**Appendix A. Spherical harmonic function**

The basic idea of the *SH* basis functions is to extend the polar radius of the particle surface from the unit sphere and to use the correlation coefficients of the SH series as a mathematical representation of the three-dimensional particle morphology, as shown in Eq. (A1).

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|  | (A1) |

where, *r*(*θ*, *φ*) denotes the distance from the center to the particle surface; and are the spherical coordinates;  is the SH function obtained by the Eq. (A2); *N* is the degree of the *SH* series,  is the *SH* coefficients, which can be obtained by the Eq. (A4).

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| --- | --- |
|  | (A2) |
|  | (A3) |

where,is the associated Legendre polynomial function given by Eq. (A3); *m* and *n* *a*re the degree and order of , *m=* (-*n*, -*n*+1, …, 0, …, *n*-1, *n*) and *n* < *N*. To visualize the variation mechanism of the spherical harmonic function, Fig. A.1 exhibits the basis image of 0-3rd order spherical harmonic function.

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| 图表, 气泡图  描述已自动生成 |
| **Fig. A.1** Example of spherical harmonic function base image of order 0-3 |

Based on the calculated radius and the spherical function of the reconstructed point, with reference to Garboczi et al.，the spherical harmonic coefficients can be solved by Eq. (A4).

|  |  |
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|  | (A4) |

Taking the surface vertices *r*(*θ*, *φ*) obtained from 3D scanning as the inputs to the left side of Eq (1), it is easy to determine  by applying standard least-squares estimation to the linear equations. Thus, the 3D morphology of particles can be succinctly characterized by a set of .