Electronic Supplementary Material (ESI) for Biomaterials Science.

Supplementary Table 1. Source, reference, company, endotoxin levels and purity of the Ch formulations used *in vitro* and *in vivo*.

Source	# Reference	Company	Endotoxin Levels	Purity (%)	Ref
Squid Pen	NR	France-Chitine	NR	*	(1)
NR	NR	Sigma-Aldrich	Negative	NR	(2)
NR	NR	Fluka Chemika	NR	NR	(3)
NR	NR	Sigma-Aldrich	NR	NR	(4)
NR	NR	Sigma-Aldrich	NR	NR	(5)
Shrimp shell	NR	Marinard Biotech	NR	*	(6)
NR	NR	Zhejiang Golden-shell Pharmaceutical	NR	NR	(7)
NR	NR	Xingcheng Biological Industrial Limited	NR	NR	(8)
NR	NR	Yuhuan County Marine Chemical Company	NR	NR	(9)
Squid Pen	NR	France-Chitine	Negative	*	(10)
NR	NR	Sigma-Aldrich	NR	NR	(11)
Shrimp shell	NR	Primex	NR	*	(12)
NR	NR	Sigma-Aldrich	NR	*	(13)
NR	NR	Ao'xing Biotechnology	NR	**	(14)
NR	NR	NR	NR	NR	(15)
Squid Pen	NR	France-Chitine	Negative	*	(16)
NR	NR	NR	Negative	NR	(17)
NR	NR	Yuhuan Ocean Biochemical	NR	*	(18)
NR	NR	Zhejiang Aoxing Biotechnology	NR	NR	(19)
NR	448869, 42344, 448877, 419419	Sigma-Aldrich	NR	NR	(20)
NR	NR	Sigma-Aldrich	NR	NR	(21)
NR	NR	Sigma-Aldrich	NR	NR	(22)
NR	NR	Sigma-Aldrich	NR	*	(23)
NR	NR	Sigma-Aldrich	NR	NR	(24)
NR	NR	Sigma-Aldrich	NR	NR	(25)
NR	NR	Aoxing Bio-Technology	NR	NR	(26)
NR	NR	Zhejiang Aoxing Biotechnology	NR	NR	(27)
NR	9012-76-4	Solarbio Company	NR	NR	(28)
NR	NR	NR	NR	NR	(29)
Crab Shell	9012-76-4	Coolaber	Negative	NR	(30)
NR	NR	Koyo Chemical	Negative	NR	(31)
Shrimp Shell	C-3646	Sigma-Aldrich	NR	NR	(32)
NR	NR	Sigma-Aldrich	NR	NR	(33)
NR	NR	Golden-shell Biochemical	NR	NR	(34)
Shrimp Shell	NR	Primex	NR	*	(35)
Shrimp Shell	NR	Primex	NR	*	(36)
NR	NR	Heppe Medical Chitosan	NR	NR	(37)
NR	NR	Golden-shell Biochemical	NR	*	(38)
NR	NR	NR	NR	NR	(39)
NR	Protasan UP CL110	Pronova Biopolymer	NR	NR	(40)
NR	NR	Immunophotonics	NR	NR	(41)

NR	Protosan UP G213	NovaMatrix	NR	NR	(42)
NR	NR	Immunophotonics	NR	NR	(43)
NR	NR	NR	NR	NR	(44)
NR	NR	NR	NR	NR	(45)
NR	NR	Intermedpharm	Negative	NR	(46)
NR	Protosan G213	NovaMatrix	NR	NR	(47)
NR	NR	Pronova Biopolymer	NR	NR	(48)
NR	Protosan G213	NovaMatrix	NR	NR	(49)
NR	Protosan G213	NovaMatrix	NR	NR	(50)
NR	NR	Koyo Chemical	NR	NR	(51)
NR	Protosan G 213	NovaMatrix	NR	NR	(52)
NR	NR	NR	NR	NR	(53)
NR	NR	NR	NR	NR	(54)
NR	NR	Pronova Biopolymer	NR	NR	(55)

Notes:

NR – Not Reported

* The authors mention performing a purification protocol.

** The authors mention not performing further purification.

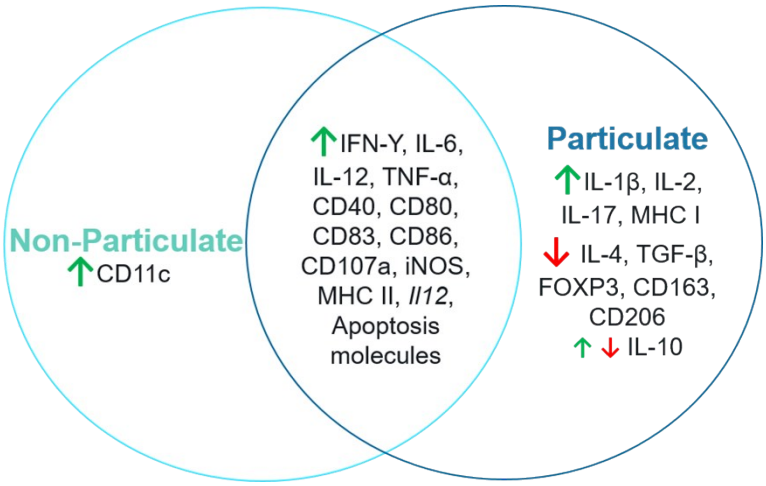
Supplementary Table 2. Ch formulations explored in vitro and in vivo, as well as the controls used to obtain the values presented in the results section.

Type of Formulation	Designation	Control	Ref
Films	Ch/γ-PGA PEMs incorporating IFN-γ	IFN-γ	(1)
	Ch solution for well coating	TCPS	(2)
Gel	Poloxamer 407-Ch hydrogel containing PLGA-OVA NPs, MPLA and QA	PLGA-OVA NPs	(3)
	Ch-DOX hydrogel containing Sig/E7/LAMP-1 vaccine	Sig/E7/LAMP-1 vaccine	(4)
	Ch hydrogel vaccine incorporating OVA and QA	DCs vaccine + OVA	(5)
	Ch thermogel	NT	(6)
Microneedles	Core-shell (Ch shell and PVA/PVP core) microneedle system incorporating anti-PD-L1 drug and 1-MT/Polymer solution	Anti-PD-L1/1-MT injection	(7)
Nanoparticles	IL-12 loaded TPP-Ch NPs	IL-12	(8)
	NKG2D-IL-21 loaded Ch NPs	*In vitro:* IL-21	(9)
		In vivo: PBS	
	Ch/γ-PGA NPs carrying IFN-γ	IFN-γ	(10)
	Ch NPs carrying CD40 and ICOSL mRNA	Lipofectamine as a transfection agent	(11)
	ChLa NPs carrying CD73 siRNA + TCL pulsed DCs	DCs vaccine	(12)
	PEG-ChLa NPs carrying A2AR siRNA	NT	(13)
	Mannose coated-Ch NPs carrying TCL	TCL	(14)
	CMCS-coated SF-CLN and IMD-CLN cationic lipid based NPs	SF-IMD solution	(15)
	Ch/γ-PGA NPs	NT	(16)
	Ch NPs carrying MG	MG or NT	(17)
	GC/GM-CSF-IL-21 NPs	GM-CSF-IL-21	(18)
	Biotinylated Ch NPs with plasmids encoding GM-CSF and IL-21	PlasmidS encoding GM-CSF and IL-21	(19)
	Ch/RA Polyplexes or Ch/RA Polyplexes + OVA	RA or RA + OVA	(20)
	Ch-conjugated green copper oxide NPs or Ch-conjugated green copper oxide NPs carrying BSA	Green Copper Oxide NPs or BSA	(21)
	Ch NPs carrying plasmid pBudCE4.1-azurin-MAM-A	Plasmid pBudCE4.1-azurin-MAM-A	(22)
	Ch coated HCuS NPs-CpG nanocomplexes + laser therapy	Laser therapy	(23)
	Ch NPs carrying OVA and poly I:C	*In vitro:* Soluble OVA	(24)
		In vivo: Soluble OVA + Poly I:C / Soluble OVA	
	Ch-dextran sulfate NPs carrying PD-1 and PD-L1 siRNA	NT	(25)
	FA-Ch NPs carrying IP-10 gene + TRP2-specific $CD8^+$ $CD28^+$ T cells	TRP2 $CD8^+$ $CD28^+$ T cells	(26)
	MCMC/HA/PS/ODN NPs	HA/PS/ODN NPs	(27)
	FA-Ch NPs carrying IP-10 gene + DC/TCL fusion vaccine	DC/TCL Vaccine	(28)
	Ch NPs carrying EphrinA1-PE38/GM-CSF Ch NPs	Saline	(29)
	Ch NPs	NT	(30)
	Chitosan-carboxymethyl dextran NPs carrying cefmetazole, 5-FU or ICG	NT	(31)
	Alginic-acid coated Ch NPs carrying legumain DNA vaccine	PBS	(32)
	Nanogel of amphoteric methacrylamide N-carboxyethyl Ch and N-(2-hydroxy)propyl-3-trimethylammonium Ch Chloride coated with RBCM and	PTX + IL-2	(33)

	carrying IL-2 and PTX		
	PEG and mannose-trimethyl Ch and citraconic anhydride grafted PC carrying VEGF and PIGF siRNA	PBS	(34)
	Ch NPs carrying plasmid encoding E7 vaccine and IL-12	E7-DNA + IL-12	(35)
	Ch NPs carrying plasmid encoding E7 vaccine	E7-DNA	(36)
	ChNPs carrying siRNA for Gal-1	NT	(37)
	TMC NPs conjugated with DOX via CA linkage and carrying IL-2	DOX + IL-2	(38)
Solution	Ch solution	NT	(39)
	GC + HIFU	HIFU	(40)
	N-dihydrogalactochitosan solution	NT	(41)
	Ch glutamate/IL-12/OVA solution	IL-12/OVA solution	(42)
	N-dihydrogalactochitosan + PDT-generated vaccine	PDT-generated vaccine	(43)
	SWNT-GC solution + PTT or SWNT-GC + PTT + anti-CTLA-4	*In vitro:* PTT *In vivo:* anti-CTLA-4	(44)
	N-dihydrogalactochitosan solution + laser treatment	NT	(45)
	WSCO and IFN-Y solution	NT	(46)
	Ch glutamate/IL-12 solution	PBS	(47)
	Glycated Ch solution	NT	(48)
	Ch/IL-12 solution	NT	(49)
	Ch/IL-12 solution	IL-12 solution	(50)
	Ch solution	Acetic Acid	(51)
	Ch/IL-12 solution	IL-12 or PBS	(52)
	COS solution	NT	(53)
	COS solution + CTX	CTX	(54)
	Glycated Ch solution + Laser	Laser	(55)

Abbreviations List:

1-MT: 1-methyl-D,L-tryptophan; 5-FU: 5-Fluorouracil; A2AR: Adenosine A2A Receptor; CA: *cis*-aconitic anhydride; CD: Cluster of Differentiation; Ch: Chitosan; ChLa: Chitosan Lactate; CLN: Cationic Lipid-based Nanoparticles; CMCS: *O*-Carboxymethyl-Chitosan; COS: Chitosan Oligosaccharide; CpG: Cytosine-Guanine; CTX: Cyclophosphamide; DCs: Dendritic Cells; DOX: Doxorubicin; Ephrin1A: Specific ligand for tyrosine kinase family receptor EphA2; FA: Folate; Gal-1: Galectin-1; GC: Glycated Chitosan; GM-CSF: Granulocyte-Monocyte Colony Stimulating Factor; HA: Hyaluronan; HCuS: Hollow Copper Sulfide; HIFU: High-Intensity Focused Ultrasound; ICG: Indocyanine Green; ICOSL: Inducible Costimulator Ligand; IFN-γ: Interferon γ; IL: Interleukin; IMD: IMD-0354; IP-10: Interferon-γ-inducible Protein 10; MCMC: Mannosylated Carboxymethyl Chitosan; MG: Methylglyoxal; MPLA: Monophosphoryl Lipid A; NPs: Nanoparticles; NT: No treatment; ODN: Oligodeoxynucleotides; OVA: Ovalbumin; PBS: Phosphate-buffered saline; PC: poly (allylamine hydrochloride); PDT: Photodynamic Therapy; PE38: *Pseudomonas aeruginosa* toxin PE38; PEG: Polyethylene Glycol; PIGF: Placenta Growth Factor; PLGA: Poly (lactic-co-glycolic acid); Poly I:C: Polyinosinic-Polycytidylic Acid Sodium Salt; PS: Protamine sulfate; PTT: Photothermal Therapy; PTX: Paclitaxel; PVA: Polyvinyl Alcohol; PVP: Poly (*N*-vinylpyrrolidone); QA: Quil A; RA: Toll-like receptor 3-recognizing RNA adjuvant; RBCM: Red Blood Cell Membrane; SF: Sorafenib; Sig/E7/LAMP-1: Vaccinia vector carrying E7 antigen and sorting signal of the lysosome-associated membrane protein 1; siRNA: Small Interfering RNA; SWNT: Single Walled Carbon Nanotube; TCL: Tumor Cell Lysate; TCPS: Tissue Culture Polystyrene; TMC: Trimethyl Chitosan; TRP2: Melanoma-specific antigen TRP2; TPP: Tripolyphosphate; VEGF: Vascular Endothelial Growth Factor; WSCO: Water Soluble Chitosan Oligomers; γ-PGA: Poly-(γ-glutamic acid);



Supplementary Figure 1. Summary of the immune-related alterations observed in *in vitro* studies, using particulate and non-particulate Ch.

	Non-particulate	Common	Particulate
TME	↑ ↓ Macrophages	↑CTL, DCs, Th1, NK cells ↓ Tregs	↑M1-macrophages ↓ M2-macrophages, MDSCs
Systemic	↑ ↓ Macrophages	↑B cells, CTL, DCs, Th, NK cells, IgGs ↓MDSCs, Tregs	↑ ↓ IL-4, IL-10, IL-17, TGF-β

Supplementary Figure 2. Summary of the immune-related alterations observed in *in vivo* studies, using particulate and non-particulate Ch.

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