Supporting Information

## Flexible Perovskite Solar Cells Fabricated by Gradient Heat Treatment Process

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## The content of supporting information

- 1. The statistics of grain size distribution of perovskite.
- 2. Possible reasons for perovskite grain size increase.

## 1. The statistics of grain size distribution of perovskite

As shown in Fig. S1, the statistics of grain size distribution of perovskite films calculated by Nano measurement software and fitted by Gaussian function. The lager fall-off rate (GHT-1 and GHT-2) led to average grain size as small as 0.6~ $0.8 \mu$ m. The grain size gradually increased to an average of 1.2  $\mu$ m as the fall-off rate slowed to GHT-4. However, when the fall-off rate dropped to GHT-5, the average grain size is 1.02  $\mu$ m, which does not increase further.



**Figure S1.** Statistics of grain size distribution of perovskite films fabricated by different GHT process from GHT-1 to GHT-5.

## 2. Possible reasons for perovskite grain size increase

By observing the top-view SEM image, the reason why the grains size became larger was speculated when the fall-off rate was reduced. As shown in Fig. S2, the circles represent the phenomenon that small grains are swallowed by large grains, and the squares represent the fusion phenomenon between grain boundaries. These two phenomena all lead to the gradual blurring and eventually disappear of grain boundaries. We guess that slowing down the fall-off rate is equivalent to prolonging the holding time. Thermal energy makes the grain boundaries of small grains have a higher interfacial energy. The diffusion of grain boundaries of small grains makes it disappear gradually, as if swallowed up by large grains. The fusion of grain boundaries between larger grains may be due to thermal energy-induced interdiffusion at grain boundaries. When the chemical potential energy on both sides of grain boundaries is equal, the grain boundaries here tend to disappear. When the fall-off rate is further slowed down, the grains that have been grown by thermal diffusion tend to grow towards a more perfect shape, which eventually leads to more distinct edges and angles.



**Figure S2.** Top-view SEM diagram of grains growth and disappearance of grain boundaries. Circles indicate the small grains are engulfed by large grains, and squares indicate the fusion of grain boundaries.