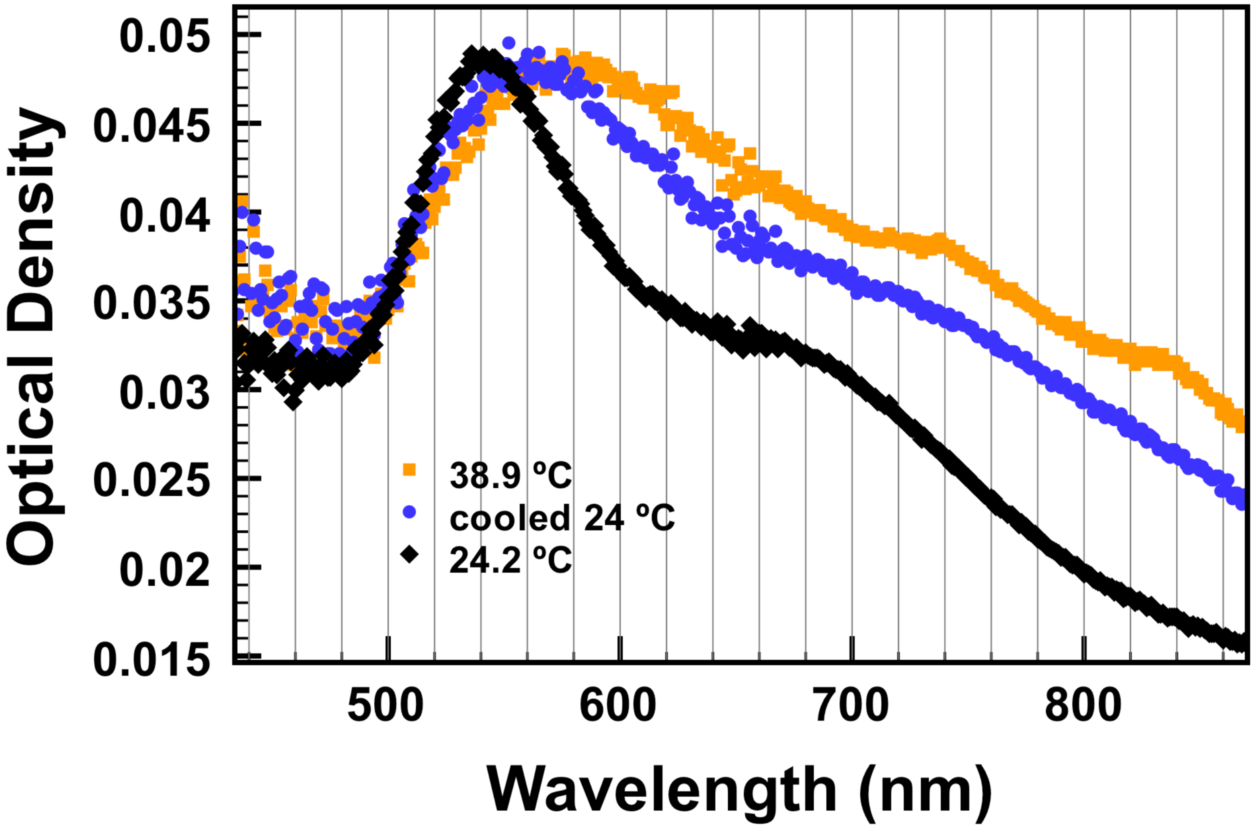

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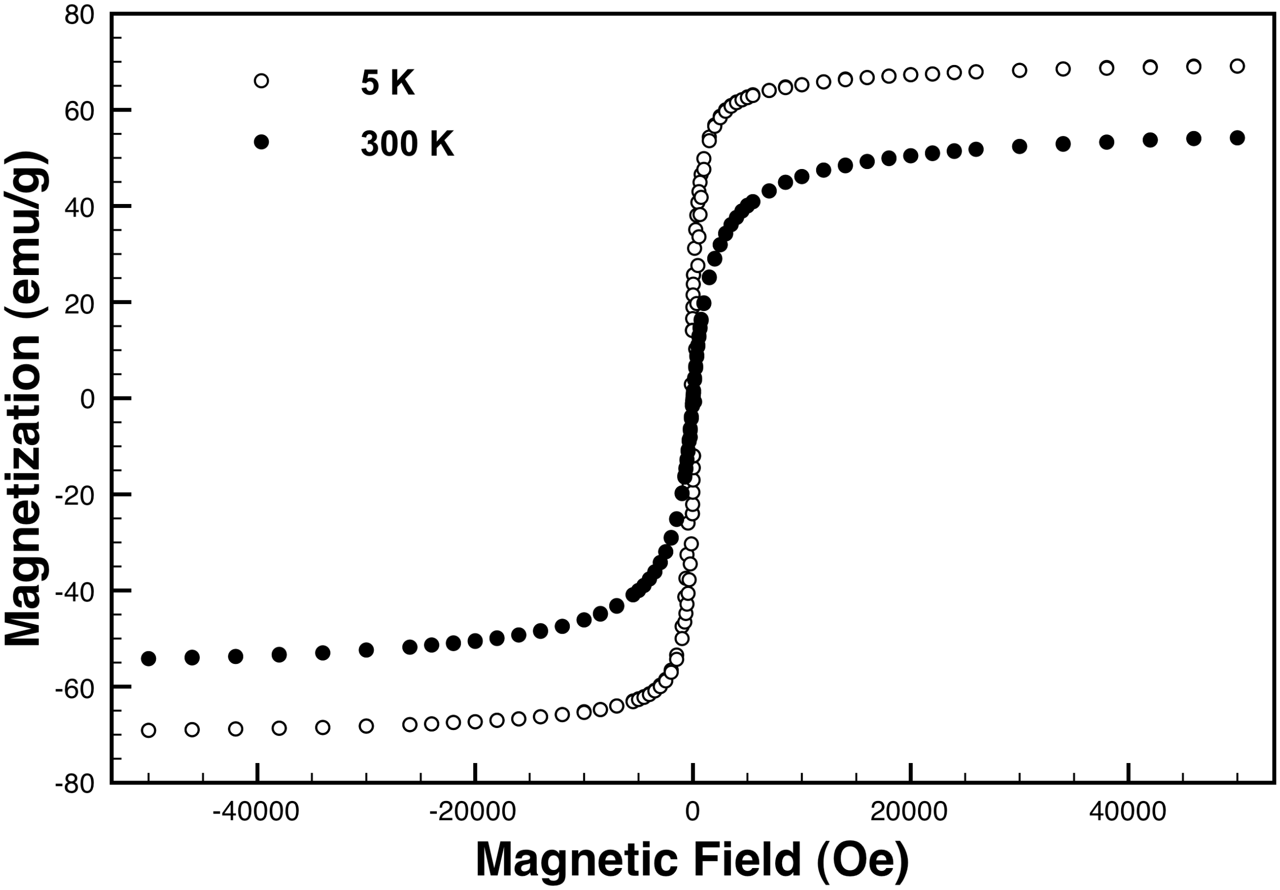

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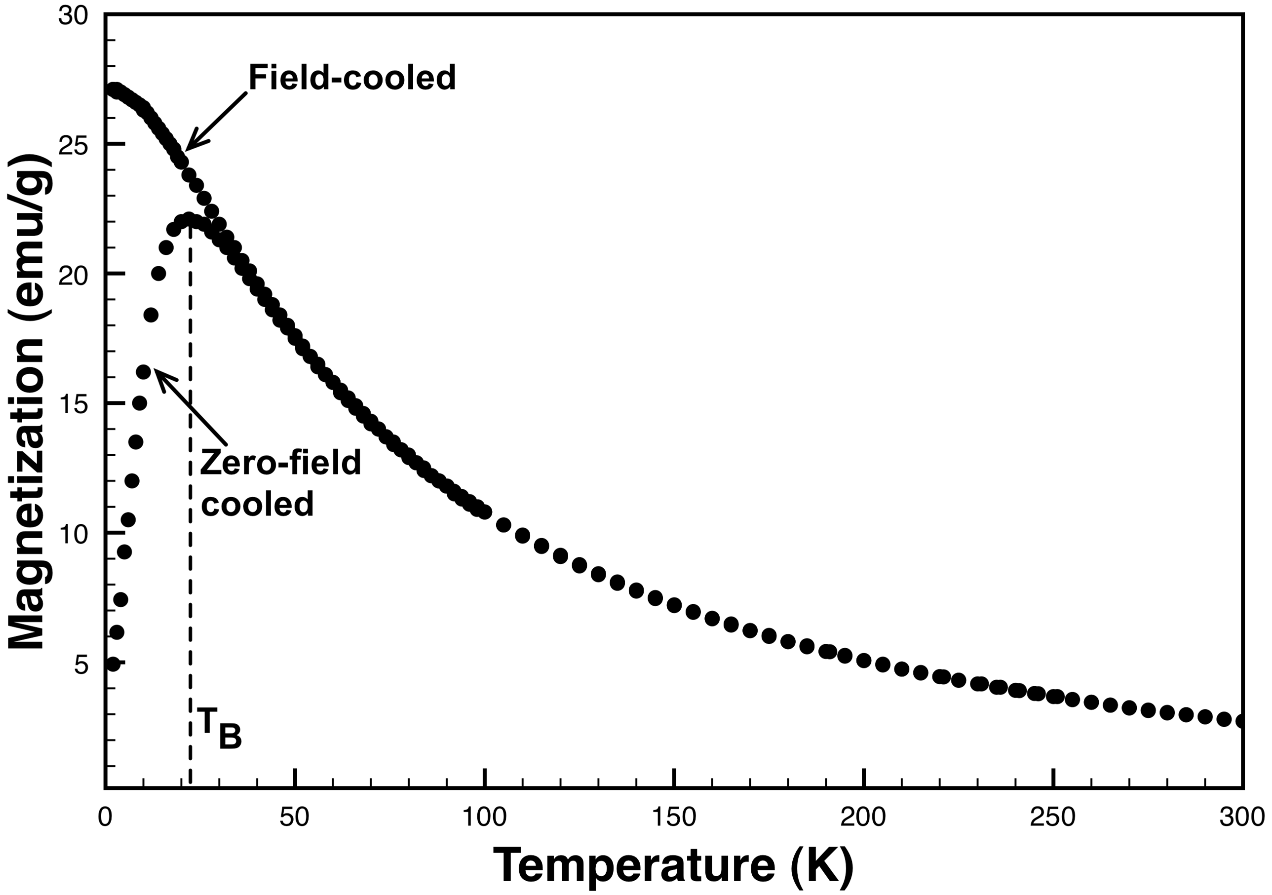

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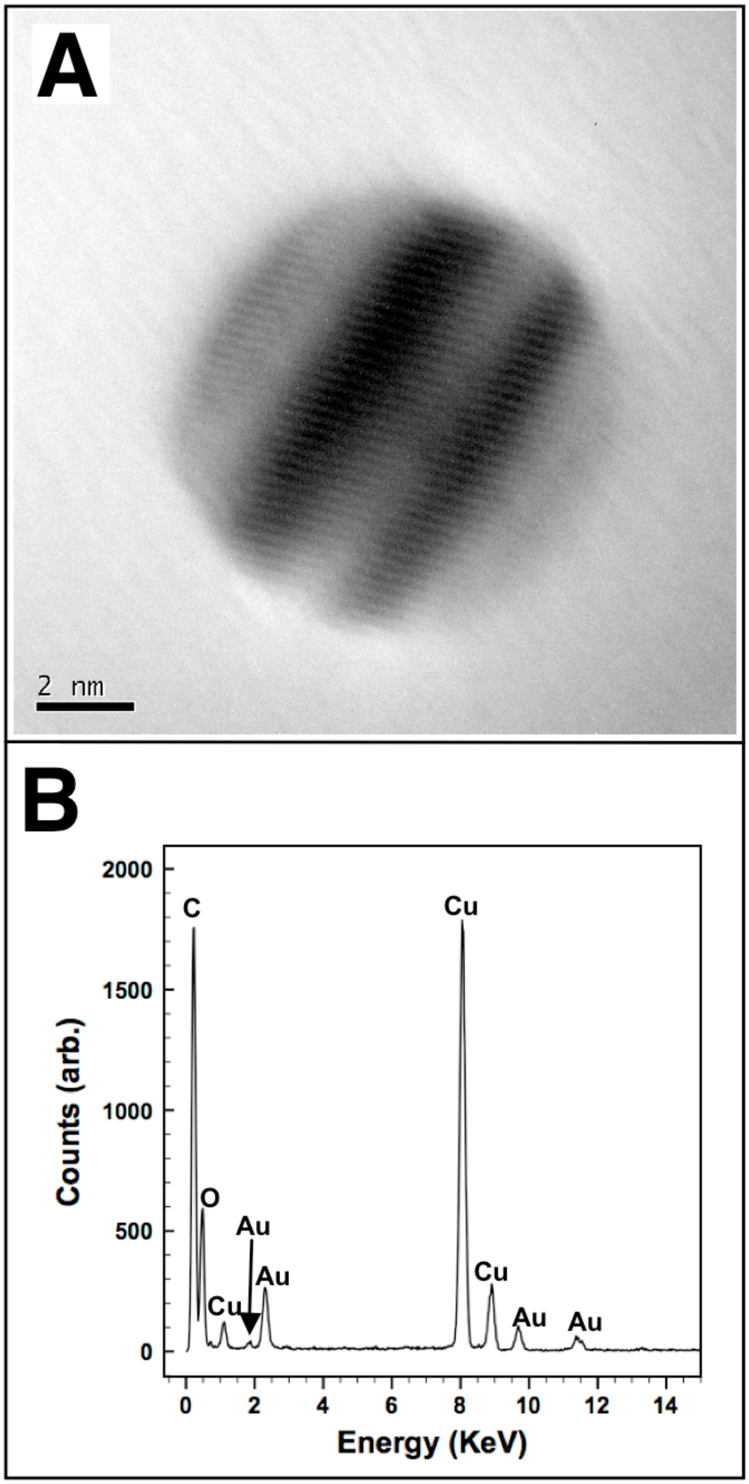

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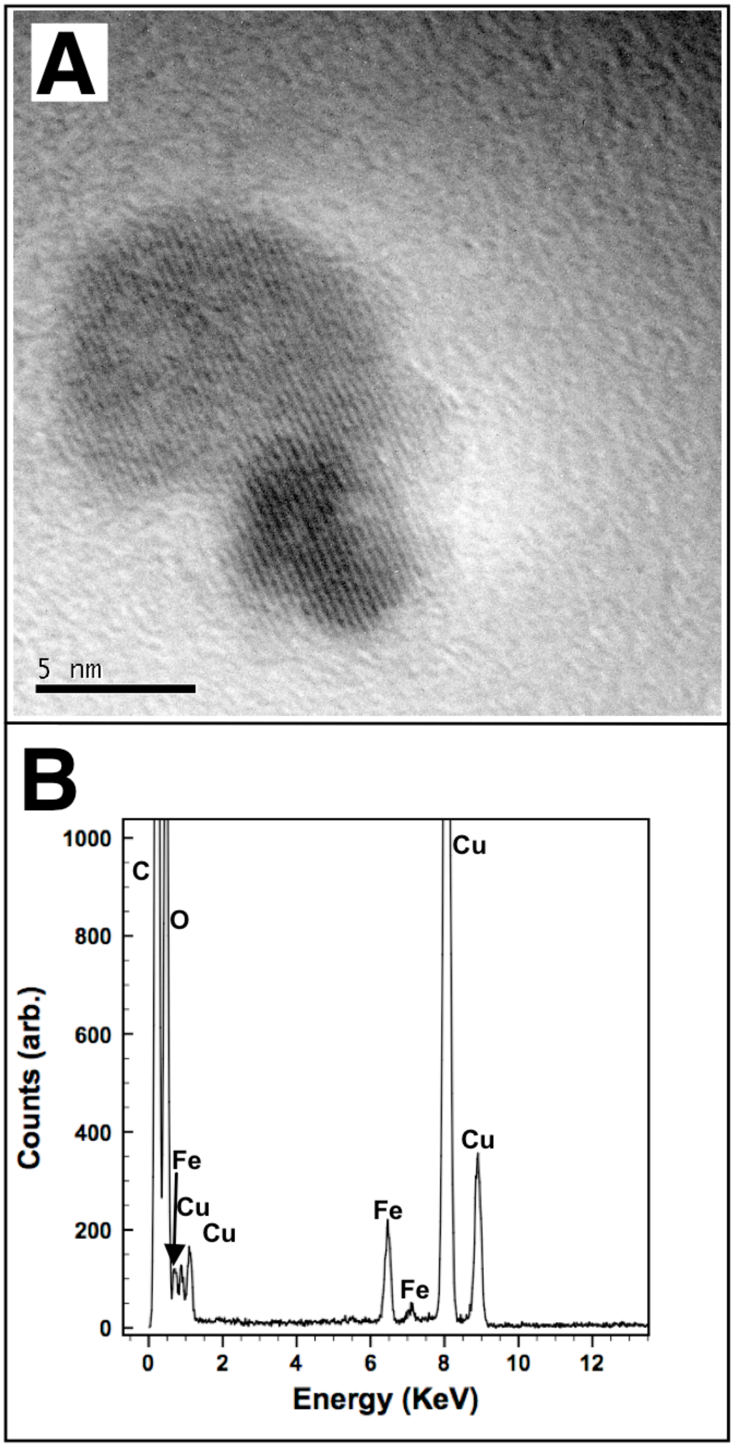

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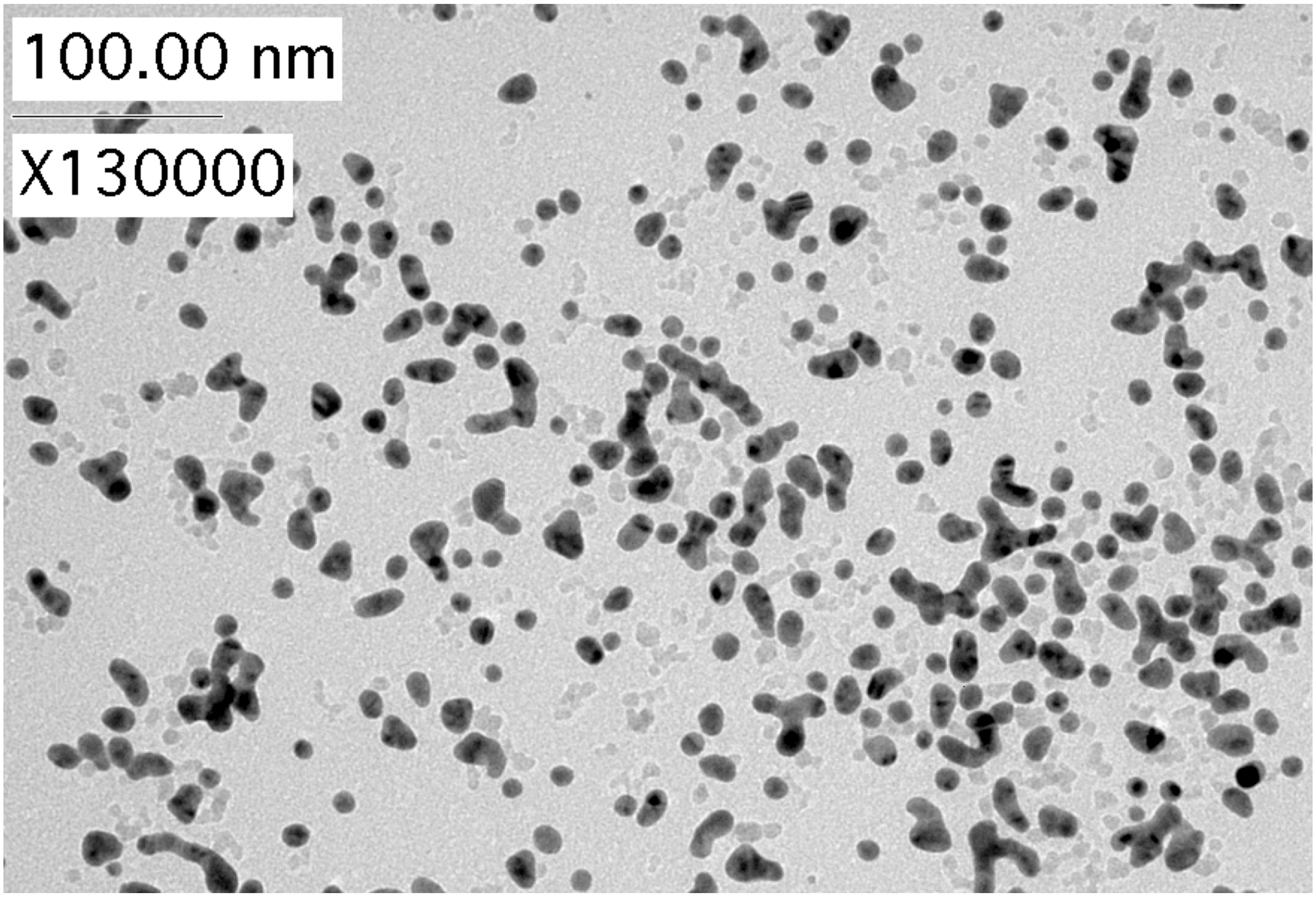

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