Answers to comments of reviewer 2

Comment from the authors to this review:

Below are given detailed answers to all specific comments and questions of reviewer 2. Most comments by the reviewer have already been covered in the original manuscript (see details below), indicating that the reviewer did not seriously read our manuscript.

A main generic accusation of this review claims that we insufficiently reference previous work. There is always the possibility of missing a relevant publication. However, we reference 67 relevant publications in our manuscript related to all relevant results and discussions in our study. If relevant publications are missing, a proper review should mention them explicitly rather than accusing authors of omitting them intentionally.

We also like to state very clearly that the language and generic accusations in this review are unacceptable for a serious scientific discussion (see also the Chief Editor Comment to this review)! Such reviews are the exact opposite of what a thorough and fair scientific discussion should be, which is the very essence of the entire peer-review process and one of the core foundations of scientific progress. We therefore hope that this review and its style of comments will serve as a pointed negative example of poor scientific discussion and reviewing process.

General comments

Overall the work is written in a vague manner without direct means for relevant confirmations. This work also made the impression for reproducing already published works published years ago but not yet fully cited them. This "gave the chance" of this work to pretend to be the initial discoveries. The morphologies of the NPs demonstrate that this study did not yet optimize the system. Only switching on the generator and collecting them for subsequent analysis cannot add scientific values, particularly ignoring the large volume of literature, which reported almost the same line of argumentations. The authors also try to use artificially texted letters to mark the different metals without showing any experimental proofs. Raising such concern is mainly due to all their similar appearances but undistinguishable nature from the TEM results.

See comments above.

1) Primary particle was defined scientifically incorrect.
   We defined Primary Particles as is usually done in similar publications in line 187-188 (see, e.g., (Tabrizi et al., 2009) p.319)

2) Where the authors showed the 1-nm particles?
   The 1 nm particles are visible on the TEM images of Figure 2 and quantified in Figure 3.

3) Melting point of the 4 metals cannot be directly used to evaluate their different particle sizes, as the impure surface of the particles also makes dramatic influences.
   The impure (mostly oxidized) particle surfaces are described in the manuscript already, affecting mostly Cu and Ni. Surface oxidation increases melting points compared to the pure metal (line 214-217). Two more publications were added in line 219 (Seipenbusch et al., 2003) and line 217 (Olszok et al., 2023) describing the oxide formation and limitations in

particle growth of oxidised particles. (line 214-219)

4) How the authors prove that the oxidation took place during NP production?
Oxidation during spark production is a process well described in literature using the same conditions as we do; e.g. a gas purity of 5.0. The relevant literature is mentioned and cited in line 215-217. One more paper was added (Olszok et al., 2023).

5) Why the mass was evacuated using micro molar? As the authors also claimed that the SDG can also deliver NPs of high quantity. How the amount of micro molar supports the aforementioned claim?
The mass was given in micromolar to be able to compare all four metals more easily. Providing mass in milligrams only would complicate this comparison given the different densities of the various metals. This is also done by multiple other studies such as (Schmidt-Ott, 2019; Tabrizi et al., 2009) The term 'high quantity' is used to define the *number* of particles (in the range of 1E8 particles per cm^3).

6) For the different production rates of the metals, previous study already clearly explained why. Please cite the relevant literature, not only saving the unnecessarily repeated work but also not making an impression of confined literature review.
The paragraph describing the different production rates was extended to include more than one literature reference (Domaschke et al., 2018; Schmidt-Ott, 2019). L254-261

7) The authors also argued that the mismatch of size distributions measured between the TEM analysis and SMPS data is mainly due to the low counting efficiency from the latter. How to explain the overestimation of diffusional deposition of smaller NPs for TEM analysis?
The overestimation of smaller particles via the deposition through diffusion is described in the manuscript (line 281-290). Therein, we also discuss in detail how to correct this collection bias and show results of this correction in Figure 4.

8) A very puzzled presentation for Fig. 5 onwards is to remove the Cu NPs. Could the authors provide any scientific reasons for such inconsistencies?
The reason why the results for Cu were not considered anymore in the main text of the manuscript can be found in line 324-325 A sentence was added on where the data on Cu can be found (i.e. appendix B) in line 325.

9) How the edges of the NPs were determined when using eq. 1 to calculate circularity?
This is described in detail in line 141-145.

10) How the authors separate the ones coated over TiO2 with those of self-coagulation?
This topic is thoroughly discussed in the manuscript in line 361-379.

Literature

Domaschke, M., Schmidt, M., and Peukert, W.: A model for the particle mass yield in the aerosol synthesis of ultrafine monometallic nanoparticles by spark ablation, Journal of Aerosol Science, 126, 133–142, https://doi.org/10.1016/j.jaerosci.2018.09.004, 2018.

Olszok, V., Bierwirth, M., and Weber, A. P.: Creation of Gases with Interplanetary Oxygen Concentration at Atmospheric Pressure by Nanoparticle Aerosol Scavengers: Implications for Metal Processing from nm to mm Range, ACS Appl. Nano Mater., 6, 1660–1666, https://doi.org/10.1021/acsanm.2c04585, 2023.

Schmidt-Ott, A. (Ed.): Spark Ablation: Building Blocks for Nanotechnology, Jenny Stanford Publishing, New York, 472 pp., https://doi.org/10.1201/9780367817091, 2019.

Seipenbusch, M., Weber, A. P., Schiel, A., and Kasper, G.: Influence of the gas atmosphere on restructuring and sintering kinetics of nickel and platinum aerosol nanoparticle agglomerates, Journal of Aerosol Science, 34, 1699–1709, https://doi.org/10.1016/S0021-8502(03)00355-0, 2003.

Tabrizi, N. S., Ullmann, M., Vons, V. A., Lafont, U., and Schmidt-Ott, A.: Generation of nanoparticles by spark discharge, J Nanopart Res, 11, 315–332, https://doi.org/10.1007/s11051-008-9407-y, 2009.