

Table S1 Raw data and peptide sequences for results shown in Figure 6C												
Antigen	Amino Acid Sequence	DR4 CithFib-1	DR4 CithFib-2	DR4 CithFib-3	DR4 CithFib-4	DR4 CithFib-5	B6 CithFib-1	B6 CithFib-2	B6 CithFib-3	B6 CithFib-4	B6 CithFib-5	
A2591 hFibB211-230cit	KLESDVSAQMEY[CIT]TPCTVS	311	217.5	2131.5	2287.5	157	-22	6.5	4	86	69	
A2583 hFibB46-65cit	H[CIT]PLDKK[CIT]JEEAPSL[CIT]PAPP	736.5	549.5	3294	2588	3085.5	22620	8322	876.5	2119	45967.5	
A2579 hFibB451-470	QYTWDMAKHGTDGCVVWNNW	154.5	440	3309	4388	2104.5	7	66.5	31.5	819	1868.5	
A2577 hFibB421-440	RKQCSKEDGGGWYVNRCHAA	4697	3915	3526	1815.5	5405	3362	275	348	2168.5	1903	
A2572 hFibB346-365	DWKGDKVKAHYGGFTVQNEA	-0.5	1	132.5	80.5	-3.5	374	36.5	122	222	360.5	
A2563 hFibB211-230	KLESDVSAQMEYCRTPCTVS	18	80.5	918.5	387	1859	-55	21.5	3	104.5	-29.5	
A2558 hFibB136-155	AVSQTSSSSFQYMYLLKDLW	530.5	1640	10899	13046.5	6789	812.5	212	176.5	888	-147.5	
A2556 hFibB106-125	CPTGCQLQEALLQQRPIRN	50.5	158.5	717.5	1187	355.5	88.5	7	22	297.5	-35.5	
A2555 hFibB91-110	DAGGCLHADPDLGLVCLPTGC	1136	2496	3506.5	2586	325	64819.5	36411	1928.5	5991.5	11568	
A2549 hFibB1-20	MKRMVSWSFHKLKTKMKHLL	19.5	-18	50	56	5	82	64	72.5	327	-97	
A2548 hFibA616-635cit	THSTK[CIT]GHAKS[CIT]JPV[CIT]GIHTS	25	76	335	251.5	130	588	25	42	89	-82.5	
A2544 hFibA541-560cit	VSETES[CIT]GSESGIFTNTKES	412	329.5	156	25.5	153	6027	4675.5	947	31168	8462.5	
A2537 hFibA406-425cit	NNPDWGTFEEVSGNVSPGT[CIT]	294.5	1916	41.5	46.5	-14	12635.5	19027	65.5	47526	13015.5	
A2536 hFibA391-410cit	GSF[CIT]PDSPPSGGNA[CIT]PNNDPW	1795	1888.5	855.5	846	22	216	15	820	256.5	64	
A2527 hFibA211-230cit	DLLPS[CIT]D[CIT]QHLPLIKMKPVP	160.5	154.5	574	202.5	10.5	1312.5	114	332	5097	4729.5	
A2522 hFibA121-140cit	NN[CIT]DNTY[N[CIT]VSEDL[CIT]S[CIT]IEV	190.5	2185	4848	8539	909	486	2601	434.5	193.5	-39	
A2514 hFibA616-635	THSTKRGHAKSRPVRGIHTS	20	26.5	43.5	-2	154.5	352.5	4.5	185.5	182.5	-71.5	
A2509 hFibA541-560	VSETESRGSESGIFTNTKES	913.5	430	395.5	244	744	19142.5	7281	1337.5	19032.5	5222.5	
A2500 hFibA406-425	NNPDWGTFEEVSGNVSPGTR	4485.5	7634.5	138.5	21	1.5	34398	34434	3	41089	11574	
A2490 hFibA256-275	QMRMELERPGGNEITRGGST	32	39.5	28.5	15	124.5	162.5	22	74	290	-3.5	
A2476 hFibA46-65	ACKDSDWPFCSDEDWDWPF	22412.5	27109.5	35470	36047	14428.5	9955	2344.5	5510	9549.5	10603.5	

Antigen	Amino Acid Sequence	B6 hFib-1	B6 hFib-2	B6 hFib-3	B6 hFib-4	B6 hFib-5	DR4 hFib-1	DR4 hFib-2	DR4 hFib-3	DR4 hFib-4	DR4 hFib-5
A2591 hFibB211-230cit	KLESDVSAQMEY[CIT]TPCTVS	123	6.5	11.5	25	-59	428	8.5	6.5	60.5	23
A2583 hFibB46-65cit	H[CIT]PLDKK[CIT]JEEAPSL[CIT]PAPP	856.5	841.5	1176.5	1568	790	369.5	1367	1527.5	624.5	1277.5
A2579 hFibB451-470	QYTWDMAKHGTDGCVVWNNW	111.5	67.5	123	145.5	442	946.5	27.5	1281	200.5	760
A2577 hFibB421-440	RKQCSKEDGGGWYVNRCHAA	2388.5	485.5	1315	326.5	1132	3758.5	1871.5	3742	59.5	1776.5
A2572 hFibB346-365	DWKGDKVKAHYGGFTVQNEA	281.5	445	154	66	337.5	8.5	7.5	142.5	0	16
A2563 hFibB211-230	KLESDVSAQMEYCRTPCTVS	77	11.5	-18	3.5	-94.5	42	18	87	60	-9.5
A2558 hFibB136-155	AVSQTSSSSFQYMYLLKDLW	288.5	274.5	309.5	294.5	1469	5503	775	1834.5	1062.5	1498
A2556 hFibB106-125	CPTGCQLQEALLQQRPIRN	90.5	-4	39.5	13	-49.5	69.5	48.5	5.5	6	90.5
A2555 hFibB91-110	DAGGCLHADPDLGLVCLPTGC	442.5	154	690.5	356.5	119	921.5	765	2606	248.5	171.5
A2549 hFibB1-20	MKRMVSWSFHKLKTKMKHLL	160	288.5	544	467	273	59	100	-202	133.5	51
A2548 hFibA616-635cit	THSTK[CIT]GHAKS[CIT]JPV[CIT]GIHTS	973	617.5	836.5	155.5	236	21	74	-64	34.5	22.5
A2544 hFibA541-560cit	VSETES[CIT]GSESGIFTNTKES	5051	875.5	2119	152	1112	23	315.5	300	127	231.5
A2537 hFibA406-425cit	NNPDWGTFEEVSGNVSPGT[CIT]	15410	3808.5	1587.5	42.5	630	322	9166	305	1991.5	335
A2536 hFibA391-410cit	GSF[CIT]PDSPPSGGNA[CIT]PNNDPW	39.5	-11	4.5	-1.5	24	532	18.5	1032	14.5	7.5
A2527 hFibA211-230cit	DLLPS[CIT]D[CIT]QHLPLIKMKPVP	852.5	1924	227.5	-2.5	-13.5	26.5	9.5	62	-1.5	-9
A2522 hFibA121-140cit	NN[CIT]DNTY[N[CIT]VSEDL[CIT]S[CIT]IEV	156.5	25.5	25	21.5	-134	5.5	26.5	387.5	-3	-43
A2514 hFibA616-635	THSTKRGHAKSRPVRGIHTS	341	164.5	124.5	331.5	493	36	73.5	66	26	50.5
A2509 hFibA541-560	VSETESRGSESGIFTNTKES	8053	3534	12075	2716	18780.5	796	-2403	1826	1946	7849
A2500 hFibA406-425	NNPDWGTFEEVSGNVSPGTR	17103	7508.5	4308	8	1614	789	16323.5	220	9727	1430
A2490 hFibA256-275	QMRMELERPGGNEITRGGST	177.5	70.5	6.5	26	-59	29190.5	17.5	1231	3	26326
A2476 hFibA46-65	ACKDSDWPFCSDEDWDWPF	14573	7323.5	13009.5	12128	8963	10735.5	24532	25156.5	2157.5	13076.5

Values represent normalized arbitrary antibody units

Table S2

Raw data and peptide sequences for results shown in Figure 6E

Antigen	Amino Acid Sequence	DR4 CithFib-1	DR4 CithFib-2	DR4 CithFib-3	DR4 CithFib-4	DR4 CithFib-5	B6 CithFib-1	B6 CithFib-2	B6 CithFib-3	B6 CithFib-4	B6 CithFib-5	t-test
A711 fibrin	Protein	0	-3	-9	6	-29	3.5	2.5	30.5	14.5	64	0.0486
A551 hnRNP-A2 (17-38)	SFETTEESLRNYYEQWGKLTDC	62.5	187.5	511.5	245	106	51.5	17	31	90.5	-74.5	0.0439
A2500 hFibA406-425	NNPDWGTFEEVSGNVSPGTR	4485.5	7634.5	138.5	21	1.5	34398	34434	3	41089	11574	0.0261
A2462 vim286-305cit	EAEWYKSKFADLSEAAN[CIT]N	247	978.5	1966.5	1323	1330	75.5	20	-19	274	111.5	0.0054
A2460 vim256-275cit	IDVDVSKPDLTAAL[CIT]DV[CIT]QQ	37.5	2643.5	2313	3678	-177	-58	-7.5	-38	-105.5	-66.5	0.0492
A2433 vim286-305	EAEWYKSKFADLSEAANRN	107.5	140	465	395	385.5	52.5	14.5	8	241	-105.5	0.0237
A2396 H2B/r 16-36	KKAVTKAQKKDGKSAHRKE	195.5	348.5	598	339.5	534.5	42	44	40	52	-32	0.001
A2371 H2A 79-98	IIPRHLQLAIRNDEELNKL	1041.5	689.5	7690.5	10433	5050.5	943.5	72.5	175	802	378	0.0444
A2353 Serine Protease 11 391-410 cit	TSSKAKELKD[Cit]H[Cit]DFPDVIS	5	178	123	206.5	246.5	-94	19.5	9	62	-25.5	0.0127
A2348 Vitronectin 223-242 cit	KGNQYW[Cit]FEDGVLDPDYP[Cit]N	24.5	35.5	52.5	131.5	223	-53.5	4	16	12.5	-66.5	0.0278
A2346 Vitronectin 196-215 cit	I[Cit]DVWGIIEGPIDAAFT[Cit]INC	35.5	69.5	633	399	719.5	5	31	11.5	76.5	-21.5	0.038
A2338 Clusterin 416-435 cit	EQFNWVS[Cit]LANLTQGEDQYY	396.5	231.5	1352.5	512	613.5	145	26	211	201.5	209	0.0465
A2321 Fibromodulin 345-364 cit	LQVV[Cit]LDGNEIK[Cit]SAMPADA	14.5	228.5	260	414	63.5	-64.5	9	4	124	-8.5	0.0469
A2317 Fibromodulin 261-280 cit	DSYF[Cit]GAPKLLYV[Cit]LSHNSL	47.5	1.5	426.5	463.5	510.5	35.5	51	8	112.5	-51.5	0.05
A2316 Fibromodulin 246-265 cit	LEQLYMEHNNVYTVPSYF[Cit]	117	130	770.5	1072.5	1912.5	-218	20.5	12	98.5	-15	0.0412
A2305 Biglycan 238-257 cit	HNKIQAIELEDLL[Cit]YSKLY[Cit]	143.5	251.5	1089	1541.5	2259.5	207	71	-22.5	228	-26	0.0428
A2301 COMP 468-487 cit	CDDDDNDGVPDS[Cit]DNC[Cit]LV	340	4137	3573.5	3649.5	1717	1226	25	259	84	228.5	0.0147
A2293 Serine Protease 11 433-452	VIISINGQSVVSANDVSDVI	4	32.5	295.5	324	192.5	1.5	14	6.5	24	-64.5	0.0333
A2285 Vitronectin 223-242	KGNQYWRFEEDGVLDPDYPRN	17.5	50.5	112.5	74	279.5	-107.5	13.5	15.5	-4	-43	0.0332
A2278 Clusterin 472-491	PVEVSRKNPKFMETVAEKAL	23	544.5	1148	736.5	404.5	-6.5	13.5	29	55.5	235.5	0.0291
A2232 COMP 498-517	DGVGDVCQDDFDADKVVDKI	36.5	37.5	184.5	135.5	31	-86.5	6.5	18.5	4.5	-12.5	0.0275
A2214 HSP90	Protein	49	244	891	547.5	294.5	82	43	156	21	-20	0.0464
A2003 cfc2	SHQESTRG[CIT]SRGRSGRSGS	309.5	220	374.5	79.5	196	106	37.5	32.5	139.5	-27	0.0157

Values represent normalized arbitrary antibody units

Table S3

Raw data and peptide sequences for results shown in Figure 6D

Antigen	Amino Acid Sequence	DR4 CithFib-1	DR4 CithFib-2	DR4 CithFib-3	DR4 CithFib-4	DR4 CithFib-5	B6 CithFib-1	B6 CithFib-2	B6 CithFib-3	B6 CithFib-4	B6 CithFib-5
A2471 vim421-440cit	LNL[CIT]ETNLDSLPLVDTHSK[CIT]	17396	65029.5	64943	65069	14256.5	56103.5	31	4657.5	1672	105.5
A2466 vim346-365cit	EMEENFAVEAANYQDTIG[CIT]L	300.5	4477	1763.5	5456	470.5	220	92	62	306	206.5
A2463 vim301-320cit	AAN[CIT]NNDAL[CIT]QAKQESTEY[CIT]	64.5	177.5	196.5	912.5	974.5	12	6.5	40.5	46	-36
A2462 vim286-305cit	EAEWYKSKFADLSEAAN[CIT]N	247	978.5	1966.5	1323	1330	75.5	20	-19	274	111.5
A2460 vim256-275cit	IDVDVSKPDLTAAL[CIT]DV[CIT]QQ	37.5	2643.5	2313	3678	-177	-58	-7.5	-38	-105.5	-66.5
A2454 vim136-155cit	EQLKGQGKS[CIT]LGDLYEEEM[CIT]	3413.5	1547	15016.5	17852.5	8289	5581	757.5	4158	5850.5	1682.5
A2452 vim106-125cit	ELQELND[CIT]FANYIDKV[CIT]FLE	2600.5	7086.5	6146	11309	5555.5	3377.5	1118.5	1393	3007	1987
A2448 vim46-65cit	PSTS[CIT]SLYASSPGGVYAT[CIT]S	2344	1269	214.5	1507.5	297.5	670.5	469.5	1018	1459	287
A2438 vim361-380	TIGRLQDEIQNMKEEMARHL	68	304	4045.5	2068	2626.5	1546.5	40	183.5	224	318
A2432 vim271-290	DVRQQYESVAAKNLQEAEEW	4540.5	4876	26704	24794.5	14245	6030	613	1151.5	2539	3669

Antigen	Amino Acid Sequence	B6 hFib-1	B6 hFib-2	B6 hFib-3	B6 hFib-4	B6 hFib-5	DR4 hFib-1	DR4 hFib-2	DR4 hFib-3	DR4 hFib-4	DR4 hFib-5
A2471 vim421-440cit	LNL[CIT]ETNLDSLPLVDTHSK[CIT]	260.5	-5.5	61	-3.5	376	295	4.5	-18	3.5	-48
A2466 vim346-365cit	EMEENFAVEAANYQDTIG[CIT]L	477.5	432	437	306	17.5	569.5	832	5251	143.5	1360.5
A2463 vim301-320cit	AAN[CIT]NNDAL[CIT]QAKQESTEY[CIT]	68.5	125.5	36.5	19.5	10	36	58	14.5	40.5	51
A2462 vim286-305cit	EAEWYKSKFADLSEAAN[CIT]N	99	74.5	962	41.5	33	808	120.5	313	63.5	225.5
A2460 vim256-275cit	IDVDVSKPDLTAAL[CIT]DV[CIT]QQ	-46	28	96	-336.5	-137	348.5	49	1136	-71.5	-30.5
A2454 vim136-155cit	EQLKGQGKS[CIT]LGDLYEEEM[CIT]	3301.5	932	763.5	364.5	1526	4431	2831.5	6628.5	807	2357.5
A2452 vim106-125cit	ELQELND[CIT]FANYIDKV[CIT]FLE	5302.5	4741	4771	6175	4827.5	1556.5	3014.5	9407.5	549	1124
A2448 vim46-65cit	PSTS[CIT]SLYASSPGGVYAT[CIT]S	2560.5	3214.5	2481.5	3480.5	839.5	203.5	3149.5	1602	119.5	172
A2438 vim361-380	TIGRLQDEIQNMKEEMARHL	186	10.5	352.5	11.5	-110	48.5	83.5	1104.5	38	253
A2432 vim271-290	DVRQQYESVAAKNLQEAEEW	1204.5	946	1382.5	1718	1377.5	6336	2964	11644	1727	7800

Values represent normalized arbitrary antibody units

Table S4. Arginine substitutions between sequences of human and mouse fibrinogen, their state of citrullination determined by mass spectrometry, and IgG antibody responses in DR4-IE tg and B6 mice immunized with CithFib.

Position of non-conserved arginines	Citrullinated in vitro	DR4 CithFib	B6 CithFib
aR123F	Yes	3147.7	623.8
aR129Q	Yes	3147.7	623.8
aR181K	No	-	-
aR216K	No	-	-
aR271-	Yes	32.8	20.6
aR287-	Yes	0	0
aR308-	No	-	-
aR353-	No	-	-
aR367S	Yes	0	0
aR427K	Yes	56.2	608.5
aR444L	Yes	0	288
aR459H	Yes	0	0
aR510S	Yes	49	0
aR547-	Yes	0	0
aR573S	Yes	270.2	48.9
aR591-	Yes	40.8	0
bR33K	Yes	7902.5	5221.5
bR124K	Yes	0	62.4
bR158K	No	-	-
bR376K	No	-	-

Human and mouse fibrinogen sequences were aligned by BLAST and regions where arginine residues in the human sequence were substituted with other amino acids in the mouse sequence were identified. For instance aR123F denotes arginine at position 123 of the alpha chain in human fibrinogen is substituted by phenylalanine in the mouse sequence. Areas that did not have surrounding regions of homology between species were denoted as (-).

Mass spectrometry data from Nakayama-Hamada et al.(25) was used to identify sites of citrullination after *in vitro* treatment of fibrinogen with PADI2 or PADI4. Mean citrulline specific antibody responses in DR4-IE tg or B6 mice immunized with CithFib at day 70. Antibody responses were determined by synovial proteome microarray analysis, and represent the mean arbitrary antibody units for 5 individual mice.