POW 2024-01

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March 17, 2024

Let us begin with the following lemma.

Lemma 1. *For any $n \geqslant 1$, let $A_{n,k}$ be the number of different outcomes we can get by rolling n dice with the sum of the faces congruent to k modulo 6. Then, $A_{n,0} = A_{n,1} = \cdots = A_{n,5} = 6^{n-1}$.*

Proof. Let $B_{n,m}$ denote the number of different outcomes we can get by rolling n dice with the sum of the faces equal to m. Note that, for any fixed n, the generating function of $\{B_{n,m}\}_{m \geqslant 0}$ is $(x + x^2 + \cdots + x^6)^n$. As $A_{n,k} = \sum_{t=0}^{\infty} B_{n,6t+k}$ for each $0 \leqslant k \leqslant 5$, it holds that

$$A_{n,0} + A_{n,1}x + \cdots + A_{n,5}x^5 \equiv (x + x^2 + \cdots + x^6)^n \pmod{x^6 - 1}.$$

Now we use induction on n, to show that $A_{n,k} = 6^{n-1}$ for each $0 \leqslant k \leqslant 5$. It is clear that $A_{1,k} = 1$ for each $0 \leqslant k \leqslant 5$. Now, suppose that for some n, it holds that $A_{n,k} = 6^{n-1}$ for each $0 \leqslant k \leqslant 5$. Then, one can observe that

$$\begin{aligned} A_{n+1,0} + A_{n+1,1}x + \cdots + A_{n+1,5}x^5 &\equiv (x + x^2 + \cdots + x^6)^{n+1} \\ &\equiv (A_{n,0} + A_{n,1}x + \cdots + A_{n,5}x^5)(x + x^2 + \cdots + x^6) \\ &\equiv 6^{n-1}(1 + x + \cdots + x^5)(x + x^2 + \cdots + x^6) \\ &\equiv 6^{n-1}(6 + 6x + \cdots + 6x^5) \qquad \pmod{x^6 - 1} \end{aligned}$$

where the last line can be verified by directly computing the product by expanding the terms. In particular, we get that $A_{n+1,k} = 6^n$ for each $0 \leqslant k \leqslant 5$, which completes the proof. □

For $k \geqslant 7$, it is clear that the probability of S_1 being divisible by k is 0, not $1/k$.

Also, when $k = 4$ or 5, it is clear that the probability of S_1 being divisible by k is $1/6$, not $1/k$.

On the other hand, when $k = 1, 2, 3$, or 6, because k is a divisor of 6, the sum of the faces is divisible by k if and only if the remainder of that sum when divided by 6 is divisible by k. Hence, from **Lemma 1**, it follows that for any $n \geqslant 1$ the probability of S_n being divisible by k is

$$\frac{\sum_{\substack{k \mid \ell \\ 0 \leqslant \ell \leqslant 5}} A_{n,\ell}}{A_{n,0} + A_{n,1} + \cdots + A_{n,5}} = \frac{\frac{6}{k} \cdot 6^{n-1}}{6 \cdot 6^{n-1}} = \frac{1}{k}.$$

Therefore, all the positive integers that satisfy the desired property are exactly 1, 2, 3, and 6.